

Subsidy Project of Decommissioning and Contaminated Water Management Started From FY2021

Research and Development for Treatment and Disposal of Solid Waste

Final Report for FY2021

September 2022

International Research Institute for Nuclear Decommissioning (IRID)

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1. R&D Approach



1. R&D approach -Background and purpose of R&D-

Basic policies of waste management in the "Mid-and-Long-Term Roadmap"

- It is necessary to <u>understand the properties</u> of solid waste including nuclide composition and radioactive concentration, so as to study its treatment and disposal.
- The solid waste generated shall be <u>stored and managed safely and rationally in</u> <u>accordance with its properties</u>.
- To ensure safety in the storage and management of solid waste, <u>a method for rationally</u> <u>selecting treatment (preceding processing) for waste stabilization and</u> <u>solidification shall be developed, and preceding processing methods shall be</u> <u>selected</u> before determining the technical requirements for disposal.
 - To effectively proceed with research and development (R&D) project related to treatment and disposal of solid waste, the R&D project teams of characterization, treatment and disposal of solid waste shall closely work together. <u>The progress and issues of each</u> <u>project shall be shared to proceed with R&D among the project members while</u> <u>obtaining an overview of activities pertaining to solid waste management and</u> <u>identifying the need for R&D tasks</u>.

Until around the end of FY2021

Technical prospects of measures for the treatment and disposal of solid waste, and their safety are indicated.

*Mid- and Long-Term Roadmap Toward the Decommissioning of Fukushima Daiichi Nuclear Power Station Units 1-4, TEPCO(September 26, 2017), excerpts from 4-5, partly revised.



1. R&D approach

Goals for achieving the milestones of the Mid-and-Long-Term Roadmap

The following specific goals for the milestones to be achieved until around the end of FY2021 are indicated in the strategic plan* of the Nuclear Damage Compensation and Decommissioning Facilitation Corporation (hereinafter referred to as NDF). R&D initiatives are undertaken to contribute to the attainment of the goals.

Specific goals related to technical prospects

- Safe and rational disposal concepts shall be formulated based on the properties and amount of solid waste generated at the Fukushima Daiichi Nuclear Power Station (NPS) and the treatment technologies that can be used for such solid waste. In addition, safety assessment methods reflecting the properties of the disposal concept shall be developed based on examples from various foreign countries.
- 2. The analysis and evaluation methods for characterization shall be clearly specified.
- 3. Treatment technologies that are expected to be introduced in actual conditions for stabilization and solidification of waste with disposal in mind for various important waste streams (e.g., secondary waste generated from water treatment) shall be clearly specified.
- 4. A system for the rational selection of treatment (preceding processing) methods for stabilization and solidification shall be established based on No. 3 above before the determination of the technical requirements for disposal.
- 5. With respect to solid waste for which treatment technology that considers disposal is not clearly specified, establishment of treatment and disposal measures using the series of methods developed prior to FY2022 shall be foreseeable.
- 6. The challenges pertaining to storage management of solid waste until waste conditioning and corresponding measures shall be clearly specified.

* 3.2.2.4. in Technical Strategic Plan 2019 for Decommissioning of the Fukushima Daiichi Nuclear Power Station of Tokyo Electric Power Company Holdings, Inc. (Nuclear Damage Compensation and Decommissioning Facilitation Corporation, September 9, 2019



1. R&D approach Goal of R&D

- Solid waste management at the Fukushima NPS shall <u>be stored and operated safely and</u> <u>rationally</u> depending on its properties.
- A rational way for selecting preceding treatment methods shall be developed.
- * Goal excerpted from "2. Contents of Project" of Guidelines for applying to the "Project of Decommissioning and Contaminated Water Management (Research and Development of Processing and Disposal of Solid Waste)" (March 1, 2021), This goal is same as it of the existing subsidized project. This research is a continued project to achieve the same goal.





1. R&D approach Reflection of R&D results on the decommissioning



*Solidification and storage of waste according to the results of treatment and disposal study

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1. R&D approach Association with other research

[Input into this project]

ID	Implementation items and specific details (use of information)	Necessary information	Time period	Source from	Remarks
1	Understanding of storage conditions of high dose waste to study for their management.	Consumables of equipment to be used for fuel debris retrieval, and specifications of water treatment and air-conditioning systems, and collecting containers.	As needed	Fuel debris retrieval project	Exchange information as necessary
2	Understanding of storage conditions of high dose waste to study for waste management.	Specifications of systems to be used for canisters of collecting, containing and storing fuel debris	As needed	Canister project	Exchange information as necessary
3	Study on waste management stream and acquisition of basic knowledge concerning approach to applicability assessment of treatment technology	Information on high-temperature treatment technology	As needed	Project proposers	Cooperate through administration office

[Output from this project]

ID	Implementation items and output details	Purpose of use at output destination	Time period	Destination	Remarks
1	Research results of subsidized projects	Acquisition of basic data on study of high- temperature treatment technology	As needed	Project proposers	Cooperate through administration office
2	Sharing of storage management results and adjusting the connecting conditions	Confirmation of compatibility with the fuel debris retrieval process	As needed	Fuel debris retrieval project	Exchange information as necessary
3	Confirmation of evaluation conditions of the amount of hydrogen generation	Comparison with evaluation of hydrogen amount generated from fuel debris	As needed	Canister project	Exchange information as necessary
4	Knowledge of waste characterization	Results of evaluations on analysis data of samples taken from Fukushima Daiichi and study on generation process of the samples	As needed	Fuel debris characterization project	Participate assessment committee

Note: Fuel debris retrieval project: Development of Technology for Further Increasing the Scale of Retrieval of Fuel Debris and Internal Structures Project Canister project: Development of Technology for Collection, Containing and Storage of Fuel Debris Project Fuel debris characterization project: Development of Technology for Analysis and Estimation for Fuel Debris Characterization Project Project proposers: Organizations that proposed to study applicability of practical high-temperature treatment technology for solid waste to establish selecting methods for advance treatment method.



1. R&D approach Implementation schedule (1/3)

Details of Implementation	Planned							F	Y2021					
	Actual		4	5	6	7	8	9	10	11	12	1	2	3
a. Storage management														
(a) Investigation and evaluation of storage methods														
① Study of storage methods for high dose waste			Drafting of t	he investiga	ation plan $ abla$		Organizatio parameteriz	on of the pro zation	perties of wa	aste and			Summary	∇
Organization of the properties of waste and parameterization														
			Drafting of	the investig	ation plan	Evaluati high dos	ion of the an	nount of hyd	rogen gas g	enerated by	v typical		Summary	∇
•Evaluation of the amount of hydrogen gas generated by typical high	dose waste													
② Study of the requirements for containers and storage facilities			Drafting of	he investig	ation plan ∇	Investigati	ion of proble	em cases 🗸	Study of n	neasures	s and ∳√		Summary	∇
Investigation and organization of problem cases					1									
			Drafting o	f the investi	gation plan	7	methods	for vent filte	mance asse	essment	∇		Summary	∇
Investigation of performance assessment methods for vent filters					-									
(b) Technological development of contamination evaluation for sorting solid	vaste			Drafting of	plans	∇		System de	sign		∇		Summary	<u> </u>
			-											
b. Establishment of concepts of treatment and disposal, and development of safety assessment methods														
(a) Establishment of selecting advance treatment methods			Drafting of	the experim	ental plan								lation	
① Low-temperature treatment technologies			formalities			∇	(outsource	d, in-house)	s and analys	ses)	and report		
Establishment of methods for examining the possibility of solidification treatment	tion by means of	low-											S	ummary 🗸
			Drafting of	the experim	nental plan			(T	
			formalities	ng out outso	ourcing	∇	outsourced	l, in-house)	and analysi	es)	and report		
ii. Investigation of degradation of solidified substances by heating, etc.													Sı	ummary 🖓
												Tost compl	otion	
			Drafting of	the experin	nental plan	∇ In	nplementatio	on of analys	is (in-house)		and report		
III. Evaluation of the relationship between the inventory of CS, etc. and temperature of solidified substances													S	ummary $ abla$
			Drafting of	the experin	nental plan							Teeteema	1	
			formalities			∇	(outsourced	d, in-house)	anu analys	62		and report		
iv. Investigation into and evaluation of factors influencing long-term de	gradation												ε	Summary $\overline{ abla}$
														I



The target completion deadline of each work step is indicated by " ∇ ".

1. R&D approach Implementation schedule (2/3)

	Planned		-			-	F	Y2021		-	-	-	
Details of Implementation	Actual	4	5	6	7	8	9	10	11	12	1	2	3
(2) Investigation of approach for assessing applicability for treatment technology	nology	Drafting of the study plan			Investigation and organization work (in-house) Test completion and rep					d report			
i. Consolidation of acquired data and evaluation method used for tec	hnological			1							1		Summary 7
comparison		Drafting of	the experim	l nental plan									
		and carryir formalities	ng out outso	urcing	∇	Implementa (outsource	ation of tests d, implemen	s and analys ited in-hous	ses e)		Test comp and report	letion $ abla$	
ii. Investigation related to Cs volatilization volume during high-tempe treatment and its control	erature		1								-		ummary 🗸
												1	
(b) Provision of disposal concepts and development of safety assessment me	ethods												
 Information organization to study the disposal concepts and to estab assessment methods 	lish safety	C) Prafting of th	e study plar			Study	of disposal	concepts	∇	7	Summary	∇
Study of disposal concepts for typical waste streams													
		Dr	afting of the	study plan	7 1	ł	Inform	nation orgar	nization	$\overline{\nabla}$	7	Summary	∇
Information organization to establish safety assessment methods	ods												
2 Development of techniques for assessing impact of affecting substance	es, etc. on disposal	Drafting	of the study	∕plan ∖	7	Org	panization o	f sorption in	npact, and s	tudy of para	ameters	7	7
Evaluation of nuclide sorption impact													
		Drafting c	of the study	plan 🗸		Sensitivity	evaluation o	of parameter	rs including	sorption im	pact	$\overline{\nabla}$	
Sensitivity evaluation of the dose evaluation parameters													
												Summary	∇

c Characterization of solid waste													
(a) Efficient characterization													
① Investigation of analytical inventory estimation methods													
i. Implementation of analysis													
(i) Collection, storage and transportation of samples						(collection of	rubbles					
Collection, storage		*Collection status of p	is carried or reparation o	out througho f the sample	ut the year. es.	The date of	transportati	ion will be c	hanged app	ropriately d	epending or	n the	
			Analysis sa	mples trans	portation (Fi	irst time) 🗸	Ar	nalysis samp	oles transpo	ortation (Sec	cond time)	ľ	
Transportation	Transportation												
										Study of	of the plan f	or the follow	ing year ∑
 Study of the plan for the following year 													

The target completion deadline of each work step is indicated by " ∇ ".

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1. R&D approach Implementation schedule (3/3)

Details of Implementation	Planned					-	F	Y2021		-			
		4	5	6	7	8	9	10	11	12	1	2	3
						Imp	lementatio	of analysis	 3				∇
(ii) Implementation of analysis			I	I	1			1	1				
		* Analysis i	s carried ou	t throughou	t the year. N	Moreover, the	e analysis p	lan is chang	ged appropi	riately as ne	eded.		
				∇	As neede	d	7	As needed	1	∇	As needed	l As	s needed V
(iii) Analytical database								-					
		*Analytical	database is	compiled a	nd then rep	orted as nee	ded.						
ii Study on analytical inventory estimation methods													
(i) Status of generation and status of storage of solid waste			Study o and sto	f classificati rage form	ion based o	n the genera	tion け ∖	7					
			ļ			-							
(ii) Contamination mechanism		;	Study relate	d to contar	nination thro	ough air	7	Stud	dy related to	o contamina	tion through	water	
									l				
(iii) Study on analytical inventory estimation methods		Drafting of	of the study	plan	Z Study rel waste in	lated to class ventory	sification of	waste and e	estimation o	of N	Compilatio of estimatio	n and sumn on issues	∇
② Proposal for mid-and-long-term analysis plan													
i Development of mid-and-long-term analysis plan		Consolidation to analytical	on of informat capabilities	ion related	Consolida	ation of samp ssioning sche	le informati dule, etc. i	ion, elated to an	alysis 🗸	term analy	ent of mid- a sis plan	and long-	Summary V
ii Study of analytical goals based on needs		Co tar	nsolidation get nuclides	of analytica , analysis m	l needs, ord nethods, etc	ler of priority, 2.	identificati	on of	7	 Study of o analytical 	uantitative goals		Summary V
								-					
		* ************			~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~								
iii Study of analytical planning methods			Trial run	of the study	of analytic	al planning m	nethods	I	7	Evaluation the trial ru	n of the valic n results	lity of	Summary 🗸
(b) Development of sampling technology		Drafting up	the study p	lan 🗸	D	Design, manu	facturing a	nd testing of	the sampli	ng system			
		-									L		
					Stu	udy and form	ulation of s	ampling plai	ns			∇ Sum	mary 🗸
d. Integration of R&D results					Church	f anna atraite		alidation -f	the view -	into for			
			Drafting up p	lans	narrow	ing down	es and cons	solidation of	une viewpo			Summary	

The target completion deadline of each work step is indicated by " ∇ ".



1. R&D approach Project organization chart



2. Project details (No.1)

- a. Storage management
 - (a) Study and evaluation of storage
 - [1] Investigation of storage methods for high dose waste
 - [2] Investigation of the requirements of storage containers and facilities
 - (b) Technological development of contamination evaluation for segregate solid waste



(a) Investigation and evaluation of storage and management methods

 According to the Mid-and-long-term Roadmap*, solid waste generated in the Fukushima Daiichi Nuclear Power Station (NPS) shall be stored and managed in a safe and effective manner considering the properties.

*Excerpted from 4-5, Mid-and-long-term Roadmap towards the decommissioning of the Fukushima Daiichi NPS, Tokyo Electric Power Company (TEPCO) Holdings, Inc. (Sep. 26, 2017)



Reflecting to the storage and management method for the Fukushima Daiichi



- Achievements up to FY2020
- Referring one example of the storage method for high dose waste, hydrogen generation rate was estimated based on decay heat that Cs-137 is set as a representative of a contaminated nuclide.

Goal

- O Issues of evaluation on hydrogen generation behaviors for high dose waste are clarified.
- Details of implementation (Details are described in the next pages.)
- Information acquisition

Parameters including properties of high dose waste and G values of hydrogen generation are clarified.

- Study of factors required for evaluation of the amount of hydrogen generation
 High-level radiation waste to be required for measures of hydrogen generation during storage is selected and factors that will affect evaluation of hydrogen generation rates are studied.
- O Evaluation of the amount of hydrogen generation
 - Parameters of the hydrogen generation evaluation model that would affect hydrogen generation rates are identified and the effects of the hydrogen generation rates are investigated.
 - Parameters for each factor that would affect the evaluation of hydrogen generation rates are set in more practical manner and the amount of hydrogen generation is evaluated.
 - On a base of evaluation results of the hydrogen generation amount of a representative high dose waste, issues of the evaluation are clarified.
- Indicators for the achievements of goal
- Issues of the hydrogen generation behaviors for high dose waste are clarified (Target at the completion: TRL 2 \rightarrow 3).





<Study on estimation of the hydrogen generation amount in previous years and in FY2020>

Evaluation formula (linear model) and parameters of hydrogen generation rate as well as concepts of setting are described as below.





**Estimated inventory of waste to be generated during retrieving fuel debris.

<Study of the storage methods for high dose waste>

Study items and the procedures for FY2021 are as below.

1. Information acquisition



1. Information acquisition

- According to the waste list that was studied based on an integration of R&D results (waste stream), properties of waste (compositions of waste, radioactivity inventory and water content rate, etc.) required to study the storage management method are clarified from aspect of hydrogen generation (high-radiation dose and water content or not).
- To examine parameters of hydrogen generation, the hydrogen generation mechanism is clarified and then information related to results of existing studies and parameters of G values are examined.

2. Study of factors required for evaluation of hydrogen generation (hydrogen generation rates)

- As a result of clarifying the hydrogen generation parameters, impact factors of the hydrogen generation rate evaluation are studied.
- Wastes needed for countermeasure during storage are selected based on the results of information acquisition for high dose waste and study of hydrogen generation factors.
- The evaluation model for selected waste is studied and parameters (impact parameters) that will affect hydrogen generation parameters especially hydrogen generation rate are extracted.

3. Evaluation of the amount of hydrogen generation (hydrogen generation rate)

- The impact parameters (practical estimation value) that will be used for specific calculation evaluation are set based on the study results and the amount of hydrogen generation for representative high-radiation waste is evaluated by calculation.
- Issues for the evaluation model and parameter setting are studied based on a result of the calculated evaluation.
- The selection of waste to be required for countermeasure are also re-examined whether it is appropriate or not based on the evaluation results.

<Clarification of hydrogen generation parameters>

Information acquisition

Hydrogen generation evaluation formula from radiolysis

(1) Linear model

Hydrogen generation rate= $\Sigma i, j$ (Decay heat of nuclide i / radiation type j × G value of radiation type j × Water absorption rate of radiation type j)

Decay heat of nuclide i / radiation type j= Inventory of nuclide i × Emission rate of radiation type j × Emission energy of radiation type j

Hydrogen generation rate is proportional to <u>1) absorption energy (decay heat)</u>, <u>2) G value, and 3) absorption rate</u>.

Decay heat depends on nuclide inventory, G value depends on radiation types and absorption rate depends on water existence (thickness of water membrane, etc.) .Hydrogen continues generating at a certain rate.

(2)
$$\frac{dC_i}{dt} = \xi g_i^{\gamma} Q^{\gamma} + \sum_l^N \sum_m^N k_{lm} C_l C_m - C_i \sum_s^N k_s C_s$$

 C_i , C_n , C_s : Concentration of material i, I, m and s (mol/I), g_i^{γ} : G value of γ ray of material i (unit/100eV),

 ξ :Number of molecules and conversion factor of mol (mol/unit), Q^{γ} : Absorption energy of γ ray (eV), N:Number of materials affecting reactions (-)

 k_{lm} :Reaction rate constant of material I and m (l/mol/sec), k_s : Reaction rate constant of material i and s (l/mol/sec)

Hydrogen generation rate depends on 1) Absorption energy (decay heat and proportion of energy absorption), 2) G value and 3) Reaction rate constant among materials and thereby it is function of time.

[1] Yoichi Wada, Masahiko Tachibana, Atsushi Watanabe, Kazushige



Fig.1 : Aging changes of degradation product concentration calculated by radiolysis model [2] *(L) and (g) in the figure represent liquid phase and gas phase.

Ishida, Nobuyuki Ota, Naoto Shigenaka, Hiromitsu Inagaki & Hiroshi Noda (2016) Effects of seawater components on radiolysis of water at elevated temperature and subsequent integrity of fuel materials, Journal of Nuclear Science and Technology, 53:6, 809-820 [2] Junichi Takagi and Seiji Yamamoto " Alpha-radiolysis Simulation to Predict Hydrogen Accumulation under Fuel Debris Condition ",The International Workshop on Radiolysis and Electrochemistry NPC 2016, Brighton, United Kingdom - October 7, 2016



Information acquisition

<Clarification of hydrogen generation parameters>

[Clarification of mechanism] Applicability of the linear and radiolysis models

Hydrogen is produced by irradiating into water. Hydrogen produced from irradiation mitigates into the gas phase of a container and the partial pressure of hydrogen is increased.

The concentration of hydrogen underwater achieves equilibrium as time passes. The concentration can be calculated by the radiolysis model.

The concentration of the gas phase hydrogen in the container is equal to the liquid phase hydrogen concentration in accordance with the Henry's Law.

When the concentration is less than 4 vol %, the countermeasure for hydrogen generation is not necessary.

On the other hand, when the hydrogen concentration of the gas phase is more than 4 vol %, hydrogen in the container has to be released into outside of the container by using a filter vent.

The hydrogen generation rate shall be conservatively calculated by the linear model.

In the both cases,

It is important to set values of decay heat, G value and absorption rate for the estimation.



Fig.1 : Illustration of hydrogen mitigation inside/outside of the container



<Clarification of hydrogen generation parameters>

Clarification of the G value parameter information^[1]

The distribution of chemical species are nearly homogeneous in space for a microsecond after physical processes that water absorbs radioactive energy (excitation and ionization), physical chemistry processes (spar formation) and chemical processes (chemical reaction in a spar).

- Chemical yield of chemical species represents as G value and [number of species //100 eV] or [mol/J] are used as units.
- After completing a spar reaction (up to 10⁻⁷ second), G value is called as the primary G value.
- On the other hand, the amount of these degradation products that interact after long time has passed is called as "apparent G value" or " effective G value." Generally, the primary G value is larger than apparent G so that the primary G value is used for evaluation.



[1] Yusa Muroya, "Radiation chemistry of water and water solution", RADIOISOTOPES vol. 66 (2017) No. 10 <u>https://www.jstage.jst.go.jp/article/radioisotopes/66/10/66_661006/_article/-char/ja</u>



<Clarification of hydrogen generation parameters>

[Clarification of the G value parameter information]

- The primary G value depends on radiation types and energy, etc. (Table 1 ^[1]).
- Handling of free water and other type of water (i.e. Cement crystal water) is accordance with rules in each country (Table 2 ^{[2}]).

[Examples]

- U.S. and U.K.: Hydrogen generation is evaluated considering the reduction of free water caused by cement solidification and changes of the water condition
- France: G value for free water is used for estimation even after cement solidification

* G values for estimating hydrogen generation are set after classification depending on properties of waste (water status, and types of organic substances and radioactivity) in each country

Types of radiation (LET [eV/nm])	-H ₂ O	e ⁻ aq	ОН	н	H ₂	H_2O_2	HO ₂ / O ₂ -
Gamma rays (0.23)	4.08	2.63	2.72	0.55	0.45	0.68	0.008
18 MeV D* (12.3)	3.46	1.48	1.78	0.62	0.68	0.84	-
1.8 MeV H* (34.5)	2.85	0.92	1.08	0.38	0.79	0.85	0.03
32 MeV He ⁺ (61)	3.01	0.72	0.91	0.42	0.96	1.00	0.05
12 MeV He ²⁺ (108)	2.84	0.42	0.54	0.27	1.11	1.08	0.07
Fast neutron (14.5 - 63.4)	2.91	0.99	1.15	0.40	0.78	0.85	0.03

Table 1. Primary G value of major radiation degradation products (molec./100eV)

ć	The G-value wa and the state of	is confirmed to be set acco water.	ording to the radioactive wa	ste classification
		US	UK	France
Gwalue	Carbonate slurry	At the time of drying (5 to 20wt%): 1.6 When cement solidifies: 0.6 to 0.85 ⁻¹	Established based on discussions with the regulatory authorities (Example of initial settings is 0.4 or 0.45 for free water)	Free water, cement hydration water: 0.452 (Documented value) Mg(OH) ₂ : 0.051 (Documented value)
G-value (H ₂)	Iron coprecipitation slurry	Same as above	Same as above	Free water, cement hydration water: 0.452 (Reference value) Fe(OH) and water: 0.00529 (Reference value)
	Reactor internal structures	 Adhesion water: 1.6 (After drying: 0) 	Same as above	Adhesion water: 0.45
Concept of G-value (H ₂)		• Organized as per the waste properties (presence of organic matter, occurrence of solidification, moisture content etc.). * ² • Even if the waste does not contain α nuclides, the α ray G- value is conservatively assumed to be the base.	G-value has been set in the past as 0.05 to 0.5 considering the change of state of water due to cement solidification. Whether to adopt the documented value or the actually measured value is discussed with the regulatory authorities.	G-value of free water is used even after cement solidification. A lower G-value can be set when actual measurement is carried out. (0.032 for the DSC ⁻³ dry sludge) The G-value of waste is calculated from the sum of products of the mass fraction of the generation source and each G-value.
	*1 Set according to t	the solidification conditions (depen	ds on the type of cement material). Ir	n the case of Type I (OPC). G-value of

Table 2. Clarification results of the concepts of G value setting in each country

0.6 is used when the ratio of water to cement by weight is 0.45.

*2 CH-TRU payload appendices (books)

*3 Dry Storage Container

[1]] Yusa Muroya, "Radiation chemistry of water and water solution", RADIOISOTOPES vol. 66 (2017) No. 10 <u>https://www.jstage.jst.go.jp/article/radioisotopes/66/10/66_661006/_article/-char/ja</u>

[2] Achievement reports for FY2018: Subsidy project of Decommissioning and Contaminated Water Management started since FY2016 "R&D for Treatment and Disposal of Solid Waste" and Subsidy Project of Decommissioning and Contaminated Water Management started since FY2017 "R&D for Treatment and Disposal of Solid Waste (R&D for advanced treatment and analysis methods) (study on waste characterization, storage, management, disposal and waste stream) https://irid.or.jp/_pdf/20180000_15.pdf?v=2



<Procedures of information acquisition of high dose waste>



(1) List of waste items

The information is organized based on the list of waste items. However, information on spent fuel and fuel debris are not acquired.

(2) Waste required for information

Waste to be required for information acquisition are selected from the aspect of the following major factor for hydrogen generation.

Water content or not

(3) Information acquisition of high dose waste

The following information of selected waste is clarified

- > Properties: Lump-like and particle-like, etc.
- Radioactivity: Concentration of radioactivity for each nuclide
- > Others: Containers to be used for storage, etc.



<Targeted information: 1. Dismantled waste, rubbles and etc.>

Was	te list: The amount of hydrogen gas	generation is evaluated for Units 1 to 3 assum	ing contamination that is diffe	rent from ordinary reactors.					
10	Dismantling waste								
	S1 Pressure vessel	S2 Containment vessel metal	S3 Containment vessel concrete	S4 Metal from inside the building	S5 Concrete from inside the building	S14 Waste from debris collecti	N1 Dismantling waste from Unit 4	N2 Dismantling waste from Units 5 and 6	N3 Dismantling waste from other facilities
1	Pressure vessel Equivalent to L2 waste	Dismantled metal	Dismantled concrete	Metal scraps	Dismantled concrete	DSP slot plug concrete	Equivalent to ordinary reactor (Refer to S1 to S5)	Equivalent to ordinary reactor (Refer to S1 to S5)	Installed before the earthquake disaster
2	Steam dryer ordinary reactor	Pipes	Contaminated dust (surface contamination)	Pipes	Contaminated dust (surface contamination)	Heat insulating material	Contaminants from the accident (Refer to S3 to S5)		Central radioactive waste treatment building
з	Steam-water separator (includes L3)	Equipment (including cables, panels, etc.)	RSW concrete (activated)	Rebars	T/B walls (inner), floors, pillars etc.	PCV head			High-temperature incineration facility
4	Upper grid plate	Rebars	BSW concrete (activated)	R/B equipment	T/B walls (outer)	Shield plug concrete	Equivalent to		Incineration work-shop building
5	Core shroud	Main body of PCV	RPV pedestal concrete (surface contamination)	R/B high-level radioactive equipment and pipes (DHC system, AC system, FCS)	Rw/B walls (inner), floors, pillars, etc.	RPV head	L2-L3 waste in an ordinary reactor		On-site bunker building
6	Core support plate	PCV internal components	Torus concrete (surface contamination)	R/B high-level radioactive equipment and pipes (DHC system, AC system, RCW)	Rw/B walls (outer)	RPV lower hemispherical dome*			Auxiliary building
7	Other equipment Equivalen	RSW steel plate	Well plug (surface contamination)	P/B high-level radioactive equipment and pipes (AC system)		Equipment for debris retrieval			Common pool building
8	Fuel support waste in	BSW rebars		Additional shielding material					SPT building
9	Jet pump an ordinary	RPV pedestal rebars		Turbine body					Miscellaneous solids incineration facility
10	Control Rod Guide Tube (includes			Generator					Installed after the earthquake disaster
11	CRD housing			Condenser			1		Additional miscellaneous solid
						(Cesium adsorption apparatus
12	CRD			Moisture separator			ma airelad in		(KURION)
13				Feed water heater		vvasie ile	ins circled in		Oil separator
14	Not studied as it in	volves fuel debris handling		Other equipment and pipes,		red or			Second cesium adsorption
				High-level radioactive pipes		red an	e studied.		Third cesium adsorption apparatus
15				(MS system)					(SARRY-2)
16				Rw/B equipment and pipes, etc.					Decontamination equipment (AREVA)
17									Reverse osmosis membrane
	(treatment unit 1
18	Inform	ation on the PCV. tu	urbine and the	e reactor buil	dina concrete	e. etc.			apparatus
19									RO unit for demineralization
20	is not	studied because the	ev are estima	ated to be disi	mantled in th	e air.			Demineralizer
21									Cylindrical flanged tank
(2) F	Rubbles, etc.								Rectangular tank
	S6 Metal rubble	S7 Concrete rubble	S8 Combustible materials	S15 Contaminated soil	N5 Other rubbles, etc.				Solid radioactive waste storage facility
1	Metal scrap	Concrete scrap	Used protective clothing	Contaminated soil	All types of rubbles not mentioned on the left				Slurry stabilization equipment
2	Pipes (metal)	Concrete	Paper and waste cloths	Gravel and quarry	Non-combustibles (PVC pipes etc.)	·,			Volume reduction facility
3	Rebars	Contaminated dust (surface contamination)	Plastics, vinyl, etc.	Dredged soil	Non-combustibles (glass, ceramics, etc.)				Unit 3 fuel discharge cover
4	Dismantled tanks		Wood	Asphalt	Equipment (including cables, batteries, etc.)				Unit 3 fuel discharge equipment
5	Wires		Felled trees and grass		Fixtures and fittings				Facilities related to debris retrieval
6	Empty containers		Incinerated ash		Building sludge (generated				Debris temporary storage facility
7	Equipment and materials required for construction work (steel plates, steel pipes, etc.)				when water is drained)				Storage tanks for contaminated water
8	Vehicles and heavy machinery								Equipment for removing sludge
6									Cesium adsorption vessel
9									temporary storage facility
10			1 1 4		1 1.4			Units 4 to 6 not studied	
12	🖿 🖌 🛛 Informatio	on on the waste that i	ncludes the re	eactor pressure	e vessel and ti	ne			
13		plug concrete concre	ting during du	, al dobrio rotria	wal in alarifian			Not studied as it involves fuel d	ebris handling
14		plug concrete genera	anng duning du		eval is clarified	l		No underwater storage and und	lerwater cutting
16	🗆 🗸 🛛 Rubbles	are not studied becau	isa tha radiatio	n laval is low				shoo wator storage and unit	
17		are not studied becat						Same as waste from ordinary r	eactors
18		1	1	Į	ļ				



<Targeted information: 2. Secondary waste generated from water treatment and other waste>

3	Secondary waste generated from co	ntaminated water treatment							
\square	S9.1 Adsorption vessel ① (KURION, SARRY)	S9.2 Adsorption vessel ② (Mobile Sr removal equipment)	S10.1 Multi-nuclide removal system ① (slurry)	S10.2 Multi-nuclide removal system ② (adsorbent)	S10.3 Multi-nuclide removal system ③ (treatment column)	S11 Sludge from decontamination equipment	S12 Filter	S13 Slurry discharged from the evaporative concentration apparatus	N4 Other water treatment facilities
1	KURION	Mobile strontium removal equipment (KMPS)	Carbonate slurry	Silico-titanate series adsorbent (FST, Sr-treat)	Chelate resin series adsorbent: ferric hydroxide (Ly-RT)	Sludge from the decontamination system	High performance ALPS	Slurry discharged from the evaporative concentration apparatus	High performance ALPS
2	Surfactant modified zeolite (SMZ)	Silico-titanate (ISM)	Iron coprecipitation slurry	Activated charcoal series adsorbent (GAC, AGC, Kuraray coal)			Barrier filter (Made of polypropylene)		Silico-titanate (HS -726, FST)
3	Hershelite (H, EH)	Second mobile strontium removal equipment		Polymer series adsorbent: Cerium hydroxide (ReadE2, Read-B)			Cs filter (made of nylon resin)		Activated charcoal series adsorbent (K-MAC)
4	Silver impregnated hershelite (AGH)	Silico-titanate (IE-911, FST) (For cleaning the drainage canal)		Polymer series adsorbent: Iminodiacetic acid chelate resin (IRC748i)			Sr filter (made of nylon resin)		Polymer series adsorbent: (CRB05, CN4000, READ-B, WA30)
5	Cobalt ferrocyanide hershelite (KH)	Silico-titanate (FST) (Cleaning of tanks)		Polymer series adsorbent: Ferric hydroxide series adsorbent (CN4000)			RO concentrated water treatment facility		Zeolite series adsorbent (AgZ, KW-500G-7)
6	Silico-titanate (TSG)			Chelate resin series adsorbent: ferric hydroxide (Ly-RT)			Barrier filter (Made of polypropylene)		RO concentrated water treatment facility
7	Filter sand (silica sand)			Zeolite series adsorbent: Silver zeolite (AgZ)			Mobile Sr removal equipment		Silico-titanate (HS -72)
8	SARRY			Titanium oxide (GX194)			SS filter (Made of polypropylene)		Sub-drain cleaning equipment
9	Chabazite (IE-96)			Cobalt ferrocyanide (Cs-treat)			Ultra filter (made of polvethersulfone)		Zeolite series (A51-JHP)
10	Silico-titanate (IE -911)						Sub-drain cleaning equipment		Silico-titanate (HS -726)
11	Silico-titanate (EN -101, HS -726, ET-501)						Barrier filter (Made of polypropylene)		Activated charcoal series (K- MAC)
12	Filtration sand + zeolite series:						Cs filter (made of nylon resin)		Polymer series (READ-B)
13	Filtration sand + activated charcoal						Sr filter (made of nylon resin)		Mobile treatment apparatus
14	SARRY-2								Silico-titanate (IE -901)(Cleaning
15	Filtration sand + activated charcoal								Silico-titanate (IE -901)
16	series: Anthracite								(Cleaning of trenches) Zeolite series (ET201) Cleaning
17									of trenches) High-performance ALPS
11/									verification test apparatus
19									Polymer series (CRB05, WA30)
20									Zeolite series (AgZ)
21									adsorbent (As-CATCH2)
22									Other
							\		Zeolite sandbag (Zeolite) Zeolite sandbag (activated
									carbon sandbag)
-									
4	Other waste								
	S16.1 Waste from before the earthquake disaster (Solid waste storage facility, etc.)	S16.2 Waste from before the earthquake disaster (on-site bunker, fuel pool, etc.)	X1 Spent fuel	X2 Debris	X3 Substances affecting the environment (segregated)				
1	Waste from operation	Granulated solidified substances (Concentrated liquid waste pellets manufactured by Hitachi)	Spent fuel	Debris	РСВ	V	Vaste items ci	ircled in	
2	Waste from operation (non- conformance to standards or properties unknown)	Granulated solidified substances (Concentrated liquid waste pellets manufactured by Toshiba)	Nuclear fuel (in-service)	Debris (components mixed)	Removed metal (lead, aluminum, etc.)		read are stu	died.	
з		Granulated solidified substances (Waste resin pellets manufactured by Toshiba)	Nuclear fuel (before being used	Debris (components adhered)	Substances containing asbestos				
4	-	Waste resin stored in tanks Waste sludge stored in tanks	Damaged fuel		Substances containing lead				
6		Liquid waste stored in tanks						Spent fuel, debris (fuel debris) is not studied	
7		Control rods		1					
8		Channel box						No underwater storage and underwater cutting	
9		Fuel support							
10		Neutron detector						Same as waste from ordinary reactors	
11		Spent fuel pool		ł	l	Ī			
12		Channel box							

Information on secondary waste generated from water treatment is selected and clarified.

Wastes generated before the Great East Japan Earthquake are classified as Level 1 waste that are equivalent to waste generated from the normal reactor.



<Information items for high dose waste1>

The information of selected items is clarified as below.

➢ Properties

Information to be used for studying evaluation model.

- ✓ Material (metal or concrete or resin, etc.)
- Shape (block-like or porous or particles less than several m diameter)
- Storage conditions

Information on the storage container (anticipated container) and water content required for evaluation

- ✓ Volume of the storage container (estimated) [m³]
- ✓ Volume of stored waste (estimated) [t]
- Amount of stored radioactivity per nuclides [Bq] (calculated from concentration of radioactivity defined from the advanced inventory estimation and amount of stored radioactivity)
- Amount of sored water content (estimated) [L] (setting estimated from water content rate, possibility of dry treatment, pre-treatment and etc.)



Vent



(for secondary waste generated from water treatment) ^[2]

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Fig.1: Storage container for waste (illustration))

IRID

treatment

 ^{[1]:} Final report of Subsidy Project of Decommissioning and Contaminated Water Management in FY 2016 Supplementary Budget, Advanced Fundamental Technology for Retrieval of Fuel Debris and Reactor Internals
 [2]: Estimation setting of the storage container based on the information related to secondary waste generated from water

<Information items for high dose waste2> Estimation of storage method for dismantled waste



Fig.1: Conceptual image of storage case for the container used for estimating the amount of hydrogen generation in FY2020



the size of the storage container.

Fig. 2: Handling flow of large waste studied in FY2020

[1]

Kunio Shiraishi, Takenori Sukekawa and Satoshi Yanagihara, (Japan Atomic Energy Research Institute)", Dismantled Waste Data Analysis of the Japan Power Demonstration Reactor (JPDR) (Commissioned Research)," JAERI-Data/Code 99-050.

> Storage condition of the storage container

A container model was created considering the shape of waste to be generated during fuel debris retrieval in FY2020 and the amount of hydrogen generation was evaluated. In FY2021, the following estimated items are studied to store in the container.

✓ Metal waste including the reactor pressure vessel, etc. A detailed cutting method is not studied as demolition methods are different depending on the installation situation. The waste is cut to fit the size of the storage container under water by remote-operation and block-like wastes are assumed to be stored in the storage container

 Concrete waste to be generated during collecting fuel debris The concrete waste is cut to fit and store the container by using a wired saw. Water is assumed to exist in the container as below.
 Adjacent to contaminated water and water impregnates in the concrete void. (The concrete voids is assumed to be less than 10%. In this case, the void is conservatively set as 10%.)

• Water adhered to the surface when lifting from water after cutting under water. (The conservative estimation of water thickness is 2mm of the whole surface although water is not evenly adhered the whole surface.)

• Water using when producing concrete is not considered (Assuming that water-cement ratio is around 50. Free water may possibly remain right after concrete production, however remaining water can be ignored as free water is assumed to evaporate after time has passed.)

> Storage volume, voids and water content of waste

The following conditions are set for the waste.

i. Storage ratio is set based on track records of JPDR demolitions ^[1]

Metal waste: 20vol% (conservative setting

rather than the record)

Concrete waste: 50vol%

ii. Storage volume of waste is set based on volume inside the container and its proportion.



25

iii. Water volume generated from the waste is

*: The internal volume is set assuming that thickness of the shield is 60mm. (Inner dimensions: $1.3m \times 1.3m \times 0.88m = 1.5m^3$)



Assuming a storage method of secondary waste generated from the water treatment



> Storage condition of the storage container

Secondary waste generated from water treatment is assumed to be stored based on results studied by Tokyo Electric Power Company (TEPCO) Holdings, Inc. as below.

✓ Slurry (carbonate and iron co-precipitation):

After the filter press dehydration treatment, slurry is assumed to be stored in the storage container described in the left figure.

✓ Waste adsorbent:

It is assumed that the waste is removed from HIC and stored in the storage container described in the left figure. Adsorbent is dehydrated through dehydration equipment during storage in HIC therefore, additional dehydration is not performed during replacement.

Storage volume, voids and water content of waste The following conditions are set for the waste. Volume inside the container

*: Assuming that thickness of the shield is 50mm, inner volume is set. (Inner diameters: $1.9m \times 1.9m \times 1.4m=5.1m^3$)

i. Storage ratio:

Slurry: 80vol.% (based on the result studied by TEPCO HD.)

Waste adsorbent: 80vol.% (regarded as same as slurry because of insufficient information)

ii. Voids

Closing of lid

Filter

unit

Pump for extraction of water from compression

> Filtrate tank

Transfe Pump for Setting by inner volume of the container and storage volume of waste

iii. Water content

Slurry: 60vol.% (based on the result studied by TEPCO HD.) Waste adsorbent: 50wt. %

(based on properties of long-term storage spent resin of JPDR^[3]) [1]: Assuming of the storage container based on the information related to secondary waste generated by water treatment.

[2]: The figure was created by reference to information provided by TEPCO HD.

[3] Kato, etc. "Disposal of spent ion replaced resin during long-term storage in JPDR and their investigation of the physical property," Journal of Atomic Energy Society of Japan, vol.22, No.6, pp.386 -391(1980)

Fig. 2: Treatment process of secondary waste for water treatment (slurry) [2]

Liquid wast

Slurry pump



ALPS transfe

Linit cleaning

Liquid waste transfer pump

Liquid waste transfer pump



<Information items for high dose waste4-1> Nuclides required to evaluate hydrogen generation

- Conventional evaluation
 - ✓ When evaluating hydrogen generation from high dose waste caused by accidental contamination, <u>Cs-137</u> is regarded as a target nuclide (evaluation for FY2020). ⇒ <u>Other nuclides except Cs-137</u> are not regarded not to contribute contamination.
 - ✓ According to analysis results of heat value of fuel debris (fuel component and fuel cladding tubes), heating values (energy) of β-, γ- and α-rays can be evaluated (evaluation for FY2020).⇒ The heat value of fuel debris cannot be used to evaluate high dose waste because there are differences in heat values (energy) of contamination between accidental contamination adding to dismantled waste and secondary waste generated from water treatment.

Concepts of the evaluation

✓ Hydrogen generation of high dose waste is evaluated considering nuclides except Cs-137 contributing to heat generation. Current data is used to evaluate wide variety of nuclides.

- ✓ Nuclides for evaluation of hydrogen generation are selected based on evaluation results of heat value of fuel debris.
- ✓ The concentration of selected nuclides is evaluated from radioactivity concentration data for each waste.

 \Rightarrow Nuclides that are used for evaluation are selected from both of the aspects of heat generation (absorption energy) and radioactivity concentration of waste. The heat generation rate for β-, γ- and α-rays is set.

- a. According to evaluation results of fuel heat value evaluated by the Japan Atomic Energy Agency (JAEA) ^[1], nuclides that contributes to hydrogen generation (absorption energy is large) are selected.
- b. According to 10-year-attenuation radiation concentration data that is back data of inventory estimation results^[2] for each waste (waste generated from demolition, fuel collection and water treatment) evaluated by JAEA and the Central Research Institute of Electric Power Industry (CRIEPI), high-level radioactive nuclides are selected.
- c. Nuclides that are attributed to evaluation on hydrogen generation from the aspect of (a) the heat generation rate and (b) concentration of radioactivity are covered and then the heat generation rate of β -, γ and α -rays are set based on multiplication of concentration and decay energy.

[Supplement]

With regard to target dismantled waste (metal and concrete), when evaluating hydrogen generation caused by nuclides that are not accidental and activated-/-secondary contamination nuclides, Co-60 must be added in evaluation target nuclides in accordance with "Requirements for the marginal depth disposal waste and its inspection method: 2015" (AESJ-SC-F014:2015).

JAEA-Data/Code 2012-018, Evaluation on fuel composition for Fukushima Daiichi Nuclear Power Station, https://jopss.jaea.go.jp/search/servlet/search?5036485
 Daisuke Sugiyama, Ryo Nakabayashi, Shingo Tanaka, Yoshikazu Koma & Youko Takahatake (2021) Stochastic estimation of radionuclide composition in wastes generated at Fukushima Daiichi nuclear power station using Bayesian inference, Journal of Nuclear Science and Technology, 58:4, 493-506,



<Information items for high dose waste4-2> Nuclides required to evaluate hydrogen generation

From the aspect of the heat generation rate

Aging variation of the heat generation rate of the Unit 3 reactor core fuel at the Fukushima Daiichi Nuclear Power Station (NPS) evaluated by JAEA^[1] describes in the figure classified by a) Radiated material, b) Actinides and c) Fission products.

The contribution to the whole heat generation rate (refer to the right figure and table) is shown Fission products > Actinides > Radiated material in an early stage. Compared with Actinides that has longer half-life time, the heat generation rate of the fission product decreases and it becomes Actinides > Fission product > Radiated material 50-100 years later.

Among the nuclides that have passed 10-50 years, major nuclides (top five nuclides) that generates a large amount of heat were evaluated from the data of fuel compositions as below.

- Radiated materials: Sb-125, Te-125m, Co-60, Ni-63, Fe-55
- •Actinides: Pu-238,Am-241,Cm-244,Pu-240,Pu-239
- ·Fission products: Y-90,Ba-137m,Cs-137,Sr-90,Cs-134



Fig.1: Aging variation of the heat generation rate of the Unit 3 reactor core fuel at Fukushima Daiichi NPS

	i iout goi	ionution rute	(11/0010)
Time period	10-year	20-year	50-year
Radiated material	1 10F+01	1 01E+00	4 16F-02
Actinides	1.31E+04	1.42E+04	1.47E+04
Fission product	5.37E+04	3.95E+04	1.91E+04
Actinides Fission product	1.31E+04 5.37E+04	1.42E+04 3.95E+04	1.47E+04 1.91E+04

Heat generation rate (W/core)

Heat generation rate (W/g)

Time period	10-year	20-year	50-year
Radiated			
material	8.78E-08	8.01E-09	3.31E-10
Actinides	1.04E-04	1.13E-04	1.17E-04
Fission product	4.27E-04	3.14E-04	1.52E-04

[1] JAEA-Data/Code 2012-018 "Evaluation on fuel composition for Fukushima Daiichi NPS" https://jopss.jaea.go.jp/search/servlet/search?5036485





<Information items for high dose waste 4-3> Nuclides required to evaluate hydrogen generation

Nuclides of dismantled waste

In reference to back data of the inventory estimation results estimated by JAEA/ CRIEPI^[1], the concentration of radioactive nuclides which half-life period have passed 10 years is accumulated and target nuclides for estimation are the ones that the accumulated concentration is up to 99%.

	Table 1 Nuc	lides from the (equivale)	reactor internal nt to L2)	structures	Table 2 Nuclides from
	<assumed< td=""><td>to be the sam studi</td><td>e as the pressu ied></td><td>re vessel</td><td><assumed td="" to<=""></assumed></td></assumed<>	to be the sam studi	e as the pressu ied>	re vessel	<assumed td="" to<=""></assumed>
	No.	Nuclide	Type of radiation	Cumulative concentration	No.
	1	Cs-137	β、γ	39.21%	1
	2	Pu-241	α	65.00%	2
	3	Sr-90	β	88.76%	3
	4	Pm-147	β	94.09%	4
ſ	5	Cs-134	β、γ	95.73%	5
	6	Pu-238	α	96.57%	6
	7	Eu-154	β、γ	97.40%	7
	8	Sb-125	β、γ	97.91%	8
	9	Cm-244	α	98.30%	9
	10	Co-60	β、γ	98.56%	10
	11	Eu-155	β、γ	98.79%	11
	12	Pu-240	α	98.98%	Ļļ
ſ	13	Fe-55	β、γ	99.17%	

to L2) <assumed as="" be="" concrete="" same="" studied="" the="" to=""></assumed>										
No.	Nuclide	Type of radiation	Cumulative concentration							
1	Cs-137	β、γ	52.37%							
2	Pu-241	α	72.73%							
3	Sr-90	β	89.67%							
4	Pm-147	β	94.14%							
5	Cs-134	β、γ	96.30%							
6	Pu-238	α	97.00%							
7	Sb-125	β、γ	97.68%							
8	Eu-154	β、γ	98.30%							
9	Cm-244	α	98.62%							
10	Co-60	β、γ	98.84%							
11	Eu-155	β、γ	99.01%							

the containment vessel concrete (equivalent

 Nuclides of secondary waste generated from water treatment

Major nuclides that are collected from water treatment of each slurry and sludge are clarified.

Table 3 Nuclides selected from the secondary waste generated from water treatment

Nuclide	KURION	SARRY	AREVA sludge	Carbonate slurry	Iron co- precipitation slurry	Silver zeolite
	Cs-137	Cs-137	Sr-90	Sr-90	Sr-90	H-3
β、γ	Sr-90	Sr-90	Cs-137			Sr-90
	Cs-134	Cs-134				
	Pu-241	Pu-241	Pu-241	Pu-241	Pu-241	Pu-241
a	Pu-238	Pu-238	Pu-238	Pu-238	Pu-238	Pu-238
u	Cm-244	Cm-244	Cm-244	Cm-244	Cm-244	Cm-244
	Pu-240	Pu-240	Pu-240	Pu-240	Pu-240	Pu-240

Nuclide	Silico-titanate series adsorbent	Cobalt ferrocyanide	Iminodiacetic acid chelate resin	Titanium oxide	Ferric hydroxide
	H-3	H-3	H-3	H-3	H-3
β、γ	Sr-90				
	Pu-241	Pu-241	Pu-241	Pu-241	Pu-241
a	Pu-238	Pu-238	Pu-238	Pu-238	Pu-238
u	Cm-244	Cm-244	Cm-244	Cm-244	Cm-244
	Pu-240	Pu-240	Pu-240	Pu-240	Pu-240

Nuclides required to evaluate hydrogen generation (plan)

Black letter: Nuclides selected based on the heat generation evaluation of fuel compositions

Blue letter: Nuclides added the above

Radiated material: Sb-125,Te-125m,Co-60,Ni-63,Fe-55,H-3,Eu-154,Eu-155 Actinides: Pu-238,Am-241,Cm-244,Pu-240,Pu-239,Pu-241 Fission products: Y-90,Ba-137m,Cs-137,Sr-90,Cs-134, Pm-147 [1] Daisuke Sugiyama, Ryo Nakabayashi, Shingo Tanaka,

Yoshikazu Koma & Youko Takahatake (2021) Stochastic estimation of radionuclide composition in wastes generated at Fukushima Daiichi nuclear power station using Bayesian inference, Journal of Nuclear Science and Technology, 58:4, 493-506,



<Information acquisition of high dose waste (investigation method)>





<Information acquisition of high dose waste(investigation status)>

廃棄物区分		1	廃棄物性状		総放射能量*(<>	保管状况				保管容器内状況(想定)								
大区分	中区分	小区分	周棄物名称	材質	形状	ペントリタ算用度化)	現在	保管前机理 (計画)	保管前処理 (想定)	保管容器什様 (想定)	容器内容積	廃棄物収納率	廃棄物収納量	廃棄物収納量	収納放射能量	空隙量	水分量	(想定根据等)
7.4=77						ment(pd/t)		DIED DO CELE	in a since a conci		(想定)[m3]	[vol%]	[m3]	[t]	[Bq]	[m3]	[L]	
解体廃棄物	S1 圧力容器		圧力容器	金属	塊状	0.105.10	据付中	-	切断	外寸1.42m×1.42m×1mh	1.4872	20	0.3	2.34	000000	1.2	20	圧力容器廃棄物(事故前L2相当+滞留水汚染)の10年後減費放射能濃度を想定
			A C C R R	金属	地法	9.10E+13	据付中	-	切町	廃棄物保官容器	(約1.5)	20	0.3	2.34	別述登理	1.2	20	比重7.8とし、L1廃棄体2.9t収納時の表面付着水等:約4~20kgから20kg(学会標準参照)
			风 不分離發	立涡	塔沃		35194	-	9781	((// ٢١١٤)		20	0.3	2.34		1.2	20	林林空間の原齋物(コンクリート)(東京的12回名」港研支汚染)の10年後は香放射新漫座を相定
解体廃棄物	S14 デブリ回収廃棄物		DSPスロットプラグコンクリート	コンクリート	多孔状	1.27E+14	据付中	-	切断			50	0.74	1.8		0.74	88	出意要は15000000000000000000000000000000000000
			保温村	金属	多孔状	0.105 . 12	据付中	-	切断			50	0.74	1.8		1.2	20	
			PCV~ v F	金属	塊状	5.100415	据付中	-	切断	外寸1.42m×1.42m×1mh	1.4872	20	0.3	2.34	DI1-0-89/200	1.2	20	江刀谷母 C 问 一 叙义。
			シールドプラグコンクリート	コンクリート	多孔状	1.27E+14	据付中	-	切断	席業衍体官容森	(約1.5)	50	0.74	1.8	別述登理	0.74	88	DSPスロットプラグコンクリートと同一股定
			RPV~ 7 F	金属	塊状		据付中	-	切断	((**)))(0)		20	0.3	2.34		1.2	20	
			RPV下编派	金属	塊状	9.10E+13	据付中	-	切断			20	0.3	2.34		1.2	20	圧力容器と同一設定
Juint 200 - Verdis Within	OB 1 (ELECTRO) (VI (DION CARDY)	KUDION	テフリ取出し関係機器項	並属	現状 (約 (2)		-(今後導人)	-	切町			20	0.3	2.34		1.2	20	
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			組合浸ハーシュライト(AGH)	使結体	粒状(数mm径)	1.27E+13	(追加遮蔽あ	情報なし	専用吸着塔保管								_	
			フェロシアン化コパルトハーシュライト(KH)	焼結体	粒状(数mm径)		り)		(ベント孔あり)のまま						/			
			^{珪チタン酸塩(TSG)}	燒結体	粒状(数mm径)										\sim	_		
			ろ退砂(硅砂)	珪砂・活性炭	粒状(数mm径)													
		SARRY	チャパサイト(IE-96)	燒結体	粒状(数mm径)						(†	青報無し)保管署	容器移し替え後	の保管は想定さ	れていない			
			珪チタン酸塩(IE-911)	燒結体	粒状(数mm径)	211E+13	専用吸着塔保管	情報た日	専用吸着塔保管									
			珪チタン酸塩(EN-101,HS-726,ET-501)	燒結体	粒状(数mm径)		(ベント孔あり)	10 14.9.0	(ベント孔あり)のまま									
			ろ送砂+ゼオライト系:クリノブチロライト	焼結体	粒状(数mm径)						/							
			ろ過砂+活性炭系:アンスラサイト	珪砂・活性炭	粒状(数mm径)	Intra I	the second data just can be	10000			-							
		SARRY-2	ろ送砂+活性炭茶:アンスラサイト	理ジ・活性炭 絶社体	赵伏(数mm径)	情報なし	専用吸着塔保管	情報なし	専用敬着培保官(ペント 7. たい)のナキ									
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	and and the constrained (all	第2七バイル型ストロンチウム除土松屋	(ホインマン(M)(1000) 注チタン酸塩((F-911 FST)(物水(防海が))	体结体	<u></u> 粒状(数mm径)	情報なし	·····································	情報なし									(余水率約50%)	SARRYの実績を兼に
			はチタン酸塩(FST)(タンク浄化)	焼結体	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし								(1)(+0)00/0)	
	510.1 多核種除去装置①(スラワー)		炭酸塩スラリー	炭酸塩	粒状(数mm径)	7.72E+11	HIC保管	フィルタープレス	-									
			鉄共沈スラリー	水酸化鉄	粒状(数mm径)	7.88E+12	HIC保管	フィルタープレス	-								2900	容器収納率、肥水物のかさ密度等より設定
	S10.2 多核種除去装置②(吸着材)		珪チタン酸塩系吸着材(FST,Sr-treat)	燒結体	粒状(数mm径)	5.53E+10	HIC保管											
			活性炭系系吸着材(GAC,AGC,クラレコール)	活性炭	粒状(数mm径)	4.81E+10	HIC保管			外寸2×2×1.5mh								
			高分子系系張着材:水酸化セリウム(ReadE2,Read-B)	樹脂	粒状(数mm径)	情報なし	HIC保管			ベントフィル付保管容器	5.1	80	4.1	5.8	-	1.0		
			高分子系系後置将:イブミジ群模型 4 レート 板板(19C748)	他的	粒((数mm))	Z.30E+10	HICIRE			(遮蔽5cmを想定)			(8675(49))	(股水利)				
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			ゼオライト系系吸着材:銀ゼオライト(AgZ)	焼結体	粒状(数mm径)	4.81E+10	HIC保管											
			酸化チタン(GX194)	焼結体	粒状(数mm径)	4.70E+10	HIC保管											
			$7 \pm \square > 7 > \ll \square < \wedge \vdash (Cs-treat)$	燒結体	粒状(数mm径)	4.65E+10	HIC保管											
	5223多信誉除由装置③()6度カラム)		キレート樹脂系吸着材:水酸化鉄(Ly-RT)	樹脂	粒状(数mm径)	1.26E+11	HIC保管											
	S11 除染装置スラッジ		除染装置スラッジ	硫酸パリウム	粒状(数mm径)	8.17E+12	貯槽D	遠心分離	-								(含水率約60%)	ALPSスラリーと同等と推定
	S12 7 1 Ng	高性能ALPS	ろ送フィルタ(ボリフロビレン製)	倒崩 軸形	多九衣 冬夏分													
			CSフィルタ(ナイロン樹脂製) Srフィルタ(ナイロン樹脂製)	樹脂	多孔状		コンクリートま									/	_	
		RO濃縮水処理設備	ろ過フィルタ (ポリプロピレン製)	樹脂	多孔状		たは金属製の保								/			
		モバイル型Sr除去装置	SSフィルタ(ポリプロビレン製)	樹脂	多孔状	情報なし	官容森(ペント	情報なし	現行の保管容器					/			_	
			・ キラフネート(ポリエーテルサルフォン製)	樹脂	多孔状		1しの リノト ス約		(~> r 11.00 9)00 # #		Г		-			ן ∎	ſ	
		サプドレン他浄化設備	過フィルタ (ポリプ (一間)				74					(清報)」) そ	管容 移 替え	の保 は恋	± tron		harpha t	armatian
			5フィルタ(ナイロ) 悲励製)	1	多孔书													
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	S13英登達翰特爾座 Z ラリー		高型連続装置座スラリー	步醇塩	粒状(数mm径)	情報なし	供道 で メンプ (ベント管付	情報なし	現行のタンク		-	- T `						
			Construction of the Constr	1 C 10 C 10	Can Provenuiti		š)	173 TA -0- 0	(ペント管あり)のまま			-						
	N4 その他水処理設備	高性能ALPS	珪チタン酸塩(HS-726,FST)	焼結体	粒状(数mm径)	情報なし	HIC保管					1						
			活性炭系系吸着材(K-MAC)	活性炭	粒状(数mm径)	情報なし	HIC保管	情報たし	航理たり									
			高分子系系張着村(CRB05,CN4000,READ-B,WA30)	樹脂	粒状(数mm径)	情報なし	HIC保管	101 102 104 10-	X24±14 U									
			ゼオライト系吸着村(AgZ,KW-500G-7)	焼結体	粒状(数mm径)	情報なし	HIC保管											
		RO濃縮水処理設備	オテタン酸塩(HS-72)	焼結体	粒状(数mm径)	情報なし	專用吸着塔保管	情報なし	処理なし						1			
		ッフトレン187尹代説開	でキッイトホ(A51-JHP) 時所ない酸物(JJC-726)	光柏体 持結(水	型(次(数mm径) 数计(数mm容)	時報なし	等用吸着培保管 市田県美花(2)	情報なし	処理なし が用た」									
			本1.7 mm(m(10.1.20) 活性炭系(K-MAC)	->GHD HP 活性炭	515(0000000) 粒状(数mm径)	情報なし	·····································	情報なし	処理なし									
			高分子系(READ-B)	樹脂	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし	N-10-10-15-1								
		モバイル式処理装置	珪チタン酸塩(IE-901)(SFP浄化)	焼結体	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし	外寸2×2×1.5mh	6	80	4.8	-	-	1.2	(含水率約50%)	現在、休止中
			珪チタン酸塩(IE-901)(トレンチ浄化)	焼結体	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし	> F 2 1 /P19 休言容森								
			ゼオライト系(ET201)(トレンチ浄化)	燒結体	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし									
		高性能ALPS検証試験装置	珪チタン酸塩(HS-716)	焼結体	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし									
			高分子系(CRB05,WA30)	樹脂	粒状(数mm径)	情報なし	専用吸着塔保管	情報なし	処理なし									
I			セオフイト糸(AgZ)	炭箱体 使計(4	粒状(数mm径)	情報なし	8月吸着塔保管 本田田美祥(2)	情報なし	<u>処理なし</u> 約mpt.l									
			小岐11.第二款未放着付(AS-UATUH2) その他	統結体 7	业(g(mm佳)	情報なし	毎用吸着哈保官 情報な .	(情報なし) 情報なし。	<u>処理なし</u> 航理なし									
			ゼオライト土要(ゼオライト)	焼結体	粒状(数mm径)	情報なし	未回収	情報なし	処理なし									
			ゼオライト土嚢(活性炭土嚢)	焼結体	粒状(数mm径)	情報なし	未回収	情報なし	処理なし									



<Selection of waste required for hydrogen generation management during storage>

- > Selection of the following waste that could possibly generate hydrogen
 - Dismantled waste that has to be stored and handled underwater, or cut underwater and water is likely to enter into the container.
 - Waste that has not been contained and stored in the container (except waste that has been stored in the container with vent)
- Selected waste and waste objects for evaluation

The following waste among the selected waste is evaluated. In case that waste has different conditions in the same classification, waste is selected from the aspect of materials, the amount of radioactivity in the storage container and water content.

Table 1: Waste that is assumed to require hydrogen management and waste objects to evaluate hydrogen generation

W	aste classification required for	or hydrogen management				
Primary classification	Classification	Type of waste	Waste objects for evaluation	Reason for selection		
Dismantled waste	Reactor pressure vessel (RPV)	RPV Steam dryer Steam separator	Metal waste including RPV, etc.	Metal waste contamination is derived from the accident, radiated from base material, and secondary contamination (equivalent to L2 waste before the accident).		
	Waste generated during fuel debris retrieval	DSP slot plug concrete Heat insulating material PCV/RPV heads, etc.	Concrete waste including DSP slot plug concrete, etc.	Concrete waste (equivalent to L2 waste before the accident and assuming that contaminated waste is derived from the accident,) and it has possibly highwater content.		
Secondary waste generated from water treatment	Mobile Sr. removal equipment (adsorption tower)	Silicate titanium salt	-	(No information on concentration of radioactivity)		
	Multi-nuclide removal equipment (slurry and adsorption tower, etc.)Carbonate slurry Ion coprecipitation slurry, etc.		Carbonate slurry generated from multi-nuclide removal equipment	High level of radioactivity among secondary waste generated from water treatment and the amount of generation is large.		
	Other water treatment facility	High-performance ALPS, RO concentrated water treatment facility, etc. (silicate titanium salt, activated carbon and zeolite. etc.)	-	(The concentration is assumed to be lower than above although there is no information on the concentration of radioactivity.)		

Issues concerning hydrogen generation of secondary waste generated from water treatment during storage can be studied based on evaluation of multi-nuclide removal equipment and carbonate slurry.



<Study of factors for hydrogen generation>

Study on the following factors and the evaluation model effecting evaluation of hydrogen generation rate.

- ✓ Type of radiation (G value)
- ✓ Radioactivity inventory
- ✓ The amount of heat generation and energy absorption ratio
- ✓ Water content rate

Type of radiation (G value)

Nuclides for evaluation are mainly radiated materials and fission products that emit β - and γ -rays. On the other hand, Actinides are α -ray emitting nuclide.

 β - and γ - rays are regarded as equivalent from the aspect of water radiolysis therefore, the common G value is used. Fuel debris is assumed based on the previous case reports and G value that was used for evaluation of mixing α - and γ - rays is used ^[1].

	H_2	eag	Н	H_2O_2	OH	H^{+}	HO ₂
γ	0.43	2.7	0.61	0.61	2.86	2.7	0.03
α	1.3	0.06	0.21	0.985	0.24	0.06	0.22

Table 1: G value to be used for evaluation

[1] Junichi Takagi and Seiji Yamamoto "Simulation study on hydrogen generation by the alpha radiolysis from the fuel debris particles " Proceedings Symposium on Water Chemistry and Corrosion in Nuclear Power plants in Asia - 2017 (AWC 2017)



<Impact parameter setting 1> <u>Radioactivity inventory</u> **Radioactivity inventory (Bq)= Radioactivity per a unit weight (Bq/t) × Waste weight (t)**

•Radioactivity per a unit weight (Bq/t)

Concentration setting of 10-year-decayed radioactivity based on a **maximum concentration** of each waste (demolition, debris collection and water treatment) according to the result of the inventory evaluated by Japan Atomic Energy Agency (JAEA) and Central Research Institute of Electric Power Industry (CRIEPI)^[1].

·Waste weight (t)

Volume inside the container $(m^3) \times Filling$ rate $(-) \times$ Waste density (t/m^3)

Mass setting for each waste is as below.

- Reactor Pressure Vessel (metal waste) : Weight 2.34 ton*
- Waste generating during fuel debris retrieval (concrete)
 : Weight 1.8 ton **
- Secondary waste generated from water treatment : Weight 2.65 ton ***
- Setting of volume inside the container 1.5m3xFilling rate 20vol%xDensity7.8 t/m3=2.34 ton

Setting of volume inside the container 1.5m³xFilling rate 50vol%xDensity 2.4 t/m³=1.8 ton
 Setting of volume inside the container 5.1m³xFilling rate 80vol%=Waste volume 4.1m³, 4.1m³xDensity 1.616 t/m³x0.4=2.65 ton

(0.4(=100wt%-60wt%); Carbonate slurry with water content rate 60xt% by filter pressing is assumed.



** Radionuclide composition was estimated by conservative calculation using the value of the detection limit, although no radionuclide was detected in the waste [30–32].

Note: For some radionuclides (e.g., C-14, Se-79, and I-129), their bimodal distributions are estimated and the range of second distributions (> 0 Bq) is described as a band in the lower graph.

Figure 9. Modeling-calculated composition of radionuclides in carbonate precipitate slurry from the multi-radionuclide removal system.

Fig.1: Example of the radioactivity inventory evaluation (concentration inside the carbonate slurry) ^[1]

[1] Daisuke Sugiyama, Ryo Nakabayashi, Shingo Tanaka, Yoshikazu Koma & Youko Takahatake (2021) Stochastic estimation of radionuclide composition in wastes generated at Fukushima Daiichi nuclear power station using Bayesian inference, Journal of Nuclear Science and Technology, 58:4, 493-506,



<Impact parameter setting 2>

The amount of heat generation and the energy absorption ratio

The amount of heat generation (W) = The amount of heat generation per radioactivity of unit nuclide (W/Bq) × Radioactivity inventory (Bq)

According to 1) the number of Bq per a nuclide (Bq/core) and 2) the amount of heat generation per a nuclide (W/core) reported in "Fuel composition evaluation of the Fukushima Daiichi Nuclear Power Station" (JAEA-Date/Code2012-018) by JAEA, the amount of heat generation per 1Bq of each nuclide (W/Bq) was calculated, and multiplying the calculation result gives by the radioactivity inventory described above. The calculation result of the amount of waste heat generation is given in the below table 1.

The inventory value is a maximum value of the evaluation. On the other hand, another evaluation using values of 95% tile and a mode value was also conducted as evaluation values of secondary waste generated from water treatment widely vary.

Most of radiation energy is known to generate heat. Additionally, early composition products becomes water due to the binding reaction therefore, some parts of the products contribute to decomposition. ^[1] Although there is no specific report case, 10% of the heat generation assumed to contribute to radiation decomposition for evaluation in this project.

Type of waste		The amount of h [W		
		βγ nuclides	α nuclide	
Reactor Pressure Vessel (metal waste)	MAX	7.96	6.98	
Waste generated from collecting fuel debris (concrete)	MAX	10.69	9.82	
	MAX	31.2	25.2	
Secondary waste generated from water treatment	(Reference) 95%ile	8.65	0.225	Considering heat
	(Reference) Mode	0.673	1.01E-4	generated from α nuclic

Table 1: Type of waste and the amount of heat generation

[1]Shinichi Ohno "Hydrogen generation by water radiolysis", RADIOISOTOPES Vol.29,No.8,P.401 (1980)


(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste Study of factors required for evaluation of hydrogen generation

<Impact parameter setting 3>

Water content rate (water volume inside the container)

Water in the container can be source of hydrogen generation. Water volume inside the container is set for evaluation in reference to the following setting.

- In case of dismantled waste
 - ✓ Reactor Pressure Vessel (RPV) (metal waste): Water adhered on the surface
 - Waste generating from collecting fuel debris (concrete): Free water (assuming that water is impregnated at all voids inside) and adhered water.
- In case of secondary waste generated from water treatment Secondary waste generated from water treatment (carbonate slurry): 60vol.% (according to the result of study by TEPCO HD.)
- > Water content rate of each waste (water volume inside the container) is set as below.
 - ✓ RPV(metal waste) :20L*
 - ✓ Waste generating from collecting fuel debris (concrete) :88L**
 - ✓ Secondary waste generated from water treatment :3980L***

* Manufacturing requirements and inspection method for the marginal disposal waste in accordance with the Atomic Energy Society of Japan standard (AESJ-SC-F014:2015): Setting as 20L (adhered water on the surface when storing the L1 waste: Approx. 4-20kg)

** Setting as 88L that totals 10vol% (74L) of voids inside the container and 2mm-thickness in the whole surface of the inner wall surface (14L).

*** Setting as filling rate 80vol% of the storage container and water content rate 60wt% of carbonate slurry by filter pressing.

(Volume inside the container $5.1m^3x$ Filling rate 80vol%= Waste volume $4.1m^3$, $4.1m^3x$ Density $1.616 t/m^3x$ Water content rate 0.6= 3980L)



(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste Study on factors required for evaluation of hydrogen generation

<Hydrogen generation evaluation model>

Evaluation model for calculating hydrogen generation rate of high dose waste

In case of dismantled waste (metal and concrete waste)

Waste and water have existed respectively, which are relatively separated. Setting of water and gas volume (air) depends on volume of waste in the container. The water absorption rate of radiation energy generating from waste depends on type and state of radiation.

In case of secondary waste generating from water treatment

Conditions are set of homogeneously existing fine particles including slurry and adsorbent, and water. Water volume is set from the water content rate depending on waste volume in the container. The water absorption rate of radiation energy generating from waste is set depending on type of radiation.



Fig.1: Conceptual image of the evaluation model for calculation of hydrogen generation rate



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(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste <u>Evaluation on the amount of hydrogen generation</u>

<Evaluation result of hydrogen generation 1>

1) Evaluated by the radiolysis model

2) Evaluated by the linear model

(1 mol of hydrogen is estimated to be equivalent to 22.4L.When hydrogen generates at a constant speed, all amount of hydrogen transfers to the gas phase and concentration of 'drogen increases.



Fig.1: Relationship between concentration and time of the gas phase hydrogen (evaluation by the radiolysis)

Fig.2: Relationship between concentration and time of the gas phase hydrogen (evaluation by the linear model)



(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste <u>Evaluation on the amount of hydrogen generation</u>

<Evaluation result of hydrogen generation 2>

Table 1: Hydrogen generation rate evaluated by the linear model

Waste		Hydrogen generation rate (L/sec)
RPV (metal waste)		2.94E-6
Waste generating from collecting fuel (concrete)	4.08E-6	
	MAX	1.09E-5
Secondary waste generated from water treatment (carbonate slurry)	95%ile	9.72E-7
	Mode	7.04E-8

Table 2: Concentration of equilibrium hydrogen evaluated by the radiolysis

Waste	Concentration of equilibrium hydrogen in the gas phase (%)	
RPV(metal waste)	14.4	
Waste generating from collecting fuel (concrete)	15.1	
	MAX	12.3
Secondary waste generated from water treatment (carbonate slurry)	95%ile	1.49
	Mode	1.31



** Radionuclide composition was estimated by conservative calculation using the value of the detection limit, although no radionuclide was detected in the waste [30–32].

Note: For some radionuclides (e.g., C-14, Se-79, and I-129), their bimodal distributions are estimated and the range of second distributions (> 0 Bq) is described as a band in the lower graph.

Figure 9. Modeling-calculated composition of radionuclides in carbonate precipitate slurry from the multi-radionuclide removal system.

Fig.1: Concentration range of radioactive nuclides (example) ^[1]

- Several digits variations are found in the inventory evaluation values of each nuclide.
- There are large variations in hydrogen generation rate and equilibrium concentration depending on evaluation values.

 Daisuke Sugiyama, Ryo Nakabayashi, Shingo Tanaka, Yoshikazu Koma & Youko Takahatake (2021) Stochastic estimation of radionuclide composition in wastes generated at Fukushima Daiichi nuclear power station using Bayesian inference, Journal of Nuclear Science and Technology, 58:4, 493-506,



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(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste <u>Evaluation on the amount of hydrogen generation</u>

<Comparison of radiolysis with linear models>

Radiolysis model

- It is effective to evaluate on the equilibrium concentration after passing long time in consideration of hydrogenoxygen recombination reaction.
- Assuming that gas-liquid equilibrium of hydrogen concentration instantly establishes, it was conservatively evaluated.

Linear model

- It is effective to evaluate the time of reaching 4% in consideration of 0.4% gas phase volume.
- Hydrogen-oxygen recombination is not considered, therefore the concentration values are excessively conservative caused by the continuous increase of the concentration.



Fig. Comparison of evaluation results of the RPV (metal waste) hydrogen concentration



(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste Evaluation on the amount of hydrogen generation

<Applicable plan of the radiolysis model in combination of the linear model>



Fig.1: Applicable plan of the radiolysis in combination of the linear plan



(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste Evaluation on the amount of hydrogen generation

Issues for the evaluation model and parameter setting>

- There are two evaluation models to evaluate the gas-phase hydrogen concentration.: Radiolysis and linear models.
- ✓ The radiolysis model has characteristics of generating hydrogen and <u>reaching equilibrium</u> <u>concentration of the gas-phase hydrogen</u> in consideration of water recombination.
- In case of the linear model, hydrogen unilaterally generates, therefore the gas-phase hydrogen concentration has no upper limit.
- ✓ For above reasons, the radiolysis model is used to evaluate the gas-phase hydrogen concentration inside the container whether hydrogen concentration exceeds 4% or not.
 - > In case of < 4%, ventilation may not be necessary.
 - In case of >4%, ventilation design is necessary. In this case, the gas-phase hydrogen concentration can be determined by a balance between the amount of hydrogen generation and air exhaustion outside of the system.
- ✓ Therefore, the linear model evaluation will be necessary to design ventilation.
- Assuming that the radiolysis has established in accordance with Henry's law, the gas phase hydrogen concentration is overestimated for the initial established region. Consequently, the linear model is appropriate to evaluate the time of reaching 4%.
- Currently, dynamic behaviors including accumulation and emission of hydrogen generated from waste of concrete and slurry are not reflected into both of the radiolysis and the linear models. A method for reflecting these hydrogen generation evaluation should be considered as a future issue.



(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste Evaluation on the amount of hydrogen generation

<Waste required for hydrogen generation measures>

- The amount of heat generation (inventory) for three kinds of waste: i.e. metal waste including the reactor pressure vessel, concrete waste such as the DSP slot plug concrete, etc. generating from collecting fuel debris and secondary waste generated from water treatment was conservatively evaluated. The result of the evaluation showed that hydrogen concentration inside the container could exceed 4% when sealing the container.
- These wastes could require measures for hydrogen generation including vent filter installation depending on hydrogen generation rate.
- ✓ The amount of heat generation (inventory) is set as maximum values in this estimation. To evaluate practical hydrogen generation rates, it is significant to appropriately set the amount of radioactivity (the amount of heat generation). For practical and reasonable planning of storage and management, it is necessary to evaluate the amount of hydrogen generation based on the latest inventory.



(a) Investigation and evaluation of storage methods [1] Storage methods for storing high dose waste

Summary

Achievements so far now

- Collecting and organizing information on waste characterization required for a storage management method (waste compositions, inventory of radioactivity, type of radiation, water content rate and etc.) and hydrogen generation parameters.
- Waste required for storage management was selected according to the result of the study of waste information and hydrogen generation factors, and evaluation models (radiolysis and linear models) and effects on the parameters were studied.
- Acquiring assumed values of equilibrium hydrogen concentration and hydrogen generation rate in the container depending on type of waste based on evaluation of hydrogen generation amount.
- ✓ According to the evaluation results, installation of an appropriate vent filter is necessary because hydrogen concentration in the container could exceed 4%.

lssues

- The amount of waste heat generation (inventory) is significant to evaluate the value, however the inventory evaluation values currently have variation in several digits. For this reason, it is difficult to evaluate hydrogen generation rate and equilibrium concentration that needs accuracy values.
- The improvement of evaluation accuracy for the waste inventory is needed for more accurate evaluation.



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

- Achievements up to FY2020
- So far now, trouble cases in Japan and overseas have not been investigated and examined for the purpose of reflecting into design of the containers, the storage facilities and the storage methods (system) for the waste of the Fukushima Daiichi Nuclear Power Station (NPS).

Goal

• Examining issues and measures during waste storage on the basis of the past trouble cases.

- Details of implementation (See details on next pages)
- Case study in Japan and overseas, and primary screening
 - Trouble cases during storing the radioactive waste container are surveyed and a reference case is extracted to conduct a detailed survey for waste storage of the Fukushima Daiichi NPS.

Detailed investigation

- Detailed investigation for type of storage containers, information on stored waste, trouble occurrence points and factors, environments of storage facilities, duration of storage are conducted and the result of the survey is examined.
- Study on issues and measures during waste storage
 - ✓ On the basis of the examined results, issues and measures for waste storage are studied.
- Indicators for the achievements of goal
- Completed examining issues and measures for waste storage based on the past trouble cases. (Target at the completion: TRL:2 \rightarrow 3)



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

< Details of implementation >

1. Investigation in Japan and overseas, and the primary screening

The information on the following investigation items is acquired from a wide variety of public information in Japan and overseas. The acquired information is examined to clarify trouble occurrence facilities and factors, etc. According to the investigation results, the primary screening is carried out to extract cases that could reflect to design of the storage container, facility and method (system) of waste generated from the Fukushima Daiichi Nuclear Power Station (NPS).

[Surveyed countries and public information]

- Japan: Website of the JDream, Nuclear Information Archives (NUCIA) of Japan Nuclear Safety Institute (JANSI) and the International Nuclear Information System (INIS) database.
- U.S.: Trouble cases of Nuclear Regulatory Commission (NRC); Report of defects and incompatibility based on the 10 CFR Part 21, and report related to the License Event Report (LER) provided by an operator.
- Europe
 - U.K.: Event notification for the Office of Nuclear Regulation (ONR) in every quarter and report for local communities at the site and stake holders
 - ✓ France: Event notification for the Nuclear Safety Authority (ASN) as needed
 - ✓ Germany: Event notification for the Federal Office for the Safety of Nuclear Waste Management (BASE) on a regular basis.

[Surveyed items]

- Trouble cases of the storage facility for waste from Level 1 certificate (Class C waste) to Level 3 and metal containers.
- Fuel casks, fuel pools, flexible containers and concrete containers are not surveyed.
- In case that a trouble case caused by a vent filter installed for hydrogen management is detected, this trouble case would be extracted as notable case.

2. Detailed survey

Investigation and information acquisition of extracted trouble cases related to occurrence factors including type of the container, storage state of waste, trouble occurrence area and environment of the storage facility as well as additional information on recurrence prevention measures.

3. Study on issues and measures at the time of waste storage

On a basis of the results of detailed investigations, potential issues that would cause a trouble when designing the storage container, storage facility and storage methods of the Fukushima Daiichi NPS are extracted to be considered and studied the measures.



[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

Survey in Japan and overseas, and primary screening

<Survey method in Japan>

According to the below database, key words regarding radioactive waste (waste material, etc.), the storage container (drum, etc.) and the storage facility (storage house, etc.) are entered and retrieved 10 trouble cases (primary screening) of the container and the storage facility of solid waste and during storage.

Database (organized by)	Overviews (survey period)	Surveyed number	Excluded survey case (except cases of solid waste)
JDream (Japan Science and Technology Agency: JST)	 Approx. 70 million literatures in all fields of science technology were acquired. Booklets (abstract journals and literatures) are collected since 1981. The survey was conducted by Japanese key word search. (Duration of survey: From the beginning of registration August 2021) 	0	 Radiation control for nuclear medicine The site improvement by manufacturing jig Report on increasing of storage volume
Nuclear Information Archives (NUCIA)(Japan Nuclear Safety Institute: JANSI)	The archive is intended to widely share the information on nuclear power stations in Japan and operation of nuclear fuel cycle facilities. Report of trouble information (reports to the Japanese government since 1966 in accordance with the regulation) and information on maintenance quality (reporting to the government is not mandatory, but useful information has been shared since September 2003) are registered. (Duration of survey: From the beginning of registration to August 2021)	9	 The leakage was discovered at the storage area for solidification of granular materials where is the intensive environmental facility in the Fukushima Daiichi (Stored materials are not solidification).
International Nuclear Information System (INIS) (Japan Atomic Energy Agency: JAEA)	INIS was established in 1970 aiming at promotion of technology information sharing related to peaceful use of nuclear energy. The INIS participating nations (24 international organizations from 132 nations) provided the nuclear literature information acquired from their own nations for IAEA/INIS headquarter. The INIS headquarter integrated these information and distributed the integrated INIS database for each participant nation. The total number of the nuclear literature provided from participated nations was approximately 4.4 million literatures. (Duration of survey: From the beginning of registration to August 2021)	1	 Radioactive waste treatment at the research institute in Tokai area of Ibaraki Prefecture. Facility management report at the Aomori R&D Center of the Mutsu Research Institute.

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[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities <u>Survey in Japan and overseas, and primary screening</u>

<Survey results in Japan1 : NUCIA >

Nuclear power plant	Date of occurrence	Summary of trouble cases	Primary screening
Tokai II	March 19, 2020	Damage caused by rust generated from a can of the Low-Level Radioactive Waste (LLW) drum bottom which was carried out by the Japan Nuclear Fuel Limited (JNFL).	Not found (Event caused from solidified materials)
Hamaoka	April 23, 2018	The chipped paint and water drops were found on the bottom of the LLW drum(5 cans) carried out by JNFL. This was caused by corrosion developed from cracks inside the drum bottom.	Yes (This event is caused by solidified materials but an issue generated from storage and transportation can be reference.)
Hamaoka	August 21, 2014	The expansion was found on the bottom of the LLW cement solidification drum that is planned to be carried out.	Not found (Event caused from solidified materials)
Shimane	June 13, 2011	Corrosion was found on the bottom of the drum at the solidified waste storage facility, which was filled with the LLW waste.	Yes (Trouble case of stored waste)
Kashiwazaki- Kariwa	November 2, 2007	The drums at the solidified waste storage facility were confirmed to have fallen due to the Chuetsu-oki Earthquake (no contamination).	Not found (Event related to earthquake measures)
Shika I	October 30, 2007	The LLW waste storage drum were inspected after the Chuetsu-oki Earthquake, and consequently the leakage of liquid was found from the drum.	Not found (Event related to earthquake measures)
Kashiwazaki- Kariwa	July 17, 2007	Approximately100 cans of drums at the solidified waste storage facility No.2 building were fallen due to the Chuetsu-oki Earthquake.	Not found (Event related to earthquake measures)
Kashiwazaki- Kariwa	July 24, 2002	The leakage traces of tar-like solidified materials were found on the drum of the solidified waste storage facility No. 2 building.	Not found (Event caused from solidified materials)
Fukushima Daiichi	September 5, 1997	When lifting the drum for condensed waste liquid solidification prior to a carry-out test, the bottom of the drum came out and the content was spilled out from the drum.	Not found (Event caused from solidified materials)



[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

Survey in Japan and overseas, and primary screening

<Survey results in Japan 2: INIS and others>

Facility	Date of occurrence	Summary of trouble cases	Primary screening
Oarai Research Institute owned by the Japan Atomic Energy Agency (JAEA)	January, 2019	[INIS] The leakage traces of asphalt were found out on the four cans of the asphalt solidification filling drum.	Not found (Event caused from solidified materials)
Waste disposal site, Nuclear Science Research Institute owned by JAEA (JAERI)	June 1, 2018 (Application for changes)	[Reference] "Response to application for storage regulation changes to confirm the integrity of the drum at the JAERI waste disposal site: Response to long-term storage of radioactive waste, JAEA" Application for storage regulation changes to confirm the integrity of long-term storage drum at the underground pit and to conduct refiling work.	Not found (This is not a trouble case. Radioactive isotope waste (RI waste) should be screened.

- The case of "To confirm the integrity of long-term storage drum at the underground pit of the JAERI" is not a trouble case but the case was handled on a voluntary basis.
- The waste surveyed was Radioactive Isotope (RI) waste but not included in from Level 1 waste (class C waste) to Level 3 equivalent waste.
- The corrosion of the drum stored in the underground pit was caused by sea breeze and rain water at the temporary building, rain water infiltration when placing concrete slab, and directly adhering the waste to the drum since a polyethylene bag such as waste cloth containing water is broken, etc. (Corrosion cases caused by water contents, etc. of the drum were similar to trouble cases of Hamaoka and Shimane Nuclear Power Plants, therefore, this case was excluded in the detailed survey and there would not be a problem in extracting issues.)

[Other JAEA trouble cases]

- There is no report of the waste storage from the press release about the accident and trouble cases provided by the JAEA website JAEA (since 2005).
- There is no reference case regarding the waste storage from report of the JAEA Mutsu Office (Safety Monitoring Committee, Aomori Prefecture)



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities Survey in Japan and overseas, and primary screening

<Primary screening in Japan>

- Most of trouble cases in Japan were caused by solidified materials after waste were solidified and not caused by waste under storage.
- A trouble case of the Shimane Nuclear Power Plant (NPP) was caused by corrosion of the drum that were filled with the Low Level Waste (LLW). This case was selected to conduct a detailed survey.
- A trouble case of the Hamaoka NPP was caused by paint removal (corrosion) of the waste container after waste were solidified, however storage of the waste was also one of the causes and a similar trouble might occur during storage.
- The following cases were selected to conduct the detailed survey.
 - 1) Shimane NPP (June 13, 2011): Corrosion was found at the drum bottom that was filled with the LLW waste at the solid waste storage facility.
 - 2) Hamaoka NPP (April 23, 2018) : Paint removal and water drop adhesion were found at the bottom of the LLW drum carried out by Nuclear Fuel Limited.
- No trouble case regarding the storage facility was found. Another survey of the Japan standard for the storage facility was added in the detailed survey.



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities Survey in Japan and overseas, and primary screening

<Survey methods in US>

On the basis of the below database, keyword searches were conducted by using keywords related to radioactive materials, metal storage container (drum can and container) and storage facility (storage house, etc.). Trouble cases of the container and storage facility for solid waste, and during storing waste were surveyed by the keyword searches.

Name of database	Overview
Report of defects and incompatibility based on the 10 CFR Part 21	The 10 CFR Part 21, the U.S. regulation, requests to report the U.S. Nuclear Regulatory Commission (NRC) when defects with safety concerns or incompatibility related to the approval and authorization are found in nuclear and radioactive facilities as well as the relevant significant facilities and equipment. The NRC reports since 1995 are available on the NRC website. (Duration of survey: From 1995 to August, 2021)
License Event Report (LER)	The 10 CFR Part 50.73. the U.S. regulation, requests providers of the nuclear facility to create the License Event Report (LER) and submit the report within 60 days when abnormal events and failures are found in the nuclear facilities. The LER should include overviews of the events, clarifications of the causes, analysis of the significance and the correction measures, etc. The LERs since 1980 are available on the NRC website. (Duration of survey: From 1995 to August 2021)
Event Notification Report (ENR)	The NRC requests to submit the Event Notification Report (ENR) the report in accordance with the 10 CFR 50.72 (emergency report case), the U.S. regulation, within 1 hour up to 24 hours depending on the events of the emergency report cases. The ENR is relatively simplified than the LER. The ENR since 1999 are available on the NRC website. (Duration of survey: From 1999 to August 2021)



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

Survey in Japan and overseas, and primary screening

< Survey results in U.S>

Database	Search key words	No. of search	Results of primary screening	Database	Search key words	No. of search	Results of primary screening
Report based on the 10CFR	container	4	These cases are related to transport and the container of battery, etc. There is no survey case.	LER	container AND waste	35	These cases are related to transport and the container of battery, etc. There is no survey case.
Part 21	package	23	These cases are related to transport and work packages, etc. There is no survey case.		package AND waste	51	These cases are related to transport and work packages, etc. There is no survey case.
	gtcc	0	—		gtcc	0	—
	Greater- than-Class C	0	_		Greater-than- Class C	0	_
	Cask (addition)	9	These cases are related to storage casks for spent fuel. There is no survey case.	ENR (addition)	waste container	64	These cases are related to transport, the medical facilities and safety of criticality of fission products. They are exempt from survey case.
	drum (addition)	3	These cases are transport and drum brakes. There is no survey case.	_	waste package	3	These cases are related to the medical facilities. There is no survey case.
	Storehouse (addition)	0			gtcc	8	Seven cases of the searched result are related to spent fuel. So, they are exempt from survey case.
					Greater-than- Class C	1	This is a duplicated data of the search result of gtcc.

 \Rightarrow Applicable case is only one case of the Event Notification Report (ENR).





[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

Survey in Japan and overseas, and primary screening

<Primary screening in U.S.>

- One following case was retrieved by the primary screening. However, this case was not reported as an abnormal event.
 - Name of power plant: Zion
 - > Date of occurrence: February 3, 2015
 - Subject: Functional loss of the safety system due to snow
 - Event number: 50787
 - Event (content of the report)
 - Ventilation parts of air flow inlets that are installed with the vertical type storage casks for spent fuel, MAGNASTOR, manufactured by NAC International Inc. were clogged with snow and the clogging rate was more than 50%: 34 casks in total including 33 storage casks for spent fuel and 1 storage cask for the GTCC waste.
 - Exelon Corporation that is approved to use Zion, submitted the Event Notification Report (ENR) to the U.S. Nuclear Regulatory Commission (NRC) within 24 hours after occurrence of the event in accordance with the 10 CFR 72.75d(1)(i).
 - ✓ On February 4, snow was removed from the ventilation parts and the clogging rate of these 34 casks were recovered to be less than 50%.
 - ✓ After that, Exelon Corporation and NAC International Inc. reviewed the Safety Analysis Report (SAR). As a result of the review, the recovering measure to reduce clogging rate less than 50% was <u>completed within 10 hours</u>, while the SAR requests to complete a recovering measure within 58 hours. Therefore, this case was not regarded as an emergency report case and the <u>ENR</u>-24 hour-report <u>was withdrawn</u> in accordance with the 10 CFR 72.75(1)(i).
- The above event was not a trouble case that can refer to waste of the Fukushima Daiichi to be stored in the building, so that a detail survey will not be conducted.



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities <Survey method in Europe> Survey in Japan and overseas, and primary screening

On the basis of the below database, keyword searches were conducted by using keywords related to radioactive materials, metal storage container (drum can, etc.) and storage facility (storage house, etc.). Trouble cases of the container and storage facility for solid waste, and during storing waste were surveyed by the keyword searches.

Nations	Database (range of survey)	Overview
UKQuarterly report of the Office for Nuclear Regulation (ONR)The report of the management site management website every quarthe INES level exerce (Duration of surver)Report for the ONR municipalities and stake holder groupsThe information of regulatory inspect authorities of municipalities of municipalities of surver)		The report of the nuclear facilities that is selected from among the ONR event reports provided by operators (responsible for the site management) should be submitted to government ministries other than the ONR. These reports are posted on the ONR website every quarter (The report that is not applied to the requirements of the ministry is not disclosed in public even though the INES level exceeds level 1). (Duration of survey: From last quarter in 2000 to first quarter in 2021)
		The information on the nuclear facilities that is subjected to the ONR regulations such as status of the operation management, regulatory inspection and experiences of the event, etc. is examined and the explanation is given to mainly administrative authorities of municipalities with nuclear plants and stakeholders every quarter (or every half year). Occasionally, a report that is not included in the quarterly event notification is provided. (Duration of survey: From 2014 to first quarter of 2021)
France	Occasional event notification of the Nuclear Safety Authority (ASN)	The event reports of the nuclear facilities that are classified as the INES level 1 or more from among the ASN event reports provided by operators (responsible for the site management) are extracted and disclosed on the ASN website as needed (The event reports are generally not available in English). (Duration of survey: From 2009 to as of August in 2021)
ASN annual re	ASN annual report	The information on trends of the ASN regulation activities, status of the nuclear facilities required regulations and others is systematically organized and annually reported (The English report is also provided.). The information on the total number of the report and details of the classification is mainly described. If there is a significant report that should be paid attention, the information on individual event is also described. (Duration of survey: From fiscal year 2009 to 2020)
Germany	Monthly report for the nuclear reactor event of the Federal Office for the Safety of Nuclear Waste Management (BASE)	In case that an event of the nuclear facility occurs, an operator (responsible to the site management) reports the authority of approval in the state and then the state authority of approval reports the Federal Office for the Safety of Nuclear Waste Management (BASE). The monthly report for the nuclear reactor events includes events of nuclear power plants and research reactors that are more than 50kW of thermal output and are summarized the BASE reports (including the INES level 0) provided from each state authority every month (no English version). (Duration of survey: From 2010 to June in 2021)
	BASE annual report for events of the non- nuclear reactor (fuel cycle facility and others)	The BASE annual report of non-nuclear reactor is summarized the BASE reports (including the INES level 0) of events of fuel cycle facilities and others provided by each state authority every year (no English version). Compared with the monthly report of the nuclear reactor events, the annual report of non-nuclear reactor includes only limited information on each event. (Duration of survey: From 2010 to 2020)





[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

Survey in Japan and overseas, and primary screening

<Survey results in U.K.>

Nuclear Power Plant	Day of occurrence	Summary of trouble cases	Primary screening
Dounreay intermediate level radioactive waste disposal facility	November 12, 2002	(INES level 0) A waste container in which spent batteries were stored was put into a flask for waste management; however, a confinement function of the flask was not effective because zinc solution leaked from spent batteries. Eventually, this case caused contamination and accidental exposure.	None (Currently a battery is not a subject to waste management. Details of the battery were not clarified and recognized as small waste.)
West Cumbria high dose waste solidification disposal facility	February 7, 2019	A lid of the container should have been welded to close after high-level waste (HLW) was solidified in the container; however cleaning solution was sprayed for decontamination without closing the lid.	None (A trouble case related to treatment after waste solidification)
West Cumbria THORP reprocessing facility	Discovered an event during April to September 2019	(INES level 1) A welding part of a container that is designed to enclose special nuclear fuel material had a quality defect (insufficient performance of resistance pressure) caused from manufacturing.	None (A trouble case related to manufacturing of the container)

<Primary screening in U.K.> Target event for detailed survey: None



[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

Survey in Japan and overseas, and primary screening

<Survey results in France>

Nuclear power plant	Day of occurrence	Summary of trouble cases	Primary screening	
The La Hague site	March 27, 2013	(INES level 1) When the container (vinyl flexible container) that was packed with radioactive waste was loaded into a transportation vehicle, the vehicle was overturned and the content of the vehicle was exposed.	None (A trouble case of the flexible container)	
Tricastin (Pierrelatte)con version plant (INB 105)	Discovered on February 7, 2019	(INES level 1) As a result of investigating a storage area of metal containers (drums) contained with materials and equipment that were no longer needed (including radioactive materials), the result showed that seal performance of the container was incompatible.	Yes	
Tricastin (Pierrelatte)con version plant (INB 105)	Discovered in July 23 and August 6, 2018	(INES level 1) Two cans of the metal containers (drums) contained with materials and equipment that were no longer needed (including radioactive materials) had lost sealing performance. Contamination had spread outside of the container.	Yes (A trouble case of the waste container was surveyed as the similar site related event)	

<Primary screening in France>

Target event for detailed survey: A trouble event of the Tricastin (Pierrelatte) conversion plant (INB 105)



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[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities Survey in Japan and overseas, and primary screening

< Survey results in Germany>

Nuclear power plant	Day of occurrence	Summary of trouble cases	Primary screening
Isar Nuclear power plant Unit 1	September 30, 2011	(INES level 0) When waste was contained in 200 liter-drum and treated by drying before sealing it, the waste content emitted smoke.	None (A trouble case of pre-storage treatment)
Brunsbüttel Nuclear power plant	August 13, 2014	(INES level 0) Corrosion of multiple drums was found, in which spent contamination removal filters were enclosed. After that, survey was conducted and showed that there is a deficiency in waste management such as handling, storage and transfer of waste.	Yes (Corrosion event of the storage container)
Gundremmingen Nuclear power plant	June 13, 2016	(INES level 0) When low-level waste (LLW) was enclosed in 180 liter-drum and treated by drying before sealing it, the waste content emitted smoke.	None (A trouble case of pre-storage treatment)
Brunsbüttel Nuclear power plant	May 25, 2018	(INES level 0) The 200 liter-drum enclosed spent contamination removal films became deteriorated and was unable to continue to use. When it was opened and the waste content was collected by using a vacuum machine, it caused radioactivity leakage.	None (A trouble case of waste transfer)
Brunsbüttel Nuclear power plant	October 6, 2020	(INES level 0) The containers enclosed dismantled structures (waste metal) generated from the decommissioning were transported to the melt-reprocessing facility. The waste contents were not corresponding to data of the container identification form that was registered in the dismantled material tracking system.	None (A trouble case of data system management system)
Brunsbüttel Nuclear power plant	February 9, 2021	(INES level 0) Specifications of concrete filling material that was used when waste was enclosed into the metal container for final treatment was not complied with designated specifications.	None (A trouble case related to specifications of solidification materials)

<Primary screening in Germany> Target event for detailed survey: A trouble case of the Brunsbüttel Nuclear power plant in 2014.



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[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

Detailed survey

<Detailed survey in Japan:1>

Subject Shimane Nuclear Power Station Verification of the corroded drums				
Date of verification of occurrence	Date of verification of occurrence June 13, 2011 Location of occurrence Solid waste storage facility Building B			

[Circumstances at the time of occurrence]

When drums stuffed with low-level radioactive waste were being moved, it was found that the base of 5 drums was corroded (through holes were present). Thereafter, when external inspection of approx. 23,000 drums that were in storage was carried out, it was found that only 5 drums had through holes in their bases due to corrosion.

[Cause investigation]

The corroded drums and their contents were checked for the following.

- Deterioration of the paint on the inner surface of the drums
- (Peeling off of the plaint due to the waste)
- Checking for water and moisture from the contents of the drums



[Cause]

As a result of checking the corroded drums and their contents it was presumed that water and moisture adhered to the portion of the inner surface of the drum where the paint had deteriorated because of which the portion got corroded and through holes developed.

[Recurrence prevention measures]

Revisions were made to incorporate the following into the internal procedures concerning management of solid radioactive waste.

• In order to prevent deterioration of paint and to prevent moisture adhesion, plastic containers with painted interiors (cylindrical inner container) shall be used.

• Guidelines for re-use of drums and the methods for packaging waste shall be clearly specified.

• If drums in which corrosion has progressed and through holes are likely to develop are identified, the waste shall be transferred to drums that are sound.

Issues of waste storage: Prevention of container damage and deterioration, and water adhesion



[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

<Detailed survey in Japan:2>

Detailed survey

Subject Ha	bject Hamaoka Nuclear Power PlantPeeling off of the paint and adhesion of water droplets at the base of the drums containing low-level radioactive				
Date of verific	cation of	April 23, 2018	Location of occurrence	INFL I ow-level radioactive waste burial center	
occurren	nce	April 23, 2018	Location of occurrence	JNI LLOW-level radioactive waste bunai center	

[Circumstances at the time of occurrence]

When preparations were being made for burying the 960 drums (waste bodies) that were handed over on March 25, it was found that the paint at the base of 1 of the waste bodies had peeled off and water droplets had adhered to it. Thereafter, when external inspection of the remaining 959 waste bodies and of the 928 waste bodies planned to be transferred in May the same year was carried out, it was found that the paint had peeled off and water droplets had adhered to the base of 1 waste body in each of the lots (total 2 waste bodies).

Further, when external inspection of the waste bodies which had been temporarily stored in the burial center for over a year was carried out based on the internal regulations of JNFL from February 25, 2019 to March 12, 2019, it was found that the paint at the base of 1 waste body had bulged and water droplets had adhered to it, and the paint at the base of 1 waste body had bulged, peeled off and water droplets had adhered to it (hereinafter referred to as "peeling off, etc. of paint"

Peeling off, etc. of paint was found in a total of 5 waste bodies. (The radioactivity in the water droplets adhered to the bases of all the waste bodies was lower than the lower limit of detection.)

[Cause investigation]

In the case of 1 of the waste bodies it was found that fine iron powder had adhered to the painted surface during the process of drying the outer surface when the drum was manufactured, which got rusted over time and expanded due to which the paint on the outer surface bulged.

The following was found in the case of the remaining 4 waste bodies.

(1) Cracks on the inner surface of the base (2) Gap between the mortar and the inner surface of the base of the drum (3) Chlorine (CI) on the inner surface of the base

(4) Corrosion from the inner surface of the base reaching up to the outer surface (5) Iron corrosion products and zinc corrosion products formed on the outer surface of the base under the presence of chloride ions (CI-) (6) Calcium (Ca) which is a component of mortar was found on the outer surface of the base (7) Water (water vapor close to the amount of saturated water vapor) is likely to be present on the inside (Document verification) (8) Corrosion from the inner surface of the base was found to have reached the outer surface of the base of the waste body

(One of the through holes was approx. 6mm)

2)Gap

④Progression of

④ Corrosion, peeling off, etc.

corrosion



and shall undergo training on recurrence preventions measures.

Issues of waste storage: Prevention of container damage and deterioration, and water adhesion



③Bleeding water

(1)Crack

Inner surface

Base of the drum

Base of

the drum

Mortar Paint Zinc

Base

Paint

[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

Detailed survey

<Relevant regulations in Japan: 1>

Requirements related to container storage of solid waste have been identified from the waste storage related articles in the Regulations for Commercial Nuclear Power Reactors

• Regulations related to installation, operation, etc. of commercial nuclear power reactors

Article 67 (Records)

In accordance to the provisions of Article 43-3-21, records pertaining to the items mentioned in upper section of the following table must be recorded for each commercial nuclear power reactor in accordance with the topics mentioned in the middle section of the table respectively, and the records must be retained for the periods mentioned in the lower section of the table respectively. (Here the table has been left out, and only the recorded items are indicated.)

- 5 Radiation control records
- h The quantity of nuclear fuel material transported outside the factory or plant, or items contaminated due to the nuclear fuel material, by type, the type of container used for transporting these, and the date/time and route of transportation.
- i The type of radioactive waste disposed at the waste disposal facility or dumped in the ocean, quantity of radioactive substances contained in said radioactive waste, quantity of containers in case said radioactive waste is enclosed in containers or in case it is integrally solidified with the container, and its specific gravity, as also the date, location and method of its disposal or dumping

Article 90 (Disposal carried out at the factory or plant)

According to the provisions of Article 43-3-22 (1), with regards to the disposal of radioactive waste carried out at the factory or plant where the commercial nuclear power reactor licensee has installed the commercial nuclear power reactor, the measures listed in each of the following items must be taken, and the status of implementation of these measures must be verified before disposal.

- 11 If disposal is carried out by the method mentioned in Item 6 c, when radioactive waste is stored and disposed at the storage and disposal facility with radiation hazard prevention effect, it must be carried out as follows.
- a If radioactive waste is enclosed in containers for storage and disposal, necessary measures must be taken to prevent the spread of contamination in case cracks develop in the container or the container gets damaged, such as wrapping said container with material that can absorb the entire enclosed radioactive waste, or installing a pan that can hold the entire enclosed radioactive waste, etc.
- b If considerable overheating is likely to occur due to decay heat, etc. of said stored and disposed radioactive waste, necessary cooling measures must be taken.
- Regulations on technical standards for nuclear reactors for commercial power generation and their auxiliary facilities Article 40 (Waste storage facility, etc.)

The radioactive waste storage facility must be installed in accordance with the following.

i) It must have the capacity to store radioactive waste generated during normal operation.

ii) The structure of the facility must be such that radioactive waste does not easily leak out from it.

iii) The facility must withstand decay heat or heat generated due to exposure to radiation, and must not get corroded considerably due to the impact of chemicals contained in the radioactive waste or due to other load.

2 The commercial nuclear power reactor where the facility for storage of radioactive waste in solid form is installed, **must be installed such that contamination from radioactive waste does not spread.**



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

Detailed survey

<Relevant regulations in Japan: 2>

An example of **"Technical Specifications for Radioactive Waste Management"** specified at the nuclear power plant in accordance with the rules (Technical Specifications of the Shimane Nuclear Power Plant) is given below.

Chapter 6 "Radioactive Waste Management"

Article 86 (Radioactive solid waste management)

3. Each Manager shall verify the following items, and if any abnormalities are found, the Manager shall take necessary measures.

(1) In order to verify the storage conditions of radioactive waste at the storage facility, the Manager (Radiation Control) shall patrol the storage facility once a week, and in addition, shall take stock of the stored items once in 3 months.

(2) The Manager (Operations) shall monitor the storage condition of the spent resin stored in the waste resin tank, etc. and the filter sludge, and shall take stock of the stored items once in 3 months.

(3) In order to verify the storage conditions of the spent control rods, channel boxes, etc. irradiated in the reactor, which are stored in the on-site bunker, the Manager (Fuel & Technology) shall patrol the on-site bunker once a month and in addition, shall take stock of the stored items once in 3 months. Moreover, the Manager shall take stock once in 3 months of the spent control rods, channel boxes, etc. irradiated in the reactor, which are stored inside the fuel pool.

6. The Manager (Radiation Control) shall ensure that at the time of transportation mentioned in the previous section, the dose equivalent rate of the container, etc. does not exceed the value specified by law and the concentration of surface contamination of the container, etc. does not exceed 1/10th of the surface contamination limit specified by law. However, if containers are transported from the zones specified in Article 92 (zones in radiation controlled areas) -1-1, the verification of surface contamination concentration can be omitted.

Issues of waste storage: Inspection (considering a layout of the containers to observe the appearance of the storage container)



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities Detailed survey

<Europe: Detailed survey 1>

Subject	France: Abnormality in the container for radioactive materials and non-conformance at the uranium conversion plant (INES Level 1)					
Date of ve of occur	rification rrence	2017	Location of occurrence	Tricastin (Pierrelatte) uranium conversion plant Shut down in 2008 (Facility registration number INB105)		
[Circumsta The operat material, et exclusively surrounding result of the performance Internationa	[Circumstances at the time of occurrence] The operator, AREVA NC (formerly COMURHEX), started a series of inspection activities pertaining to metal containers (containers in which equipment and material, etc. that are no longer required and which contain radioactive materials are placed) that are stored for several years at the storage buildings exclusively for storing containers with radioactive materials. Essentially, the container was supposed to function as the primary containment barrier and the surrounding structure (shaped like a vault, pit) in which the containers are placed was supposed to serve as the secondary containment barrier. However, as a result of the inspections, non-conformances related to the containment barrier were found in several containers. Further, in spite of concerns about the sealing performance of the containers being impaired, the service entrance of the surrounding structure was not closed. This was classified as Level 1 on the International Nuclear Event Scale (INES).					
[Cause inv The contain Also, the responsible Further, closed.	[Cause investigation] The container with said storage zone issue had not undergone safety analysis. Also, the container had not undergone periodic inspection contrary to the general operation rules of the facility and the perspective of the department responsible for radiation protection on the protection policy. Further, a part of the service entrance of the surrounding structure was not properly closed in reality but was mistakenly believed to have been properly closed					
[Cause] • Inadequate container management • Failure to conduct periodic inspection						
 [Recurrence prevention measures] Improvement in container management Implementation of safety analysis 						

Issues of waste storage: Pre-study of incompatibility management during storage



[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

<Europe: Detailed survey 2>

Detailed survey

 Subject
 France: Loss of sealing performance of the container with radioactive materials at the uranium conversion plant and resulting diffusion of contamination (INES Level 1)

 Date of verification of
 July 23, 2018
 Location of occurrence
 Tricastin (Pierrelatte) uranium conversion plant
 Shut down in 2008

[Circumstances at the time of occurrence]

August 6, 2018

The operator, Orano (formerly AREVA NC) discovered that the sealing performance of a total of 2 containers with radioactive materials (containers in which equipment and materials, etc. that are no longer required and which contain radioactive materials are placed) was impaired. The measuring instrument of the radiological monitor in the vicinity of the storage facility was showing an abnormal reading.

It was decided to collect and treat the contents from the containers in question and transfer them to different containers, and until then the containers were moved inside a robust air-lock facility (surrounding structures with static and dynamic containment functions respectively) in which negative pressure was maintained. However, of the 2 surrounding structures, 1 had not been properly closed, and contamination diffused to the outside from there.

When this event was discovered, maintenance work was not being performed and hence the event did not affect any personnel. Also, based on the level of radioactivity detected outside the surrounding structure, it was concluded that there was no impact outside the site. However, since the surrounding structure not being closed properly and contamination being diffused to the outside is a violation of the general operation rules, and since the discovery was delayed, the event was classified as LEVEL 1 of the International Nuclear Event Scale (INES).

[Cause investigation]

occurrence

As the temperatures are high in said storage zone (inside the surrounding structure) in the summer season, and since hydrofluoric acid was present in the containers in gaseous form, the internal pressure of the container increased and sealing performance deteriorated leading to leakage from the head of the container. The containers had not undergone safety analysis.

The alarm equipment that was supposed to be linked to the measuring instrument for the radiological monitor was not configured properly and as a result did not function, due to which the anomaly was overlooked.

[Cause]

• Inadequate container management

• Increase in internal pressure of the container due to high temperatures in storage zone (inside the surrounding structure) in the summer season, and presence of

hydrofluoric acid in gaseous forms in the containers

• Inappropriate closing of the surrounding structures, and inappropriate configuration of the alarm equipment

[Recurrence prevention measures]

• Improvement in container management

Issues of waste storage: Pre-study of incompatibility management during storage, and study and measures of the cause of inner pressure increase



(Facility registration number INB105)

[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities <u>Detailed survey</u>

<Europe: Detailed survey 3>

Subject	Subject Germany: Inadequacies concerning the management of containers (drums) for solid radioactive waste at Brunsbüttel (INES LEVEL 0)						
Date of verification of occurrence Augus		August 13, 2014	Location of occurrence	Brunsbüttel nuclear power plant (Decommissioning underway)			
[Circumstan	ces at the	time of occurrence))				
Inadequacies disposal of t observed in	Inadequacies were found in the management of containers (drums) for solid radioactive waste. The waste containers in question were generated during the disposal of the decontamination filters in FY2012. One of the containers was significantly corroded (its entire contents were retrieved), and corrosion was observed in some of the other containers as well.						
[Cause invest The operator evaluations of medium level generated du As a result the protectiv	[Cause investigation] The operator carried out a series of evaluations to check whether there are any issues in the handling, treatment and storage of solid radioactive waste. The evaluations were carried out focusing on 6 solid waste storage facilities located in underground zones in the controlled areas. Containers in which low and medium level radioactive waste (mainly, mixed waste containing filter resin, residue from the vaporization process, hazardous chemical substances) generated during operation is placed, are stored in these zones. As a result of evaluation, it was found that corrosion of the container was caused due to increase in moisture, inadequate corrosion prevention measures, the protective coating getting damaged during handling (coating material on the outside and inside), etc						
[Cause]							
	• Inadequate container management						
 Increase in Inadequate 	 Increase in molecule in the storage facility Increase corresion prevention measures 						
 Damage to the protective coating during handling 							
[Recurrence prevention measures]							
Improvement in container management							
 Enhancement of corrosion prevention 							
 Prevention of damage to the protective coating and early detection 							

Issues of waste storage: Prevention of container damage and deterioration, water adhesion and early detection of the container damage



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities <u>Study on issues and measures at the time of waste storage</u>

Source of the primary screening and results of the detailed survey, requirements of the storage containers and storage facilities, trouble cases that were needed the requirements and issues to be reflected into further study are indicated as below.

No.	Requirements	Trouble cases that were needed the requirements	Issues
1	Storage management of radioactive solid waste	 Safety regulations France: Tricastin uranium conversion facility Germany: Brunsbüttel Nuclear power plant 	 Early detection: Regular inspection 1 time/week (considering layout of the container to observe the appearance of the storage container) Pre-study on incompatibility management at the time of storage
2	Fall prevention at the time of earthquake	 Japan: Chuetsu-Oki Earthquake (Kashiwazaki-Kariwa and Shika Nuclear Power Plants (NPP)) 	 Measures to prevent falling when earthquake occurs.
3	Ensuring the integrity of the container such as corrosion management, etc.	 Japan: Shimane NPP, corrosion of drum Japan: Hamaoka NPP, corrosion of drum France: Tricastin uranium conversion facility Germany: Brunsbüttel Nuclear power plant 	 Prevention of container damage and deterioration Prevention of water adhesion Verification and treatment of the content (whether substances including corrosive substance that should be removed exist or not. Pre-assessment and measures for generation factors of inner pressure

[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities <u>Study on issues and measures at the time of waste storage</u>

<Responding to issues>

Responding to issues are described as below.

No.	Requirements	Issues	Proposed measures
1	Storage management of radioactive solid	Regular inspection	Installation of effective inspection passage. Specifically, work efficiency shall be enhanced considering risks of the container integrity; In case of high risks, the inspection passage shall be arranged at a position that can be observed the appearance and sampling inspection shall be conducted for lower risks.
	waste	Pre-study on incompatibility management at the time of storage	Incompatible factors during storage shall be clarified and responding to issues shall be considered in advance. Installing of alarm and confinement system shall be considered if necessary.
2	Fall prevention at the time of earthquake	Fall prevention measures	Fall prevention measures shall be implemented to prevent from falling with acceleration of assumed earthquake and analysis assessment of the measures shall be carried out to make sure that the measures have no problem. When the containers are pilled up, a connecting measure shall be implemented.
		Prevention of container damage and deterioration	A storage method to prevent from causing damage of the container surface (i.e. Method for tilting the container, protruding objects, etc.)shall be carried out. Therefore, workers shall be trained.
	Ensuring the integrity of the container such as corrosion management, etc.	Prevention of water adhesion	 A plastic container shall be used to store waste containing water. At the storage area, temperature and humidity shall be controlled.
3		Verification of the content (whether substances including corrosive substance that should be removed exist or not.	•Corrosion factors shall be verified: Chloride ion that is the cause of metal container corrosion, existence of sulfate ions, dissimilar metals that may cause galvanic corrosion resulting from contact with the metal. Then, neutralization and plastic containers shall be used.
		Pre-assessment and measures for generation factors of inner pressure	The inner pressure factors shall be considered. On the basis of the inner pressure assessment, ,measures (ventilation) shall be installed.



[2] Investigation of requirements of storage containers and facilities : Survey of trouble cases of containers and storage facilities

Study on issues and measures at the time of waste storage

<Concrete measures 13

No.	Requirements Issues		Examples of concrete measures (reference cases)	Proposed plan of concrete measures (draft)
1	Storage management of radioactive solid waste	Regular inspection	[Management of solid waste storage houses in Fukushima Daiichi] Stored drums were verified abnormalities as much as possible by same regular inspection as before the Great East Japan Earthquake. The "Inspection flow of the drum" ^[1] covers a specific method to verify the drums that cannot be observed because of their position.	Regular inspection for existing stored waste is being studied. ^[2] When storage containers and their storage condition are needed to revise in future, specific inspection and verification methods will be crystallized and methods for installing the storage container will be considered.
		Pre-study on incompatibility management at the time of storage	[Performance evaluation of vent filter] (under this project) Filtering events and measures that were needed to consider were studied.	On the basis of specifications for the storage facility, the storage container and waste, incompatibility factors will be clarified and necessity of specific measures will be considered.
2	Fall prevention at the time of earthquake	Fall prevention measures	【Fall analysis of low-level storage drum at the time of earthquake】 The Central Research Institute of Electric Power Industry (CRIEPI) verified the impact of the measures by evaluating bearing force against falling. ^[3]	Existence of waste that may spread radioactive materials due to the fall will be investigated, and radiation exposure assessment due to the spread of radioactivity and fall prevention measures will be conducted.

[1]: TEPCO Holdings, Inc. Fukushima Daiichi NPS, "Storage status of radioactive solid waste (drums, etc.)" July 23, 2018, File No.4, Regulatory Council of Radioactive Waste from Nuclear Facilities (7th),

[2]:" Implementation Plan of Nuclear Facility, Fukushima Daiichi NPS", Maintenance of Nuclear III, Supplementary of Radioactive Waste Management 2". [3]: CRIEPI, "Analysis of Falling Low Level Storage Drum at the time of Earthquake", 2011, the result of research for FY2020 issued by CRIEPI Research Institute for Earth and Engineering, Environment and Science.



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

Study on issues and measures at the time of waste storage

<Concrete measures 2>

Practical reference cases and requirements that are needed to crystallize proposed plan of waste storage for Fukushima Daiichi are indicated as below.

No.	Requirements	Issues	Examples of concrete measures (reference cases)	Proposed plan of concrete measures (draft)
3	Ensuring the integrity of the container such as corrosion management, etc.	Prevention of container damage and deterioration	[Guideline of waste storage management designated by utility companies] Points to consider during storage are described in the waste storage guideline.	At the stage of clarifying waste that will be stored in the container, crystallization of storage methods will be considered (Ex. Protruding objects should not be stored at the container bottom. Board-shaped waste should be stored at the container bottom, etc.).
		Prevention of water adhesion	[Power plant and use of the plastic container] Use of polyethylene inner containers for drums [Nine storage houses in Fukushima Daiichi] Control of temperature and humidity	Water volume to be adhered will be understood and necessity of water treatment will be considered. In case of using inter containers, design and the application method of the container will be considered (When storing high dose waste in the container, deterioration effects caused by radioactivity will be evaluated. In addition, if there is a possibility to generate hydrogen gas, specifications of non-airtight container will be considered). Necessity of temperature and humidity control will be studied when storing waste.
		Verification of the content (existence of corrosion substances)	[Guideline of waste storage management designated by utility companies] Clarified substances to be removed including corrosive substances are provided in the guideline of waste storage.	Investigation of corrosive substances in waste stored, clarification of substances to be removed and refection into the storage management guideline.
		Pre- assessment and measures for generation factors of inner pressure	[Study of storage methods for high-level radioactive materials] (under this project) Evaluation of hydrogen generation amount based on investigation of waste properties and study of ventilation deterioration factors	Detailed investigation of waste properties, study on factors of inner pressure increase and evaluation of the inner pressure (study on specifications of the storage container and storage methods, and conducting of tests, etc.)



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage <Results of the survey> facilities

- Trouble cases of waste storage facilities and metal containers classified from the L1 waste level (class C waste) to the L3 level were searched for survey among the public reports posted on the website including the regulation authority website in Japan and overseas.
- The detailed survey regarding the waste storage was conducted while referencing the following trouble cases.
 - > Japan
 - Shimane Nuclear Power Plant: Corrosion of the drum bottom in which the low level radioactive waste (LLW) is stored.
 - Hamaoka Nuclear Power Plant: Coating removal and water adhesion of the drum bottom for LLW that was carried out by Japan Nuclear Fuel Limited (JNFL).
 - > France

Tricastin (Pierrelatte) Conversion Plant: Loss of airtight functions of the container enclosed with unnecessary materials and equipment

Germany

Brunsbüttel Nuclear Power Plant: Corrosion of the filter installed drums

- According to the results of the survey, issues were clarified in the aspect of storage management, fall prevention at the time of earthquake and ensuring the integrity of the container. For responding to the issues, the following measures were studied and study items for reference cases and specific measures are indicated as below.
 - Storage management: Regular inspection and study on responding to incompatibility cases in advance. ⇒ Crystallization of inspection and confirmation methods
 - Fall prevention at the time of earthquake: Analysis evaluation and connection installment. ⇒ Radiation dose evaluation of scattering waste and fall prevention measures
 - ➤ Ensuring of the container: Prevention of damage and deterioration, confirmation of the content, pre-evaluation of inner pressure and prevention of water adhesion ⇒ Crystallization of storage methods, removal of corrosive substances, and study, evaluation and measures for factors of inner pressure increase



[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities

Summary

Achievements until now

- Trouble cases in Japan and overseas regarding the storage containers and the storage facilities for waste equivalent from L1 to L3 class were searched. Then reference cases of waste storage for the Fukushima Daiichi Nuclear Power Station were clarified by primary screening.
- Among the clarified cases, surveys were conducted regarding the information on the container types, storage waste, trouble occurring place and occurrence factors as well as measures for preventing re-occurrence. The results of the survey were organized.
- On the basis of the organized results, issues when storing waste were considered from the aspect of storage management, fall prevention when earthquake occurs and ensuring the integrity of the container. Reference cases for the measures responding to the issues were studied and the study items were examined to clarify specific measures.

Issues

- According to the specifications of the container and waste properties, it is necessary to specify a storage method that would not cause damage to the container and a removal method for specific substances set by the guideline of the storage management.
- ✓ Study of water management such as the use of inner container when storing waste and others is needed.
- Each waste is required to evaluate fall resistance at the time of earthquake, radiation dose when spreading the content in the container, inner pressure generation, etc. and also specific measures.



[Reference]

(a) Investigation and evaluation of storage methods

[2] Investigation of requirements of storage containers and facilities: Survey of trouble cases of containers and storage facilities <Example of the survey>



Fig.1: Example of the case survey in Japan



Fig.2: Example of the case survey in overseas
Achievements up to FY2020

- The information of the containers with filter ventilation that are \bigcirc used for responding to hydrogen gas generation in foreign countries was organized.
- Events of filters that are needed to consider and the measures Ο for the events were studied while referencing the process flow. Tentative specifications of the filters were set.

Goal

Functions of the filters when high level radioactive waste is stored in the container with the filters and evaluation of service life of filters were examined.

Details of implementation (See details on next pages)

High level of radioactive waste that is needed to be stored in Ο the container with filters was selected on the basis of properties of high level radioactive waste. The site environment in which the storage container with filters is placed and conditions of the storage facility were studied, and test methods and conditions regarding functions and service life of the filters were also considered. In addition, a verification method for maintaining the filter functions during storage of the container with filters was considered.

Guideline to determine goal achievements

Evaluation method for the functions and service life of the filters \cap during storage of the container with filters are clarified. (Target at the completion:TRL: $2 \rightarrow 3$)

Table 1: Measures of hydrogen gas generation conducted in foreign countries

In other countries, the moisture content (free water) is reduced to the extent possible by means of drying and cement olidification, etc. and the generated hydrogen gas is passed through a filter, released from the vent, and managed in

	US	UK	France
Hydrogen gas generation countermeasures	 At the WCS as well as the WIPP disposal sites, a moisture content of 1vol% or less is accepted. (Documents are checked at the disposal sites) The installation of a vent filter is mandatory. (Although TRU waste was disposed in sealed containers in the past, there are instances where the containers were retrofitted with vent filters thereafter) 	- Ferric hydroxide sludge After cement solidification, the w aste surface is capped with grout and then covered with a lid that is designed to diffuse hydrogen through the screw threads. - Barium carbonate slurry The process is almost similar to the process for the ferric hydroxide sludge. A lid integrated with the filter is installed. - Legacy mixed waste Stored temporarily in 3 m ² containers (with a double wall to mitigate the risk of sw elling, lined with concrete to prevent corrosion, and with a filter installed on the lid to release the gas) for several decades. - Plutonium contaminated material · Several super-compacted 200 liter drums are stored in 500 liter drums and stuffed with grout. A lid integrated with the filter is installed.	 Moisture content is decreased by drying (CSD-C: There is a track record for moisture content of 5% or less in the hull and end piece wastes compressed bodies) Although the execution in case of DSC (sludge waste) is undecided, a drying process wherein the moisture content will reduce to 5% or less is being researched and developed.

Table 2: Events of filters that are needed to consider and the proposed measures

Work step	During transfer	During temporary (long-term) storage
Proposed countermeasures*	 Clogging of filter Multiple filters should be installed (multiplexing) so that even if one of the filter gets clogged, hydrogen can be released through the remaining filters. The filter installation structure should be in the form of a labyrinth, etc. so that the contents do not directly reach the filter. As the clogging of the filter is assumed to occur because the contents get stirred up during transfer, a lid with attached filter to be used exclusively during transfer should be used (when storing, that lid should be replaced with a lid with attached filter to be used exclusively during transfer, should be used (when storing, that lid should be replaced with a lid with attached filter to be used exclusively during storage). The impact on the storage container should be assessed assuming a situation wherein there is an explosion with the expected hydrogen concentration (4 vol%). (2) Contamination due to dispersion of the contents For fine particles, a particle collection efficiency equivalent to that of HEPA (99.97% particle collection rate for 0.3 µm particles) is required. Gas component should be processed by means of a ventilation system. Area classifications (red, yellow, green) should be developed based on the assumed dispersion for the filters to contor contamination. Surface contamination of the storage container should be inspected (decontamination should be carried out if necessary) before transfer inside the premises and before storage. 	 (3) Corrosion and deterioration of filter SUS316L or carbon composite materials that are corrosion-resistant in atmospheric environment should be selected for the filter media (sintered porous filter). Corrosion resistance should be confirmed with test pieces of the same material under the same conditions (or harsher conditions). (4) Performance degradation due to filter condensation The temperature and humidity should be controlled using ventilation system of the storage facility to prevent condensation. If the ventilation system of the storage facility stops functioning and condensation occurs, it is critical to restore the ventilation system and leliminate condensation before the hydrogen concentration in the storage container exceeds 4 vol%. It should be ensured that the hydrogen concentration inside the storage container does not exceed 4 vol% within the estimated time that is required for restoring the ventilation system is running at all times by multiplexing the ventilation system or by preparing an emergency pow er supply.

* Whether or not the vent filter can be used and the extent of measures depends on discussions with the authorities.



(a) Investigation and evaluation of storage methods

[2] Investigation of requirements of storage containers and facilities : Performance evaluation of vent filters

<Study of performance evaluation of vent filters>

In accordance with the flow chart below, the deterioration of filters caused by long-term use was studied.



* Evaluation of the filter functions aims to provide the guideline of performance evaluation and methods for selection.









(a) Investigation and evaluation of storage methods [2] Investigation of requirements of storage containers and facilities:

<Waste objects selected from the waste list>

Organizing of input conditions

Demolition waste			Organizin	lizing of input conditions				
S1 RPV	S2 PCV metal	S3 PCV concrete	S4 Metal in the building	S5 Concrete in the building	S14 Debris collection waste	N1 Unit 4 demolition waste	N2 Unit 5 & 6 demolition waste	N3 Other facility demolition waste
RPV	Demolition metal	Demolition concrete	Metal camera	Demolition concrete	DSP slot plug concrete	generated from the normal	generated from the normal	Installed before the earthquake
Steam dryer	Piping	Contaminated dust(surface	Piping	Contaminated dust(surface	Moisturizer	Accident contaminated		Intensive environment
Steam separator	Equipment (cables and	RSW concrete (activation)	Reinforcing steel	T/B wall(inside),floor, column	PCV head			High-temperature incinerator
Upper grid plate	Reinforcing steel	BSW(activation)	R/B equipment	T/B wall(outside)	Shield plug concrete			Incineration building
Core shroud	PCV main part	RPV pedestal	R/B high level radioactive	Rw/B wall(inside),floor,	RPV bottom plate*			Onsite bunker building
Core support plate	PCV inner equipment	Torus concrete(surface	R/B high level radioactive	Rw/B wall(outside)	Equipment for retrieving fuel			Auxiliary building
Other equipment	RSW steel plate	decontamination) Well plug(sunace	equinment R/B nign level radroactive		debris			Common pool building
Fuel support	BSW reinforcing steel	contamination	Additional shielding		Excluded	obiects becau	se 🔺	SPT building
Jet pump	RPV pedestal steel		Turbine main body		thoy are c	auivalant ta v		Incinerator for miscellaneous
Control rod guide pipe	· · · · ·		Generator			quivalent to w	asie	Installed after the
CRD housing			Steam condenser	27	generated	I from the nori	nal	Additional incinerator for
CRD			Moisture separator		reactor.			cesium adsorption
			Water supply humidifier					oil separator
			Other equipment, piping	Exclu	ded objects be	cause they have	ve no contact	Second cesium adsorption
			High level radioactive	with f	uei debris and	contaminated	water.	Third cesium adsorption
			ainina (MS. contom) RW/B equipment, piping					Decontamination
								Reverse osmosis membrane
Excluded o	biects because	thev are						Evaporation concentration
renarded as	fuel debris							system Desalting RO unit
								Desalting equipment
								Cylindrical flange tank
								Horizontal tank
bris, etc.								Square tank
S6 Debris, metal	S7 Debris concrete	S8 Combustibles	S16 Contaminated soil	N5 Others, debris, etc.				Solid waste storage facility
Metals	Concretes	Used protective clothing	Contaminated soil	Incombustible				Slurry stabilization
Piping(metal)	Concrete	Paper	Rubble, collected stones	Noncombustible				Volume reduction equipment
Reinforcing steel	Contaminated dust(surface	Plastic, vinyl	Dredged soil	Equipment(cables,battery,etc				Fuel removal cover for Unit 3
Demolition tanks	contamination)	Lumber	Asphalt	.) Appliances				Fuel removal equipment for
Wires		Cut trees, grass		Building studge(generated by				ะใช้มีผู้ภายกรางกายยายยุงกร
Empty containers		Incinerated ash						Temporary storage facility for
Construction								Contaminated water storage
Wachinen/steel.plate.steel Vehicles, heavy machinery								Decontamination equipment,
								Sludge removel equipment
								cesium adsorption tower



(a) Investigation and evaluation of storage methods [2] Investigation of requirements of storage containers and facilities: Performance evaluation of vent filters Organizing of input conditions <Waste objects selected from the waste list>

3.Secondary waste generated from water treatment									
	S9.1 Adsorption tower (KURION,SARRY)	S9.2 Adsorption tower (mobile Sr.removal equipment)	S10.1 Multi-nuclide remov al system①(slurr	S10.2 Multi-nuclide removal system@(adsorbent)	S10.3 Multi-nuclide removal system® (treatment column)	S11 Decontamination equipment sludge	S12 Filter	S13 Evaporation concentration equipment	N4 Other water treatment system
	1 KURION	Mobile type strontium	Carbonate slurry	Silicide titanate adsorbent(FST,S treat)	 Chelate resin type adsorbent: iron 	Decontamination equipment sludge	High-performance ALPS	Evaporation concentration	High-performance ALPS
:	2 Surfactant modification	Silicide titanate(ISM)	Iron coprecipitation slurr	Activated carbon y adsorbent(GAC.AGC.kurarav.c Polymer system adsorbent.Cer	nal). Ium		Filter(made from polypropylene)		SILICIDE titanate(HS-
	3 Harsh light(H,EH) Sliver impregnated harsh 4	Smealer transfer (IE-		hydroxides(Read E.2 Read.B) Polymer system adsorbent			Cs filter(made from nylon resin) Sr filter(made from nylon resin)		edsorbeot(K-MAC) Polymer system
	Feliocyanide cobalt homb.light/KH)	911.EST)(water.diccbare Silicide titanate(FST)(tank clean-up	A	Polymer system adsorbent iron hydroxide adsorbent (CN4000)			RO concentration water treatment equipment		adsorbentiCRH05.CN4000.Kead: Zeolite adsorbent(AgZ,KW-5000G- Z)
	6 Silicide titanate(TSG)			Chelate resin type adsorbent in hydroxide(Ly.RT) Zeolite type adsorbent silver	on		Filter (made from polypropylene)		RO concentration water treatment equipment
-	B SARRY			Titanium oxide(GX194)		SS filter (made from polypropylene)		SITICIDE TRANATE(HS-72) Sub drain and other filtering
	9 Chabazite(IE-96)			Ferrocyanide cobalt(cesium-tre	at)	4	Ultra filter(polyether		Zeolite adsorbent(A5-
1	0 Silicide titanate(IE-911) Silicide titanate(EN-101,HS-726,ET-	Ex.		cis because	they have b	een	Sub drain and other filtering		Silicide titanate(HS-726) Activated carbon type
1:	501) 2 Filtered sand+zeolite:clinoptilolite	ste	ored in the co	ontainer with	ventilation	holes	Cs filter(made from nylon resin)		adsorbent(K-MAC) Polymer system (Reau-
1:	3 Filtered sand activated carbon: anthracite	an	d filters and	are not plar	ned to be		Sr filter (made from ny lon resin)		Mobile type treatment
1	4 SARRY-2 5 Filtered sand+activated carbon:	rei	placed						uo) Silicide Manale(1E-901)(trench
1	6 Silicide titanate(ET-501)								Zeolite system (ET201, trench
1	7								High-performance ALPS
18	B								Silicide titanate(HS716) Polymer system (CRB-
1	9								05.WA30)
2						****			Ferric hydroxide
2									adsorbent(As-CATCH2)
4 .	0ther waste			ļ				1	Others
	before the earthquake(solid waste	before the earthquake(site	X1 Spent fuel	(2 Debris s	3 Environment impact ubstances(sorted)				Zeolite soil(zeolite)
	1 Operation waste	Granulated	Spent fuel	Debris P	СВ				Zeolite soil(activated carbon soil)
:	2 Operation waste(non- comformity.or	Granulated solidification(concentrati	Nuclear fuel(supplied)	Debris (mixed with R components) n	emoved netals/lead.alminium.etc. sbestos-containing				
	4	solidification(waste resin Tank storage waste regin	Nuclear fuel(before use)	components) n	naterial ead-containing material				

ank storage waste liquid

Tank storage waste

ludae

Control rod

Channel box

uel support

leutron detector

Spent fuel pool

Channel box

Poison curtain

Otters



Excluded objects because they are equivalent to waste generated from the normal reactor.



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<Selection of waste objects based on their properties>

	Waste to be studied	1	2	3	ludament
Large classification	Medium classification	Presence of water	Particulate matter	Contains corrosive ions	Judgment
	S1 Pressure vessel	0	0	0	0
Dismantling waste	S2 Containment vessel metal		0	0	0
	S3 Containment vessel concrete	—	0	—	0
	S14 Waste from debris collection	0	0	0	0
	N3 Dismantling waste from other facilities (After the earthquake disaster)	0	0	0	0
	S9.2 Adsorption vessel ② (Mobile Sr removal equipment)	0	0	0	0
Secondary waste	S10.1 Multi-nuclide removal system ① (slurry)	0	0	0	0
generated from	S10.2 Multi-nuclide removal system 2 (adsorbent)	0	0	0	0
contaminated water treatment	S10.3 Multi-nuclide removal system (3) (treatment column)	0	0	0	0
	S11 Sludge from decontamination equipment	0	0	0	0
	N4 Other water treatment facilities	0	0	0	0

➤ Water existence

- -Underwater cutting is assumed if dismantled waste has high radiation dose.
- •Waste generated from secondary water treatment is assumed to be stored in the condition having water content because of preventing the spread of waste.
- Having particulate substances
 - Particles are assumed to generate from cutting dismantled waste.
 - Particles are assumed to generate from crashing the secondary waste of water treatment.
- ➤ Having corrosive ion
 - It is assumed that both of dismantled waste and the secondary waste of water treatment have experience of contact with sea water.



<Results of the investigation for dismantled waste>

Organizing of input conditions

Wa	ste classification			Waste prop	perties	0		0		3		4			
Large	Intermidiate	Small	Name of waste	Material	Shape	Total radiation	Water	Estimated	Component	Component Particle Occurrence factor		Chloride	Other	pН	Contacted with sea
Demolition waste	S1 RPV		RPV	Carbon steel	Block-shape		7	Draining	Carbon steel	N.A	Cutting and chipping	N.A	N.A	N.A	0
	01101		Steam drver	Stainless steel	Block-shape	9.10E+13		Draining	Stainless steel	N.A	Cutting and chipping	N.A.	N.A.	N.A.	0
			Steam separator	Stainless steel	Block-shape		7	Draining	Stainless steel	N.A	Cutting and chipping	N.A	N.A	N.A	0
Domolition wasta	62 DCV		Demolition metal	Carbon steel	Block-shape		ΝΔ	Draining	Carbon steel	ΝΔ	Cutting and chipping	ΝΔ	ΝΔ	ΝΔ	\cap
Demonuon waste	32 F C V		Pining		Blockshape		ΝΔ	Draining	Carbon steel	ΝΔ	Cutting and chipping	ΝΔ	ΝΔ	ΝΔ	\sim
			Equipment(cables boards etc.)	Iron	Block-shape		ΝA	Draining	Iron	N A	Cutting and chipping	NA	N A	ΝA	$\overline{0}$
			Reinforcina steel	Iron	Block-shape		N A	Draining	Iron	N A	Cutting and chipping	N A	N A	N A	0
			PCV main body	Carbon steel	Block-shape		N A	Draining	Carbon steel	N A	Cutting and chipping	NA	N A	N A	0
			PCV inner equipment	Iron	Block-shape		N.A	Draining	Iron	N.A	Cutting and chipping	N.A	N.A	N.A	Ő
			RSW copper plate		Block-shape		N A	Draining		NA	Cutting and chipping	NA	NA	N A	Ō
			BSW reinforcing steel	Iron	Block-shape		N.A	Draining	Iron	N.A	Cutting and chipping	N.A	N.A	N.A	0
			RPV pedestal reinforcing steel	Iron	Block-shape		N.A	Draining	Iron	N.A	Cutting and chipping	N.A	N.A	N.A	0
Domolition wasto	S3 PCV concrete		Demolition concrete	Concrete	Porous shane		ΝA	Draining	Concrete	ΝΑ	Cutting and chipping	NA	ΝA	ΝA	0
Demonuon waste			Contaminated dust(surface contamination)	Concrete	Porous shane		N A	Draining	Concrete	NA	Cutting and chipping	NA	N A	N A	0
			RSW concrete(activation)	Concrete	Porous shape		N A	Draining	Concrete	N A	Cutting and chipping	NA	N A	N A	0
			RPV pedestal concrete(surface	Concrete	Porous shape		N A	Draining	Concrete	NA	Cutting and chipping	NA	N A	N A	0
		~~~~~	Torus concrete(surface contamination)	Concrete	Porous shape		N.A	Draining	Concrete	N.A	Cutting and chipping	N.A	N.A	N.A	0
			Well plug(surface contamination)	Concrete	Porous shape		N A	Draining	Concrete	NA	Cutting and chipping	NA	N A	N A	0
Demolition waste	S14 Debris		DSP slot plug concrete	Concrete	Porous shape	1.27E+14	12	Draining	Concrete	N.A	Cutting and chipping	N.A	N.A	N.A	0
Demontron waste			Moisturizer	Carbon steel	Porous shape	0.405.42	3	Draining	Carbon steel	N.A	Cutting and chipping	N.A	N.A	N.A	0
	collection waste		PCV head	Stainless steel	Block-shape	9.10E+13		Draining	Stainless steel	N.A.	Cutting and chipping	N.A.	N.A.	N.A.	0
			Shield plug concrete	Concrete	Porous shape	1.27E+14	12	Draining	Concrete	N.A	Cutting and chipping	N.A	N.A	N.A	0
			RPV head	Low allov steel	Block-shape			Draining	Low allov steel	N.A	Cutting and chipping	N.A	N.A.	N.A.	0
			RPV bottom head	Low allov steel	Block-shape	9.10E+13		Draining	Low alloy steel	N.A.	Cutting and chipping	N.A	N.A	N.A.	Ω
			Equipment for retrieving debris	Stainless steel	Block-shape		7	Draining	Stainless steel	N.A	Cutting and chipping	N.A	N.A.	N.A	0
Demolition waste	N3 Other demolition		Radioactive waste incineration facility	N.A	N.A	N.A	N.A	N.A	N.A	N.A	N.A	N.A	N.A	N.A	0
	wooto		Cesium adsorption system(KURION)	N.A.	N.A.	N.A.	N.A	N.A.	1	N.A	N.A.	N.A	N.A	N.A	0
	wasie		Oil separator	N.A.	N.A.	N.A.	N.A.	<u>N.A.</u>	N.A.	N.A	N.A	N.A	N.A	N.A	0
			Cesium adsorption system(SARRY)-2	N.A	N.A	N.A	N.A	<u>N.A</u>	N.A	N.A	N.A	N.A	N.A	<u>N.A</u>	0
			Cesium adsorption system(SARRY-2)-3	N.A.	N.A.	N.A.	N.A.	<u>N.A.</u>	N.A.	N.A	N.A	N.A	N.A	<u>N.A</u>	0
			Decontamination system(AREVA)	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A	N.A	N.A	<u>N.A</u>	<u>N.A</u>	0
			Reverse osmosis membrane treatment	N.A.	N.A.	N.A.	<u>N.A</u>	<u>N.A.</u>	<u>N.A.</u>	N.A	N.A	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	Q
			Evaporation concentration equipment	N.A.	N.A.	N.A.	N.A.	<u>N.A.</u>	N.A.	N.A	N.A	N.A	<u> </u>	<u> </u>	0
			Desalting RO unit	<u>N.A</u>	N.A	N.A	<u>N.A</u>	<u>N.A</u>	N.A	N.A	<u>N.A</u>	N.A	<u>N.A</u>	N.A	<u> </u>
			Desalting apparatus	<u>N.A.</u>	<u>N.A</u>	N.A.	<u>N.A</u>	<u>N.A.</u>	N.A.	<u>N.A</u>	N.A.	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	Q
			Cvindrical flange tank	N.A.	N.A.	N.A.	N.A.	<u>N.A.</u>	N.A.	N.A	N.A	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	<u>0</u>
			Horizantal tank	N.A.	<u>N.A.</u>	N.A.	<u>N.A.</u>	<u>N.A.</u>	<u>N.A.</u>	<u>N.A</u>	N.A	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	<u> </u>
			Square tank	N.A.	N.A	N.A.	<u>N.A</u>	N.A.	N.A.	N.A	N.A	<u>N.A</u>	<u>N.A</u>	N.A	<u>Q</u>
			Solid waste storade facility	N.A.	N.A.	N.A.	<u>N.A.</u>	<u>N.A.</u>	<u>N.A.</u>	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	<u>N.A</u>	<u>0</u>
			Siurv stabilization eduloment	N.A.	<u>N.A.</u>	N.A.	<u>N.A.</u>	<u>N.A.</u>	N.A.	N.A	N.A.	N.A.	<u>N.A</u>	<u>N.A</u>	<u>Q</u>
			Volume reduction equipment		N.A.	N.A.	N.A.	N.A.	N.A.	N.A		<u>IN.A</u>	N.A	IN.A	
			Fuel removal cover for Unit 3		N.A.	N.A.	NA.	N.A.	N.A.	N.A.	N.A.		N A	N.A	<u>0</u>
			Equipment for fuel debris retrievel	<u>Ν.Α.</u> Ν Λ	N A	<u>Ν.Α.</u> Ν Λ	N A	<u>N.A.</u>	N.A.		N A	N A	N A	N A	
			Tomporpry storage facility for fuel debrie	U.A.	N A	NI A	N A		N A	N.A.		N A	N.A		·····
			Contaminated water storage tank	Ν.Α. ΝΙ Λ	N A	ΝΛ	N A	<u>Ν</u> Α.	N A	N A	N A	N A	NI A	N A	<u> </u>
			Containinated water storage talk										13.43	131.23	·
			Decontamination equipment, studge	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A	N.A	N.A	N.A	N.A	0
1			removal equipment												
			Temporary storage facility for cesium	NΔ	NΔ	ΝΔ	ΝΔ	ΝΔ	ΝΔ	ΝΔ	ΝΔ	ΝΔ	NΔ	ΝΔ	0
1			adsorption tower	IN.73.	N.A.	N.A.	11.71.	11.75.	N.A.	11.4	11.7	11.7	11.7	N.A	

The inventory data is cited from back data of inventory estimation results^[1] for each waste (generated from dismantlement, debris collection and water treatment) estimated by Japan Atomic Energy Agency (JAEA/) Central Research Institute of Electric Power Industry (CRIEPI).

#### There are many uncertainties in non-material information.

[1] Daisuke Sugiyama, Ryo Nakabayashi, Shingo Tanaka, Yoshikazu Koma & Youko Takahatake (2021) Stochastic estimation of radionuclide composition in wastes generated at Fukushima Daiichi nuclear power station using Bayesian inference, Journal of Nuclear Science and Technology, 58:4, 493-506,



#### <Compiled results of secondary waste generated from water treatment>

	Waste classification			Waste pro	perties	Φ		0		3		4			
Large	Intermidiate	Small	Name of waste	Material	Shape	Total radiation dose*(advanced	Water content %	Estimated dehydration	Component	Particle diameter(µm)	Occurrence factor	Chloride ions(ppm)	Other corrosive	рН	Contacted with sea
Water treatnebt	S9.2 Adsorption tower(mobile	Mobile type Sr.removal equipment(KMPS)	Silicide titanate(ISM)	Silicide titanate	Granular(several mm diameter)	N.A.	Approx.50 %	Draining	Silicide titanate	N.A.	Pulverized powder	17000	N.A.	N.A.	O
secondary waste	Sr.removal equipment)	Mobile type Sr removal	Silicide titanate(IE- 911 EST)(discharge channel	Silicide titanate	Granular(several mm diameter)	N.A.	Approx.50	Draining	Silicide titanate	N.A.	Pulverized powder	2300	N.A.	N.A.	0
		equipment(KMPS)	Silicide titanate(FST)(tank clean-	Silicide titanate	Granular(several	N.A.	Approx.50	Draining	Silicide titanate	N.A.	Pulverized powder	2300	N.A.	N.A.	0
	S10.1Multi-nuclide	-2	Carbonate slurry	Calcium	Granular(several	7.72E+11	Äpprox.eo	Filter pressing	Calcium carbonate	N.A.	Pulverized powder	13000	N.A.	9.2-11.2	0
	removal system ①		Iron cosedimentation slurry	Iron hydroxide	Granular(several	7.88E+12	Approx.60	Filter pressing	Iron hydroxide	N.A.	Pulverized powder	13000	N.A.	N.A.	0
	S10.2Multi-nuclide	ĺ	Silicide titanate	Silicide titanate	Granular(several	5.53E+10	Approx.50	Draining	Silicide titanate	N.A.	Pulverized powder	13000	N.A.	N.A.	0
	removal system (2) (adsorbort)		Activate carbonate	Carbonate	Granular(several mm.diameter)	4.81E+10	Approx.50	Draining	Carbonate	N.A.	Pulverized powder	13000	N.A.	N.A.	0
	(adsolbent)		Polymer system adsorbent:	Cerium	Granular(several	ΝA	Approx.50	Draining	Cerium bydrovides	Ν Δ	Pulverized powder	13000	Ν.Α	Ν Δ	0
			Cerium hydroxides(Read E- Polymer system adsorbent:	hvdroxides	mm diameter)		%	Diaming				10000			
			iminodiacetic acid type chelate resin/IRC748)	Chelate resin	mm diameter)	2.30E+10	%	Draining	Chelate resin	N.A.	Pulverized powder	13000	N.A.	N.A.	0
			high and a set of the	Chelate resin	Granular(several mm.diameter)	N.A.	Applox.su	Draining	Chelate resin	N.A.	Pulverized powder	13000	N.A.	N.A.	0
			Chelate resin type adsorbent: iron hydroxide(Ly:RT)	Chelate resin	Granular(several mm diameter)	1.26E+11	Approx.50 %	Draining	Chelate resin	N.A.	Pulverized powder	13000	N.A.	6-7	0
			Zeolite type adsorbent: silver	Zeolite	Granular(several mm diameter)	4.81E+10	Approx.50	Draining	Zeolite	N.A.	Pulverized powder	13000	N.A.	N.A.	0
			Titanium oxide(GX194)	Titanium oxide	Granular(several	4.70E+10	Approx.50	Draining	Titanium oxide	N.A.	Pulverized powder	13000	N.A.	N.A.	0
			Ferrocyanide cobalt(Cs-treat)	Ferrocyanide	Granular(several	4.65E+10	Approx.30	Draining	Ferrocyanide	N.A.	Pulverized powder	13000	N.A.	N.A.	0
	S10.3Multi-nuclide removal system 3		Chelate resin type adsorbent: iron hydroxide(Ly:RT)	Chelate resin	Granular(several mm diameter)	1.26E+11	Approx.50 %	Draining	Chelate resin	N.A.	Pulverized powder	13000	N.A.	N.A.	0
	S11 Decontamination system		Decontamination system sludge	Barium sulfate	Granular(several	8.17E+12	Approx.60	Centrifugal	Barium sulfate	N.A.	Pulverized powder	N.A.	N.A.	N.A.	0
	N4 Other water	High-performance	Silicide titanate(HS-726,FST)	Silicide titanate	Granular(several mm diameter)	N.A.	Approx.50 %	Draining	Silicide titanate	N.A.	Pulverized powder	6350	N.A.	N.A.	0
	treatment system	ALPS	Carbonate type adsorbent(K=	Carbonate	Granular(several	N.A.	Approx.50	Draining	Carbonate	N.A.	Pulverized powder	6350	N.A.	N.A.	0
		Polymer system adsorbent(CN400	Polymer system adsorbent(CN4000.READ-	Chelate resin	Granular(several mm diameter)	N.A.	Approx.50 %	Draining	Chelate resin	N.A.	Pulverized powder	6350	N.A.	N.A.	0
			Zeolite type adsorben(AgZ.KW-	Zeolite	Granular(several mm diameter)	N.A.	Approx.50 %	Draining	Zeolite	N.A.	Pulverized powder	6350	N.A.	N.A.	0
		RO concentration	Silicide titanate(HS-72)	Silicide titanate	Granular(several mm.diameter)	N.A.	Approx.50	Draining	Silicide titanate	N.A.	Pulverized powder	6350	N.A.	N.A.	0
		Sub drain and	Zeolite system(A51-JHP)	Zeolite	Granular(several mm.diameter)	N.A.	Approx.50	Draining	Zeolite	N.A.	Pulverized powder	17000	N.A.	N.A.	0
		equipment	Silicide titanate(HS-726)	Silicide titanate	Granular(several mm.diameter)	N.A.	Approx.50	Draining	Silicide titanate	N.A.	Pulverized powder	17000	N.A.	N.A.	0
			Activate carbonate (K-MAC)	Activate	Granular(several mondiameter)	N.A.	Approx.50	Draining	Activate carbonate	N.A.	Pulverized powder	17000	N.A.	N.A.	0
			Polymer system(Read-B)	Chelate resin	mm.diamates)	N.A.		Draining	Chelate resin	N.A.	Pulverized powder	17000	N.A.	N.A.	0
		Mobile type treatment system	Silicide titanate(IE-901)(SFP purification)	Silicide titanate	Granular(several mm diameter)	N.A.	Approx.50 %	Draining	Silicide titanate	N.A.	Pulverized powder	17000	N.A.	N.A.	0
			Silicide titanate(IE-901)(trench clean-up)	Silicide titanate	Granular(several mm diameter)	N.A.	Approx.50	Draining	Silicide titanate	N.A.	Pulverized powder	17000	N.A.	N.A.	0
			Zeolite type(ET201)(trench clean-	Zeolite	Granular(several	N.A.	Approx.50	Draining	Zeolite	N.A.	Pulverized powder	17000	N.A.	N.A.	O
1		ALPS verification	Silicide titanate(HS-716)	Silicide titanate	mm.diameter)	N.A.	Approx 50	Draining	Silicide titanate	N.A.	Pulverized powder	6350	N.A.	N.A.	°
		system	Polymer system(CRB05,WA30)	Chelate resin	mm diameter) Granular(several	N.A.		Draining	Chelate resin	N.A.	Pulverized powder	6350	N.A.	N.A.	0
			Zeolite type(AgZ)	Zeolite	.mm diameter)	N.A.		Draining	Zeolite	N.A.	Pulverized powder	6350	N.A.	N.A.	0
			Ferric hydroxide adsorbent(As- CATCH2)	Ferric hydroxide	Granular(several mm diameter)	N.A.	Approx.50	Draining	Ferric hydroxide	N.A.	Pulverized powder	6350	N.A.	N.A.	0
			Zeolite soil(zeolite)	Zeolite	Granular(several mm diameter)	N.A.	Approx.50	Draining	Zeolite	N.A.	Pulverized powder	N.A.	N.A.	N.A.	0
			Zeolite soil(activate carbon soil)	Activate carbon	Granular(several mm diameter)	N.A.	Approx.50	Draining	Activate carbon	N.A.	Pulverized powder	N.A.	N.A.	N.A.	0

The inventory data is cited from back data of inventory estimation results^[1] for each waste (generated from dismantlement, debris collection and water treatment) estimated by JAEA/CRIEPI. Water quality data was cited from a project report of a guideline of secondary waste generated from water treatment^[2], FY 2015 subsidy project of safety technology measures for nuclear power station.

- Conservative evaluation of water quality (chloride ion, etc.) was conducted and the initial values calculated at beginning
  of treatment were extracted.
- There are many uncertainties in non-material information.

[1] Daisuke Sugiyama, Ryo Nakabayashi, Shingo Tanaka, Yoshikazu Koma & Youko Takahatake (2021) Stochastic estimation of radionuclide composition in wastes generated at Fukushima Daiichi nuclear power station using Bayesian inference, Journal of Nuclear Science and Technology, 58:4, 493-506,

[2] Project report of a guideline of secondary waste generated from water treatment, FY 2015 subsidy project of safety technology measures for nuclear power station. Project report Japan Atomic Energy Agency, March 2016





Fig.1 Illustration of dismantled waste

•Dismantled waste is placed in the inner container and then stored into the storage container.

•The inner container is assumed to be a grid-like rack to avoid accumulating hydrogen.

• Materials of the storage and inner containers are not yet decided. Carbon steel may be used same as for the secondary waste generated from water treatment.

*1: Final report of subsidy project of Decommissioning and Contaminated Water Management in FY2016, Upgrading of Fundamental Technology for Retravel of Fuel Debris and Reactor Internals.

*2: Drawing the illustration while referencing the information provided by TEPCO Holdings.

Fig.3: Illustration of filtering part of the storage container

Filter surface

Inside of the HIC

Filters should be installed in the lid of the storage container by tightening a screw after placing in the housing part with similar uses as the HIC installation.

> The dismantled waste and secondary waste generated from water treatment are stored in dedicated containers installed ventilation.

Filter lid (serving as face plate

Filtering material



Lateral gap (1.9mm)

Retainer ring



<Clarification of storage conditions: Storage container>

Using carbonate slurry that is assumed to be the highest dose, temperatures inside HIC were measured when storing in HIC.^{*1}

Table 1: Result of measuring temperatures of carbonate slurry



*1: Resource from the TEPCO Holdings website (Verification of long-term integrity of HIC, 2018), p.7

Fig.1: Simplified drawing of High Integrity Container (HIC)

## Temperatures inside HIC are almost the same as outside temperatures.

Temperatures inside the container are assumed to be same value as temperatures of the storage environments.

#### IRID



Fig.1: Dismantled waste treatment process*1



Fig.2: Processing of secondary waste generated from water treatment  \ast_2 



#### Placing of waste in the storage building

*1: Final report of subsidy project of Decommissioning and Contaminated Water Management in FY2016, Upgrading of Fundamental Technology for Retravel of Fuel Debris and Reactor Internals.
*2: Drawing the illustration while referencing the information provided by TEPCO Holdings.

> The entire process from placing to storing waste is assumed to be conducted inside the building.

The storage building is assumed to be equivalent to the No.9 storage house for solid waste that is currently under operation.

•Having shielding functions of the building and responding to measures for preventing the spread of radioactive materials.

Installing ventilation system (with air supply)

•Assuming that outside air temperatures are 0 - 40 °C, though the temperature control system is not indicated.



<Definition of deterioration mode>

Functions required the ventilation filter of the storage container are indicated below.

- Functions to release hydrogen that would generate in the process of water radiolytic = Hydrogen permeable function (hydrogen permeable rate)
- Functions to confine particles including radioactive substances= Particle collecting function (collection efficiency)



Fig.1: Conceptual image of filtering function

The deterioration mode defines filter clogging and damage. The cause and event of filter clogging and damage were examined based on the storage environments of solid waste.



#### (a) Investigation and evaluation of storage methods [2] Investigation of requirements of storage containers and facilities: Performance evaluation of vent filters <u>Study of deterioration factors</u>

#### <Study on the causes and events of filter deterioration (damage and clogging)>

The storage environments of solid waste were classified into five environmental categories while referencing the safety evaluation scenario creation method (FEP^{*1}). (Radioactivity in the environment and thermal, stress, chemical environments)
 Assumed causes and events of the filter deterioration were compiled based on each environmental classification.

Environmental classification	Causes	Events	Detailed events	Deterioration mode	
		Hydrogen	A filter would be clogged by hydrogen generation due to radiolysis of solid waste and particle rise.	Clogging	
Radioactivity	Radiolysis	generation	A filter would be damaged by filter clogging and increase of inner pressure, which is caused by hydrogen generation and particle rising due to radiolysis.	Damage	
in environment		Hydrogen embrittlement	Hydrogen embrittlement would occur and damage by a long-term exposure of hydrogen that is generated from radiolysis of water accompanied by waste.	Damage	
	Radioactivity	Radiation deterioration	Filter materials would deteriorate and be damaged by radiation of solid waste.	Damage	
		Thermal alteration	Waste temperature would increase due to decay heat. Heat conduction would increase filter temperature and the filter would be damaged by thermal alteration.	Damage	
Thermal environment	Decay heat	Thermal alteration (container)	Decay heat would increase waste temperature and the head generated by increased temperature would deform the container. A fixed filter would be damaged by stress.	Damage	
		Heat convection	A filter would be clogged by flying particles due to heat convection.	Clogging	
Stress	Vibrations caused by	Impact	When transporting filters or earthquake occurs, the container and filters would be deformed and damaged by impacts of collision / fall, and the filter functions would be lost.	Damage	
environment	earthquake	Dust production by vibration	A filter would be clogged by flying particles which are caused by vibrations of transportation and earthquake.	Clogging	
		Thinning	A filter would be clogged by adhesion of corrosive substances that generate in the thinning process.	Clogging	
Chemical			Oxide film generation	A filter would be clogged by generating oxide film on the surface of a filter fiber due to corrosion and increasing a fiber diameter (decreasing a hole diameter).	Clogging
environment	Corrosion	Salt deposition	A filter would be clogged by accumulating corrosive substances on the filter that is exposed with steam containing corrosive substances for a long-term.	Clogging	
		Electric corrosion	A film would be clogged by corrosion products generated from a filter electric corrosion.	Clogging	
		Microbial film	Microbe such as mold would generate inside/outside a filter and a biofilter would be created, therefore, a filter would be clogged and hydrogen cannot pass through the filter.	Clogging	
		Water film	A water film would be created on the filter surface due to differences of temperatures inside/outside a filter, therefore, a filter would be clogged and hydrogen cannot pass through the filter.	Clogging	
External environment	inside the container and external environments	Freezing	A water film would be frozen when a filter is placed under circumstances below freezing point due to air conditioning failure after a water film is created on the filter surface due to condensation. A filter would be clogged when inside a filter is filled with ice.	Clogging	
		Aerosol deposition	A filter is clogged by accumulating flying aerosol, such as sea salt particles, on the external surface of a filter.	Clogging	
		Spread of particles	A filter would be clogged by flying particles due to the occurrence of the flow caused from differences of pressure inside/outside a filter.	Clogging	
	UV in the environment	UV deterioration	Filter materials would be deteriorated and damaged by UV in the environment.	Damage	
	*** 0 1				

*1 A method for compiling scenario <u>Feature</u>, <u>Events</u> and <u>Process</u> are compiled to create a list.

#### 18 deterioration events in total were extracted.





Tentative specifications were provided under the previous project (FY2018) as below.*1

 $\succ$  Filtering materials  $\rightarrow$  Corrosion resistance materials (SUS316L and carbon composites)

> Particle collection function  $\rightarrow$  Equivalent to HEPA function

> Hydrogen permeable function  $\rightarrow$  Hydrogen concentration in the container should not exceed more than 4%.

*1: Result of subsidy project of Decommissioning and Contaminated Water Management for FY2018 (Research on Treatment and Disposal of Solid Waste), report of FY2019 research meeting p.33



In addition to the above tentative specifications, specifications of general air filters were also investigated. The following items were investigated and the filter requirements were clarified.

- A) Use applications of major manufacturers
- B) Manufacturing methods of filtering materials
- **C)** Particle collection functions
- **D)** Hydrogen permeable functions



<Investigation of air filter specifications: Ex.1>



Fig. 1 Sintered metallic filter manufactured by Poral*1

#### *1 Poral catalog (brochure-filtres-poral.pdf)

#### B) Manufacturing methods and filtering materials

The following materials were used in the term of corrosion resistance.

Sintered metal

Carbon composite

The details of the manufacturing method are not opened to the public. Generally, when using sintered metal, a filter is manufactured by connecting metal fibers and metal particles that are processed in high temperatures.

# Sintering

Schematic representation

of sintering mechanisms :

Fig.2: Conceptual image of connection by high temperature processing

*2 DOE/WIPP 11-3384 "CBFO Approved Filter Vents" U.S. Department of Energy Revision 2 March 2011 https://www.osti.gov/biblio/1041227 *3 Terry Wickland and John Schierloh "HYDROGEN DIFFUSIVITY THROUGH DRUM AND LINER VENTING FILTERS" WM Symposia 1998 **SESSION 27,37** 

https://xcdsystem.com/wmsym/archives//1998/html/sess27/27-37/27-37.htm

#### A) Use applications of major manufactures

Examples of use applications in nuclear facilities are indicated as below.

- •U.S.: TRU waste container at the Waste Isolation Pilot Plant (WIPP) site.
- France: Hull end pieces and sludge waste container at the La Hague site
- U.K.: Waste containers such as for sludge and cladding tube
- -Japan: High Integrity Container (HIC) for secondary waste generated from contaminated water processing

Major manufacturers are indicated as below.

- Nuclear Filter Technology (NFT)
- UltraTech
- Poral etc.

#### C) Particle collection functions

In case of using the US WIPP waste container, specification values of the particle collection functions are indicated for each type of the container.^{*2} The particle collection functions are indicated as an example of certification product as below.

>99.97%@0.3-0.5µmDOP(specified amount of flow: 210ml/min@330 Pa) Equivalent to the HEPA functions.

Table 1: Example of collection functions of the filter for the waste

C	ontainer at the US V	/VIPP.			CH-:Contact-Handled
		CH-TRAMPAC: The TRU waste container that can be directly handled	RH-TRAMPAC: that the TRU waste container that can be remotely handled.	TRUPACT-III: TRAMPACTRU: The waste transfer container for waste	RH-:Remote-Handled TRAMPAC: Transuranic Waste Authorized Methods for Payload Control
	Particle collection function (%)	>99.9	>99.5	>99.9	TRUPACT-III:Transuranic Package Transporter III

### D) Hydrogen permeable functions

When using the filter for the waste container stored at the U.S. WIPP(Waste Isolation pilot Plant), Hydrogen diffusivity mol/sec/mol-fraction is defined as the number of hydrogen mol that mol concentration gradient is set to pass through a filter per one second. These values are measured by tests.*3 Filters with high-hydrogen permeability are included from x/2 to 5x and 25xas a standard of NFT products and  $X(3.70 \times 10^{-6} \text{m/s/mf})$ .

Hydrogen spreading factors of the Poral filters made in France are used as an index. The index of the hydrogen spreading factors  $3.66 \times 10^{-6}$  (m²/s) is included.



#### Clarification of filter requirements

#### <Investigation of air filter specifications: Ex. 2>





Cross-sectional structure of filter Fig. 1 (i) Sintered product in powder form (ii) Long fiber sintered product (iii) Short fiber sintered product (iv) Multi-layered wire mesh sintered product

Sintered metallic filter manufactured by Nippon Seisen Co., Ltd.*1

*1 Nippon Seisen Co., Ltd. Website (https://www.n-seisen.co.jp/products/filter/metal/)

#### C) Particle collection functions

Filtration precision differs depending on the manufacturer or type of filter, but with short fiber sintered product of 0.1 to several hundred µm, sub-micron order particles (equivalent to HEPA) can be collected as well.

Filtration precision is just the official name. It is a value that the manufacturer arbitrarily decides based on the weight removal efficiency with respect to all particles that are of a certain size or larger. Its evaluation method has not been specified in JIS, etc.

#### Use applications

- ·Film forming, synthetic fiber spinning
- ·Synthetic fiber (Filtration of raw materials)
- ·Various air filters (High temperature gas, exhaust gas, etc.)
- •Air equipment filter (Rectification, dispersion)

- Major manufacturers
  - ·Fuji Filter Mfg. Co., Ltd.

- Etc.

- Kodan Kinzoku Co., Ltd.
- ·Nippon Seisen Co., Ltd.
- Nichidai Corporation

Etc.

•Mossfil Co., Ltd.

#### B) Manufacturing methods and filtering materials

A) Use applications of major manufactures

Table 1

Type, manufacturing process, characteristics, filter material of the filter

Туре	Manufacturing process / Characteristics	Filter material
(i) Sintered product in powder form	Sintered metal powder High strength	SUS316, SUS316L, SUS304, Al, Bronze, Cu, Ti, Ni, 600 alloy, etc.
(ii) Long fiber sintered product	Sintered long metal fibers Extensive filtration precision Various shapes can be made	SUS316, SUS316L, SUS304
(iii) Short fiber sintered product	Sintered short metal fibers Sub-micron filtration is possible Suitable for gas and low viscosity fluids	SUS316L
(iv) Multi-layered wire mesh sintered product	Sintered metal wire mesh High strength	SUS316, SUS316L, SUS304, Cu

#### D) Hydrogen permeable functions

Although it has not been clearly specified, since the pores of the sintered product are to the order of several µm, and the porosity is 40 - 80%, the size of the pores is 5µm, air flow rate is 170mL/min, and differential pressure is 294Pa, it is presumed that its hydrogen permeation function is comparable with vent filters used overseas.

(All values are in accordance with Nippon Seisen catalog values)



#### <Investigation of air filter specifications: Ex. 3>

Fig. 1 Ceramic filter manufactured by Nihon Gaishi*1

#### *1 NGK Insulators, Ltd. Website (https://www.ngk.co.jp/product/dpf.html)

B) Manufacturing methods and filtering materials * 2, 3

•Weighing of raw material, mixing, kneading, extrusion, drying, finishing

(sealing), firing, outer diameter polishing and coating

•The following materials are used.

Alumina  $(Al_2O_3)$ : 0.004 - 15µm Zeolite: nm level (molecular sieve) Cordierite (2MgO, 2Al_2O_3, 5SiO_2): 2 - 100µm Silicon carbide (SiO_2): 10µm

# *2 "Systematization of industrial techniques pertaining to porous fine ceramics"

Systematization Report by the National Museum of Nature and Science vol 12 March 2008 *3 Transactions of The Japan Society of Mechanical Engineers Series B 72-721 (2006-9) Use applications

- Deodorization
- ·Removal of foreign material

A) Use applications of major manufactures

- from high-temperature gas
- Gas separation

Etc.

Major manufacturers •NGK Insulators, Ltd. •IBIDEN Co., Ltd. •Noritake Co., Ltd. Etc.

#### C) Particle collection functions

The collection efficiency is low for a particle size in the range of 100nm - 300nm, but as the particles get collected and the pores get filled with them, collection efficiency is almost 100% for all particle sizes. Hence it is presumed that collection efficiency equivalent to HEPA is achieved by controlling the pores.



Fig. 2 Initial performance of DPF (Diesel particulate filter)

(If mock particles with an average particle size of 170nm, number density  $6 \times 10^{6}$ [cm⁻³] are supplied at 10[NL/min])^{*2}

#### D) Hydrogen permeable functions

Although it has not been clearly specified, it is presumed that the hydrogen permeation function is sufficiently high since the resistance at the time of gas permeation is low at 40-60Pa when exhaust gas is blown at 10NL/min.



Particle diameter (nm)

and collection efficiency

Fig. 3 Correlation between the particle size

(The collection efficiency increases when the

amount of particles collected increases) *2

#### <Investigation of air filter specifications: Ex. 4>





Fig. 1 HEPA filter for air-conditioning facilities manufactured by Cambridge Filter Corporation *1

#### *1 Cambridge Filter Corporation website (https://cambridgefilter.com/pdf/)

#### B) Manufacturing methods and filtering materials

Fibers of diameter 1µm or less are spun by means of melt blowing^{*1} or electrospinning^{*2}, with a filling factor of approx. 10% (porosity 90%), and pore size of about several µm.

The material used is mainly glass fiber, but depending on the use application, polymers (polypropylene, olefin, PTFE, etc.) are used.

#### #1 Melt blowing

A technique in which molten polymer is drawn into fibers using high speed and high temperature gas, and the fibers are spun.

#2 Electro-spinning:

A technique in which high voltage is applied between the nozzle in front of a container of polymer solution and the target electrode, due to which the electrically charged polymer solution turns into extremely thin fibers that emerge from the nozzle, and are then spun.



- Use applications
  - ·Clean room
  - ·Food processing plant
  - Hospital
  - Nuclear facility

- Major manufacturers
  - ·Japan Air Filter Co., Ltd.
  - Kondo Kogyo Co., Ltd.
- (Cambridge Filter Corporation)
  - Nitta Corporation
  - •Air Products Ltd.
  - •Oshitari Laboratory Inc.

#### C) Particle collection functions

In the case of HEPA filter, it is specified in JIS8901 that

```
for particles of size 0.3µm, the collection efficiency is 99.97% or more.
```

In the case of ULPA filter, it is specified in JIS Z 8122 that

for particles of size 0.15μm, the collection efficiency is 99.9995% or

#### <u>more</u>.

Also, it is specified that the initial pressure drop is 245Pa or less respectively. In the case of a HEPA filter, when it is used in a nuclear facility in particular, it is specified in JIS Z 4812 that

for particles of size 0.15µm, <u>the collection efficiency is 99.97% or more</u>. Also it is specified that the initial pressure drop is 245Pa or less when there is standard air volume (0.4 - 32m/s), whereas in the case of large air volume type HEPA filter (28-56m/s), the initial pressure drop is 300Pa or less respectively. This performance is not unequivocally decided for the filter material. It can be specified depending on how the material is woven.

#### D) Hydrogen permeable functions

Although it has not been clearly specified, even when the gas flow rate is several tens of m/s, since the pressure drop is only of about several 100Pa, and the porosity and the size of the pores are comparatively large, it is presumed that hydrogen permeability function is sufficiently high.



#### <Results of the investigation related to fine particle collection function>

Filters in the range of medium efficiency to ULPA having HEPA equivalent efficiency comparable to overseas vent filters, can be manufactured using any of the metals, ceramics, glass fiber, polymers that are used in general industry.



*1 Ueshima, "Air filter unit efficiency", Aerosol research, Vol. 4, No. 4 (1989)

#### IRID

<Results of the investigation related to hydrogen permeation function>

The actual hydrogen permeation and diffusion coefficient for the filters used in the general industry has not been mentioned in the catalogs, etc.

However, when compared with overseas vent filters, in principle, the efficiency of metal filters is the same, and that of filters of other materials is presumed to be equivalent or more.







Hydrogen can be discharged easily

Estimation of failure mode



#### Fig. 1 Study of events leading to filter deterioration (damage, blockage)

Based on the input conditions (No. 81-82), since internal temperature = outdoor temperature (0-40°C),

- Thermal alteration/ heat distortion: No notable change in metal/ ceramics/ glass / polymer at said temperature.
- •Oxide film: Oxide film of significant thickness is not formed at said temperature.
- ·Heat convection: Significant heat convection does not occur at said temperature.



According to the input conditions (No. 82), all processes from collection to storage are carried out indoors.
UV deterioration: Does not occur as there is no exposure to UV because the processes are carried out indoors.

Aerosol deposition: Aerosol deposition does not occur due to the air-conditioning system (with air filter).
Microbial film: Microbes do not get mixed up due to the air-conditioning system (with air filter).

#### 18 deterioration events $\Rightarrow$ Of which 7 were excluded leaving 11 events to be studied.



#### Table 1 Study of failure mode scenarios by filter degradation events

		Failure mode scenarios	
	During collection	During transportation	During storage
Hydrogen generation	Unlikely to get clogged due to hydrogen generation in a short time	Unlikely to get clogged due to hydrogen generation in a short time	As hydrogen gas stream resulting from radiolysis of water is generated over a ong period of time, fine particles of solid waste get dispersed, the filter gets clogged and filtration functionality is lost.
Hydrogen embrittlement	Hydrogen embrittlement is unlikely in a short time.	Hydrogen embrittlement is unlikely in a short time.	The generated hydrogen gets absorbed, water is formed and the concentration becomes such that embrittlement occurs during the storage period, leading to damage and thereby loss of filtration functionality.
Radiation deterioration	Radiation deterioration is unlikely in a short time.	Radiation deterioration is unlikely in a short time.	The radiation energy gets absorbed leading to deterioration, the filter material gets damaged and filtration functionality is lost.
Impact	When the lid is being closed, the lid or filter gets distorted / damaged due to the impact of a collision/fall, etc. in the event of an earthquake, and the filter loses its functionality.	During transportation, the container or filter gets distorted / damaged due to the impact of a collision/fall, etc. in the event of an earthquake, and the filter loses its functionality.	During transportation, the container or filter gets distorted / damaged due to the impact of a collision/fall from a height of several m, etc. in the event of an earthquake, and the filter loses its functionality.
Dust formation due to vibrations	Unlikely to get clogged because of dust formation due to vibrations in a short time	Unlikely to get clogged because of dust formation due to vibrations in a short time	During transportation, if the container falls in the event of an earthquake, fine particles scatter and adhere to the surface of the filter. The filter gets clogged and the filtration functionality is lost.
Wall thinning	Wall thinning is unlikely to occur in a short time.	Wall thinning is unlikely to occur in a short time.	Moisture adheres to the filter due to condensation or due to the water in the waste, and along with it chloride ions or particles of waste adhere to the filter as well. Pitting corrosion or crevice corrosion takes place, corrosion products are formed that clog the filter due to which filtration functionality is lost.
Salt deposition	Salt deposition is unlikely to occur in a short time.	Salt deposition is unlikely to occur in a short time.	Water contained in waste gushes towards the outside of the container due to differential pressure or impact, it adheres to the filter and after drying salts get deposited on the filter thereby clogging it, due to which filtration functionality is ost.
Electric corrosion	Electric corrosion is unlikely to occur in a short time.	Electric corrosion is unlikely to occur in a short time.	The filter gets wet due to condensation or due to the water in the waste, and chloride ions adhere to it or the pH becomes alkaline (if the contents contain concrete), as a result of which electric corrosion occurs. The formed corrosion products clog the filter and the filtration functionality is lost.
Water film	Condensation is unlikely to occur in a short time.	Condensation is unlikely to occur in a short time.	The filter gets clogged due to the water film formed because of condensation, and the filtration functionality is lost.
Freezing	Freezing is unlikely to occur in a short time.	Freezing is unlikely to occur in a short time.	During winter, if formation of water film due to condensation and failure of the air conditioning system occur simultaneously, the filter gets clogged due to freezing and the filtration functionality is lost.
Dispersion of fine particles	During collection, the operation is carried out in an environment with uniform pressure. Since the environment is such that differential pressure does not get generated, the filter does not get clogged.	During transportation, the operation is carried out in an environment with uniform pressure. Since the environment is such that differential pressure does not get generated, the filter does not get clogged.	Due to the impact of the ventilation system of the storage facility, a current gets generated inside the storage container, due to which fine particles scatter and adhere to the surface of the filter. The filter gets clogged and the filtration functionality is lost.



: Scenarios wherein occurrence of failure is considered possible

Degradation events and failure mode scenarios were developed considering the input conditions and the processes leading up to collection.



#### Estimation of failure mode

# Table 1 Evaluation of occurrence of degradation by degradation event and filter material of the filter (determination of whether or not the testing methods needs to be studied)

		Metal			Ceramics		Glass					Polymer	
	Stainless steel	Nickel-based allov	Titanium allov	Alumina	Cordierite	Zeolite	E Glass	C Glass	S Glass	D Glass	Polyethylene	Polypropylene	PTFE
Hydrogen generation	-	-	-	_	—	-	—	—	_	-	-	—	—
Hydrogen embrittlement	-	_	x	_	х	x	_	_	_	Ι	_	x	_
Radiation deterioration	1	1	1	1	1	1	1	1	1	1	x	x	x
mpact	х	x	х	х	х	х	х	х	х	х	x	х	х
Dust formation due o vibrations	x	x	х	х	х	х	х	х	х	х	x	x	x
Vall thinning	х	x	х	x	х	x	x	х	х	х	x	х	x
Salt deposition	х	x	х	х	х	х	х	х	х	х	x	х	х
Electric corrosion	_	_	_	_	_	_	_	_	_	—	-	_	_
Vater film	х	x	х	х	х	х	х	х	х	х	x	х	х
Freezing	х	х	х	х	х	х	x	х	х	x	x	х	x
Dispersion of fine particles	-	_	_	_	_	_	_	_	_	_	_	_	_

#### Typical filter materials were selected based on the results of systematic screening of filters

11 degradation events remaining upon checking against the input conditions

- Testing methods do not need to be studied as there is no possibility of this occurring

Testing methods do not need to be studied as even though the event occurs, the extent of progress of generation can be estimated based on the investigation results

(# In this case, it is necessary to evaluate filter performance over time using theoretical formulas)

- x Testing methods need to be studied
- > Hydrogen generation, electric corrosion, dispersion of fine particles are with no impact on filter degradation (study not required)
- > Hydrogen embrittlement is for all except titanium alloy, cordierite, zeolite, and polypropylene, with no impact on filter degradation (study not required)
- Radiation deterioration is I for all except polymer material. Testing methods do not need to be studied as the extent of progress of generation can be estimated
- > Testing methods need to be studied for the above-mentioned events that are marked as x other than those that are marked as 🗸 or -

#### Details pertaining to the reasons that led to such an evaluation are provided in the following slides.



#### Estimation of failure mode

Investigation of the impact that each degradation factor has on the filter

#### **O** Hydrogen generation

•As hydrogen gas stream resulting from radiolysis of water is generated over a long period of time, fine particles of solid waste get dispersed, the filter gets blocked and the filtration functionality is lost.

• The impact at the time of storage of secondary waste generated from contaminated water treatment which has a higher proportion of fine particles than dismantled waste was considered*.

* If the system of secondary waste generated from contaminated water treatment does not have any impact, it is believed that there will be no impact from dismantled waste either.

(An event that occurs inside the storage container)

•Hydrogen is generated from the entire filled layer of secondary waste generated from contaminated water treatment, and that increases the gaps between the waste.

• If the hydrogen gas stream is large, fine particles start floating within the layer.

•The speed at which the particles start floating is the same as the speed at which the particles settle, and can be calculated using the Stokes' Law given below.

Estimation formula^[1]:  $v_s = d_p^2(\rho_p - \rho_f) g / 18\mu$ 

d_p: Average particle size (m), ρ_p: Particle density (kg/m³), ρ_f: Fluid density (kg/m³), g: Gravitational acceleration (m/s²),  $\mu$ : Fluid viscosity (Pa•s)



Internal dimensions 1.3m × 1.3m^[1]

Fig.1 Schematic diagram of the generation of hydrogen stream





# Incipient fluidization speed is calculated and it is studied whether or not fluidization can happen due to hydrogen gas stream

[1]: Assumed using information related to secondary waste produced during contaminated water treatment as reference

[2]: Kohei Ogawa, "Series <New Chemical Engineering> 1, Analysis of the movement of fluids" First Edition, Asakura Publishing Co., Ltd., (FY2011), p51, 58



Investigation of the impact that each degradation factor has on the filter

#### O Hydrogen generation

(Terminal velocity when fine particles float)

Calculated for the range of ordinary waste density and particle size

<Computational conditions>

•Particle size (0.1µm - 10µm)

The size of small particles that can easily get dispersed is set at  $0.1\mu m$  which is inclusive of the standard particle size of 0.3,  $0.15\mu m$  with respect to which the performance of HEPA and ULPA was evaluated.

Particle density (1-8g/cm³)

Set up in the range that is inclusive of carbonate slurry (approx. 3g/cm³) that is assumed to be stored, concrete (2-3g/cm³), stainless steel (approx. 8g/cm³)

 $\,\cdot\, Fluid$  viscosity and density: The values at 20  $^\circ\,\,$  C are used

 $\Rightarrow$  The terminal velocity when fine particles float is in the range of 10⁻⁷ - 10⁻²(m/s)

(Flow velocity resulting from hydrogen generation)

• The hydrogen generation rate is considered to be 1.6E-6(L/s) which is assumed while evaluating the amount of hydrogen generated.

•The hydrogen generation flow rate is calculated using the following formula

Flow rate (m/s) resulting from hydrogen generation

= Hydrogen generation rate/1000 (m³/s) / Cross-sectional area of passage (m²)

The secondary waste generated from contaminated water treatment (□1.9m × 1.9m)^[1], dismantling waste (□1.3m × 1.3m)^[2] are used for the cross-sectional area of the passage
 The cross-section of the container for dismantling waste which has a higher flow rate resulting from hydrogen generation (smaller cross-sectional area) is used for calculation

⇒ The flow rate resulting from hydrogen generation is 9.5E-10(m/s), which is sufficiently lower than the speed at which particles start floating



Fig. 1 Correlation between the flow rate at which fine particles start floating and the flow rate resulting from hydrogen generation

It was determined that the filter degradation effect caused by dispersion of fine particles due to hydrogen generation can be ignored.

[1]: Assumed using information related to secondary waste produced during contaminated water treatment as reference

[2]: Subsidy Project of Decommissioning and Contaminated Water Management in the FY2016 Supplementary Budget

"Upgrading of fundamental technology for retrieval of fuel debris and internal structures" Final Report



Estimation of failure mode

Table 1

Hydrogen generation rate

Investigation of the impact that each degradation factor has on the filter

#### O Hydrogen embrittlement

- Evaluation of the partial pressure of hydrogen in the container
- The pressure inside and outside the container is assumed to be the same.
- The hydrogen pressure is 0.05Pa since the hydrogen concentration in air is 0.5ppm.

The partial pressure of hydrogen inside the container is calculated using PV=nRT based on the hydrogen generation rate in Table 1.

Assuming that the temperature is  $30^\circ$  C, the hydrogen pressure is

0.013Pa in the case of waste (concrete) generated while collecting the pressure vessel and fuel debris, and 0.0006Pa in the case of secondary waste generated from contaminated water treatment. The hydrogen pressure at 300° C becomes approx. double.

Thus, the hydrogen pressure inside the container at 30° C is 0.063Pa in the case of waste (concrete) generated while collecting the pressure vessel and fuel debris, and 0.0006Pa in the case of secondary waste generated from contaminated water treatment.

- In the case of stainless steel and nickel-based alloys
  - Considering the hydrogen pressure inside the container, based on Table 1, the hydrogen solubility of SUS304L is lower than 5ppm even at 300° C*. As the hydrogen solubility of austenitic stainless steel is almost the same for different kinds of steel, it is believed that the solubility is the same in the case of SUS316L as well.
  - •Hydrogen embrittlement does not occur** even if SUS316L absorbs several hundred ppm at room temperature or several dozen ppm at 85° C.
  - •The solubility of nickel-based alloys differs depending on the composition, but at 400° C the volume of Ni is 85%, which is about 5 times that of Fe.

It has been reported*** that hydrogen solubility of Fe does not change at all even in the case of SUS. Based on this, it is estimated that the hydrogen solubility at 300° C when the volume of Ni is 85% is 30ppm or less.

At this concentration hydrogen embrittlement is very unlikely to occur.

As hydrogen solubility tends to decrease as temperature reduces, it is believed that hydrogen embrittlement does not occur even at room temperature.

*Nomura et al, Bulletin of the Japan Institute of Metals and Materials, Volume 15, Issue 9, p.563 - 570 (1976) **Omura, Nakamura, Materials and Environment, 60, 241-247 (2011)

*** Yoshida et al, Bulletin of the Japan Institute of Metals and Materials, Volume 11, Issue 7, p.533 - 548 (1972)





Fig. 1 Correlation between hydrogen solubility of SUS304L and hydrogen pressure*



Fig. 2 Impact of surface concentration of hydrogen on hydrogen embrittlement characteristics **



#### Ti

Based on Table 1, hydrogen solubility of Ti is 10ppm at room temperature, and that of Ti for industrial use (containing Fe: 0.3%) is 100 mass ppm. Considering that hydrogen embrittlement occurs at a concentration of 100 - 300 ppm, hydrogen embrittlement is likely to occur at room temperature depending on the type of Ti.*

* Fujii et al, Materials and Environment, 60, 248 - 253 (2011)



Table 1 Amount of hydrogen content in pure titanium ASTM Gr. 3 for industrial use which contains 0.3% Fe, and its phase structure*

Table 1 An example of a report on a film with hydrogen barrier function

Type of membrane	Rate of reduction of amount of hydrogen permeation						
$AI_2O_3$	10 ~ 10000						
TiC	10 ~ 10000						
TiN	10 ~ 10000						
$Cr_2O_3$	10						
BN	100						

**98** 

#### In the case of ceramics

Alumina which is a ceramic is capable of reducing the amount of hydrogen permeation. Hence it is being considered for use as a film for suppressing hydrogen embrittlement. An example of a report on a film with hydrogen barrier function is shown in Table 1**.

It has been reported that alumina does not get embrittled in a hydrogen environment***

There were no reports on hydrogen embrittlement of cordierite and zeolite.

** NEDO Fundamental Research Project on Advanced Hydrogen Science Interim Evaluation Report (2008) *** JST / Strategic Funds for the Promotion of Science and Technology, "Research on the function of hydrogen in environmental embrittlement of structural material", 2001 – 2002: Term 2



#### Glass fiber

There were no reports on the degradation behavior of glass fiber due to hydrogen. It has been reported that crystalline quartz glass fiber does not get embrittled in a hydrogen environment*

* JST / Strategic Funds for the Promotion of Science and Technology, "Research on the function of hydrogen in environmental embrittlement of structural material", 2001 - 2002: Term 2.

#### Polymer

The High Pressure Gas Safety Institute of Japan has reported that polyethylene can be used for hydrogen gas pipes.**

There are no reports on polypropylene.

PTFE is generally recognized for use in hydrogen gas environment.***

** Tabata, Hydrogen Energy System, Vol 35, No. 4, (2010) *** Source: Website of the Chunichi Giken Products Co., Ltd.

Fluid	PTFE	CS	SUS 304	SUS 316	Brass	Penetration		
Di-isobutylene	-	-						
Diethylphthalate	•	-	•	•	•			
Carbon tetrachloride		×	$\triangle$	$\triangle$	$\Delta$			
Dioctyl phthalate				•				
Cyclohexanone		-			—			
Cyclohexane		•	•		•			
Dimethylaniline	•	-	-	-				
Dimethyl phthalate		-	-	-				
Potassium dichromate		-			-			
Oxalic acid	•	×	Δ		×			
Bromine water		×	×	×	×			
Nitric acid 10%	•	×	Δ	Δ	×			
Nitric acid 70%	•	×	Δ	Δ	×			
Ferrous nitrate	•	-	•		-			
Ferric nitrate	•	×		•	-			
Potassium nitrate	-	×	$\Delta$	•	-			
Calcium nitrate	•	•		•				
Silver nitrate		Δ		•	$\Delta$			
Sodium nitrate			$\Delta$	$\Delta$	$\Delta$			
Zinc acetate		•	•		•			
Zinc chloride		×	Δ	•	×			
Vinegar	•	×	Δ	•	×			
Mercury	•			•	×			
Magnesium hydroxide				•	-			
Hydrogen gas				•		С		
Steam				•	Δ	A		
		$\Delta$	-	$\Delta$	$\Delta$			

•: Good △: Can be used for a limited age of use ×: Not recommended —: Test data not available

Fig. 1 Usability of PTFE in various environments**

#### Estimation of failure mode

- Investigation of the impact that each degradation factor has on the filter
- O Radiation deterioration

#### Radiation generated from waste

• The nuclides selected during the hydrogen generation evaluation are considered as radiation emitting nuclides.

•Alpha rays: Energy is high at 5 to 7MeV, but its impact can be ignored as its range is short (several cm in air).

•Beta rays and gamma rays: Its range is long, and hence it is necessary to consider that radiation is received even from waste that is at a distance from the filter.

#### Radiation impact on various materials

(1) Metal, ceramic material (stainless steel, nickel-based alloy, alumina, etc.)

- Stainless steel and nickel-based alloys are extensively used inside the reactor. Hence it is believed that there are no issues.
- Upon conducting a three-point bending test after subjecting alumina to electron beam irradiation of 300MGy, a slight change in strength was observed.

The change was about 15% at most (Fig. 1), and hence it is believed that this issue can be avoided by selecting material with some extra margin [1].

#### (2) Glass fiber

• When glass is subjected to radiation exposure, its mechanical strength changes. Depending on the elements that glass contains, the mechanical strength either decreases or increases [2].

However, as glass material is not ductile, it is believed that degradation due to radiation exposure is not an issue.



Fig. 1 Results of the test on irradiated alumina material [1]

[1] Michikazu Kincho and others "Irradiation Effects on Alumina Ceramics for Vacuum Duct" 41st Session of the Vacuum Society of Japan Proceedings, Vacuum,

P131. Vol. 44, No. 3, 2001

[2] Norihide Takeyama "The Integrated Design Approach of the Lens-Based Space Optical Systems" Optics, Vol 40, Issue 7, P. 331 (2011)



- Investigation of the impact that each degradation factor has on the filter
- **O** Radiation deterioration

#### Radiation impact on various materials

(3) Organic material (Polyethylene, PTFE, etc.)

- When organic material is subject to radiation exposure, its mechanical strength changes [3].
- When the HIC lining is simulated, polyethylene is exposed to gamma rays at 60° C, and the environment is changed (air, water, iron hydroxide, calcium carbonate, magnesium carbonate), breaking elongation decreases [4]
- In the case of PTFE, radiation resistance is even lower than polyethylene, and mechanical strength reduces with a lower radiation dose [5].

#### Summary

Since the mechanical characteristics of organic material change due to radiation, the usage conditions need to be carefully studied.





Fig. 13 Changes in the maximum tensile strength (figure on the left) and the maximum length (figure on the right) of PE and PTFE when exposed to various types of radiation



Fig. 2 Results of material testing of the irradiated polyethylene PTFE [5]

[3] Seiichi Saiki and others "Special Issue New Radiation Chemistry (Basic edition) 9 Effects of ionizing radiation for polymers" RADIOISOTOPES, 66, 469–477 (2017)
 [4] Results of tests related to radiation deterioration of polyethylene National Institutes for Quantum and Radiological Science and Technology (February 19, 2018) (<u>https://www2.nsr.go.jp/data/000223970.pdf</u>)

[5] Masayuki Sekiguchi and others, "Evaluation of the deterioration of polymer material due to X-ray irradiation and the absorbed dose" Tokyo Metropolitan Industrial Technology Research Institute Research Report No. 7 (2004)



- Investigation of the impact that each degradation factor has on the filter
- Impact

#### Events that have an impact

During transportation, the likelihood of the container or filter getting distorted / damaged due to the impact of a collision/fall, etc. in the event of an earthquake, and the filter losing its functionality, are studied.

#### Waste storage container

Specifications are assumed considering only the metal part

The weight of the storage container is calculated based on the above-mentioned specifications. The study is conducted assuming 4 ton and 7 ton.

Table 1Material, external dimensions, etc. of<br/>the waste storage container (assumed)

	For dismantling waste	For secondary waste generated from contaminated water treatment
Material	Carbon steel (density 7.8g/cm ³ )	Carbon steel (density 7.8g/cm ³ )
External dimension s	1.42m × 1.42m × 1.0m ^[1]	2.0m × 2.0m × 1.5m ^[1]
Thickness	0.06m	0.05m
Vent part	Φ100mm×1	Ф100mm × 2



*1: Subsidy Project of Decommissioning and Contaminated Water Management in the FY2016 Supplementary Budget "Upgrading of fundamental technology for retrieval of fuel debris and internal structures" Final Report

*2: Storage container is assumed based on information related to secondary waste produced during contaminated water treatment



#### Estimation of failure mode

Investigation of the impact that each degradation factor has on the filter

#### O Impact

#### Work that is believed to have lead to an impact

- ✓ Fall after the storage container is lifted up (including earthquakes)
- ✓ Lateral collision of the storage container due to swinging while the storage container is being transported or shaking during an earthquake





#### Impact on the filter

- ✓When the storage container (4 ton, 7 ton) falls from a height of several m, the filter is likely to get distorted/damaged and lose its functionality due to the impact of the fall or due to distortion of the lid of the storage container. Further, the condition of the distortion / damage differs depending on the height from which the fall occurred, the speed of movement, the portion that comes in contact with the ground (storage container falls with its bottom facing down, gets overturned, etc.), the way in which the filter is fixed, etc.
- ✓ If there is a collision, the filter may get distorted / damaged and is likely to lose its functionality. Further, the condition of the distortion / damage differs depending on the the speed of movement when the collision occurs, the portion that collides, the way in which the filter is fixed, etc.
- ✓ It needs to be considered that depending on the specifications of the inner container, the specifications of the place where the filter is installed (shape of the installation location, whether or not a baffle plate is installed, its specifications, etc.), the waste inside the storage container could move up and down due to which the filter could get distorted / damaged or blocked.

[1] Quoted from FY2020 Final Report (March 2021) on the "Subsidy Project of Decommissioning and Contaminated Water Management in the FY2018 Supplementary Budget (R&D for Treatment and Disposal of Solid Wastes)", and edited.

[2] Quoted from FY2020 Research Report (Final Report) on the "Subsidy Project of Decommissioning and Contaminated Water Management in the FY2018 Supplementary Budget (R&D for Treatment and Disposal of Solid Wast and edited.



(a) Investigation and evaluation of storage methods [2] Investigation of requirements of storage containers and facilities

Estimation of failure mode

Buoyance F

Initial velocity

resulting from

vibrations vo

Investigation of the impact that each degradation factor has on the filter

#### O Dust formation due to vibrations

During transportation, due to vibrations in the event of an earthquake, or if the container falls, fine particles scatter and adhere to the surface of the filter. The filter gets blocked and its filtration functionality is lost.

(Method of evaluating dust formation due to vibrations)

The highest reaching point of the fine particles was evaluated Fluid resistance R_f using a physical model in which it is assumed that the fine particles gain upward initial velocity due to the vibrations, leading to an upward motion while being subject to fluid resistance and buoyance in the gravitational field. (Figure 1)

(Initial velocity resulting from vibrations during transportation or in the event of an earthquake)

Seismic ground motion: Vibration acceleration during the Great East Japan Earthquake 2933 (gal=cm/s²)^[1]

Period of vibration  $0.3(s) \Rightarrow$  Initial velocity: approx. 4.5m/s

Vibrations during transportation ^[2]:

Vibration acceleration during transportation 1.5 (m/s²)

Period of vibration 0.6 to  $20(Hz) \Rightarrow$  Initial velocity: 0.27 to 1.6m/s

⇒ Conservatively considered to be 1.6m/s



Fig. 3 Sweep acceleration during vibration testing at the time of transportation^[2]

[1] Quoted from the National Research Institute for Earth Science and Disaster Prevention, K-NET & KiK-NET, Earthquake Alert, Maximum acceleration distribution map [2]: Kohei Ogawa, "Series <New Chemical Engineering> 1, Analysis of the movement of fluids" First Edition, Asakura Publishing Co., Ltd., (FY2011), p51



Investigation of the impact that each degradation factor has on the filter

#### O Dust formation due to vibrations

• The more the density and larger the particle size the higher is the reach. A maximum distance of 9.23cm is reached due to vibrations during transportation and a maximum distance of 46.7cm is reached due to seismic motion.

•Based on the distance between the waste and filter, the seismic motion is likely to have an impact on clogging of the filter.



IRID

Estimation of failure mode

Investigation of the impact that each degradation factor has on the filter

#### O Wall thinning

(1) Metal filter

Since crevice corrosion takes place at a potential lower than pitting corrosion, wall thinning of the metal filter was evaluated based on crevice corrosion. According to Figure 1, under atmospheric environment the chloride ion concentration that gives rise to crevices in SUS316L is 500ppm or more.

As nickel-based alloys and Ti alloys are highly corrosion resistant, the concentration at which crevice corrosion occurs in these is presumed to be higher, but its occurrence cannot be denied if it is assumed that particles originating from waste or corrosive ions, water adhere to them under storage environment.

Under alkaline environment, if stainless steel becomes a strong alkali as shown in Figure 3, its corrosion resistance decreases. Nickel-based alloys and Ti alloys are highly corrosion resistant to alkalies, but their specific amount of corrosion cannot be estimated.

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corrosion potential and chloride ion concentration (50° C)

Fig. 2 Crevice corrosion occurrence threshold potential for Ti and various types of stainless steel in a saturated aqueous sodium chloride solution environment

(2) In the case of ceramics

Alumina (AL203) · · Corrosion resistant in acid solution but has poor corrosion resistance in an alkaline solution.

Higher the extent of purity better is the corrosion resistance

Cordierite (2MgO·2Al2O3·5SiO2) ·· Has poorer corrosion resistance than alumina, but has been reported to have stronger corrosion resistance to alkaliies.

Zeolite (SiO4 and AIO4 basically with a tetrahedral structure) · · There are no reports on corrosion resistance. * Yoshio et al, Materials and Environment, 44, 405 - 415 (1995)

Proceedings of the Japan Concrete Institute, Vol.30, No.1 (2008)



Fig. 3 Correlation between salinity threshold and pH

Table 1 Corrosion resistance of ceramics with respect to acids and bases 7



Fig. 4 Corrosion resistance of ceramics with respect to acids and bases *



■ Investigation of the impact that each degradation factor has on the filter

#### **O Wall thinning**

(3) In the case of glass fiber

E (for general use), C (acid resistant), S (high strength), D (low dielectric constant) are the types of glass fiber filters, but only C is manufactured considering its corrosion resistance.

Glass fiber is not very alkali resistant. This is believed to be because the Si-O bonds are broken.

Moreover, there are no reports on corrosion in neutral diluted sea-water, under water, etc.

#### (4) Polymer

Polyethylene, polypropylene, PTFE indicate good corrosion resistance to saline water and alkali solution.

Type of plastic material	Vinyl chloride resin		Vinylidene chloride	Polyvinyl alcohol	Polyvinyl acetate	Polystyrene	ABS	Polyethylene	Polypropylane	olyisobutylene	Polyamide (Nylon)	Polyacetal (Delrin / Duracon)	Methacryl (Acryl)	Polycarbonate	Fluorine (Tefion)	Cellulose acetate
	Soft	Hard														
Oil, solvent, chemicals (according to the order of Japanese syllabary) [mass concentration % and temperature * C]	PVC	PVC	PVdC	PVA	PVAc	PS	ABS	PE	PP	PIB	PA	PCM	PMMA	PC	PTFE	CA
Potassium chloride	0	0	0	0	0	٥	٥	0	0	0	0	0	0	0	0	0
Calcium chloride	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Marcurio chloride	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
Stannic chloride	0	0	0	0			0	0	0	x	0	0	0	0	0	
Ferric chloride	0	0	0	٥	0	٥	٥	0	٥	0	0	0	0	0	0	0
Copper chloride	0	0	0	0	0	٥	٥	0	٥	٥	0	0	0	0	0	0
Thionyl chloride	х	х					х	x	х	х	х				0	
Nickel chloride	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Barium chloride	0	0	0	0	٥	0	0	0	0	0	0	0	0	0	0	0
Benzyl chloride	х	Δ					х			x	х				0	
Magnesium chloride	0	0	0	0	0	٥	٥	0	0	٥	0	0	0	0	0	0
Methyl chloride	х	х	х	х	х	х	х	x	Δ	x	х	х	х	х	0	х
Hydrochloric acid [10·RT]	0	0	0	0			0	0	0	0	0		0	0	0	
Hydrochloric acid [20-RT]	0	0	0	0			0	0	0	0	Δ		0	0	0	
Hydrochloric acid [20·SO]	x	Δ	Δ	Δ	х	x	Δ	Δ	0	Δ	х	х	Δ	Δ	0	x
Hydrochloric acid [38-RT]	Δ	0	0	0			Δ	0	٥	٥	х		0	Δ	0	
Salt water	0	0	0	0	0	٥	٥	0	0	0	0	0	0	0	0	0
Chloride gas (dry)	Δ	0	0			0	х	Δ	Δ	x	х	х	0	х	0	
Chloride gas (wet)	Δ	0	0				х	Δ	Δ	x	х	х	Δ	х	0	
Chlorinated solvents	x	х	x	х	х	x	х	x	х	x	х	х	х	х	00	x
Aqua regia	х	Δ	х	х	х	х	х	x	Δ	x	х	х	х	х	0	x
Octyl alcohol	х	0	0				0	0	٥	0	Δ	Δ	Δ	0	0	
Ozone	0	0	0				Δ	Δ		0	х		Δ		0	
Olive oil	Δ	0	0				٥	0	0	x	0		0	0	0	
Oleic acid	Δ	0	0	0			0	Δ	0	x	0	0	х	Ox	0	
Perchloric acid	0	0	0					0	Δ	0				0	0	
Hydrogen peroxide [5·RT]	0	0	0				Δ	0	٥	0				0	0	
Hydrogen peroxide [5·50]	0	0	0				Δ	0	0	Δ				0	0	
Hydrogen peroxide [30 · RT]	0	0	0				х	0	0	0				Δ	0	
Sodium peroxide	0	0	0					0	0	0	х		х	х	0	
Sodium peroxide [10·RT]	0	0	0	0	0	٥	٥	0	0	٥	0	0	0	Δ	0	
Sodium peroxide [30·RT]	0	0	0	0	0		0	0	0	0	0		Δ	Δ	0	
Sodium peroxide [30-70]	x	Δ	Δ	Δ	Δ		OΔ	0	0	0	х		Δ	x	0	



Fig. 1 Alkali resistance of glass fiber

Kasuga, Journal of the Japan Research Association for Textile end-uses, Vol. 6, No 4 (1965)

Table 1 Corrosion resistance of polyethylene, polypropylene, PTFE is good and is indicated by ©

Excerpts from Kayo Corporation http://www.kayo-corp.co.jp/


# Estimation of failure mode

■ Investigation of the impact that each degradation factor has on the filter

#### **O Salt deposition**

• An event wherein water contained in waste gushes towards the outside of the container, it adheres to the filter and after drying salts get deposited on the filter thereby blocking it, due to which its filtration functionality is lost.

 $\Rightarrow$  Water containing salts coming in contact with the filter is a necessary condition

(Dispersibility of droplets of water containing salts)

- Droplets get dispersed due to hydrogen generation and due to seismic motion.
- ① Dispersion due to hydrogen generation
- Hydrogen bubbles in the water contained in waste rise and reach the water surface. When the bubbles burst, micro droplets are formed.
- ·Micro droplets are generally called film drop, and their size is about 5-10µm^[2]
- As the density of water is 1g/cm³, based on the results of evaluating the floating of fine particles due to hydrogen generation, even if micro droplets are formed they do not reach the filter.

(2) Dispersion due to seismic motion

- Water containing salts is highly likely to come in contact with the filter due to vibrations caused by seismic motion.
- The size of salt particles that are formed when water containing salts dries is assumed to be 200-800µm^[3]
- HEPA filter is mainly made up of around 1µm glass fibers, and its porosity is about  $90\%^{[4]}$ . Hence the size of the pores is believed to be roughly about 1µm / (1-0.9) =10µm

# $\Rightarrow$ Water containing salts is likely to come in contact with the filter due to seismic motion, and the salt formed when this water dries is likely to

## block the filter.

[1]: The conceptual diagram has been drawn assuming the storage container based on information related to secondary waste produced during contaminated water treatment

[2] Satoshi Kadowaki, "Sea-salt Particles in the Atmosphere", Aerosol research, Volume 6, Issue 2 (1991)

- [3] Akira Yokochi, "Recent Tendency of High Efficiency Particulate Air (HEPA) filters", Health Physics, 19, 249258 (1984)
- [4] Li Bao "Influence of Fiber Size Distribution on the Collection. Performance of HEPA Filter", Aerosol research, 12(1), 38 44 (1997)



Fig. 2 Grain size of the crystals formed when saturated salt water vaporizes ^[3]



 $\begin{array}{c} \overline{d_f} = 0.96 \ \mu m \\ \alpha = 0.09 \\ \sigma = 0.72 \\ \delta_F = 0.94 \\ H = 0.31 \ mm \end{array}$ 

Fig. 3 Example of the structure of HEPA filter^[4]

Investigation of the impact that each degradation factor has on the filter
 O Electric corrosion

The filter gets wet due to condensation or due to the water in the waste, and chloride ions adhere to it as a result of which electric corrosion occurs. The formed corrosion products block the filter.

• Following are hypothetical conditions

The storage container is made of carbon steel

Structure of filter is according to Figure 1

The housing of the filter is made of stainless steel or nickel-based alloys

Since ceramics, glass and polymers are insulators, electric corrosion does not occur.
Hence only metal filters were evaluated

Hence only metal filters were evaluated.



#### Table 1 Material of the contact area and occurrence of electric corrosion

Storage container material	Filter housing	Filter material	Occurrence of electric corrosion of the filter
Carbon steel		SUS316L	Absent
	Made of SUS	Nickel-based alloy	Absent
		Ti alloy	Absent



Filter surface

Fig. 1 Example of the structure of the filter

Threaded portion

Filter

Filter housing

Filter lid

Taper threaded portion

Retainer ring

Fig. 2 Potential difference in metals under sea water (temperature 25°C)* * Urban Steel Research Group "Steel frame landscape material guide" July 1997



# Estimation of failure mode

- Investigation of the impact that each degradation factor has on the filter
- O Water film

# Formation of water film

• A water film is likely to be formed on the filter surface due to condensation. Let us assume that the temperature inside the container falls from 30° C to 10° C.

Assuming that the amount of saturated water vapor: 30.4g/m³@30° C, 9.4g/m³@10° C, capacity of the waste container is approx. 1.5m³, area of the lid is approx. 1.69m², the amount of condensed water is 31.5g. If the condensed water adheres only to the surface of the lid, the amount of water adhered to the surface of the filter is 31.5/1.69=18.6g/m².

If the specific gravity of water is assumed to be 1g/cm³, the water film will be 18.6µm which is extremely thin, but if it piles up, the filter element will get blocked by water.

• As the formation of water film due to condensation does not depend on the material of the filter, this will be discussed for all types of filter material.

# If the filter gets blocked due to moisture

• Condensation can be reduced by appropriately controlling the temperature and humidity, but blockage for a short time cannot be denied.

In that case, whether or not the increase in internal pressure due to hydrogen generation can push out the moisture from inside the filter element becomes the performance indicator.

• After the filter is blocked due to condensation when the pressure inside the container is 1 atm and hydrogen is 0vol%, when the hydrogen concentration becomes 4 vol% due to hydrogen generation, the overall pressure becomes approx. 1.04 atm. Hence, it is believed that a filter that has gotten wet can be used as long as the pressure is lower than this and air starts flowing. Meanwhile, if air cannot flow, hydrogen is likely to get accumulated inside the container.

# Summary

When the filter element is covered with a water film due to condensation, whether or not ventilation can be brought about before the hydrogen concentration reaches the lower concentration limit for combustion, needs to be verified by means of a test.



- Investigation of the impact that each degradation factor has on the filter
- Freezing

## Occurrence of freezing

- When a water film is formed on the surface of the filter due to condensation and the air conditioning equipment fails, the filter gets exposed to below freezing conditions resulting in freezing. If the entire filter is covered with ice, the filter element gets blocked. Also, in the case of organic material, the mechanical characteristics are likely to change due to exposure to low temperature.
- As blockage due to freezing does not depend on the material of the filter, this will be discussed for all types of filter material.

## If the filter gets frozen

 Under appropriately controlled temperature and humidity, condensation is very unlikely, and as long as the temperature is 0° C or more freezing does not occur.

However, if the filter is completely blocked due to freezing, hydrogen is likely to get accumulated.

- In that case, it can be evaluated whether or not the hydrogen concentration will become 4% based on the hydrogen generation rate and the time for which freezing continues.
- If organic material will be used, it is necessary to investigate its mechanical strength at low temperatures.

#### Summary

It needs to be verified by means of tests whether or not the prescribed mechanical strength is met if the filter element gets frozen.

Whether or not the ice melts and ventilation can be brought about before the hydrogen concentration reaches the lower concentration limit for combustion, does not need to be verified through tests, but can be evaluated based on the hydrogen generation rate and the time for which freezing continues.



■ Investigation of the impact that each degradation factor has on the filter

# **O Dispersion of fine particles**

• Due to the impact of the ventilation system of the storage facility, a current gets generated inside the storage container. As fine particles scatter and adhere to the surface of the filter, the filter gets blocked and the filtration functionality is lost.

(Conditions in which the impact becomes apparent)

• When fine particles disperse from the surface of waste due to the current generated as a result of ventilation, there is an impact.

• As the impact of ventilation does not depend on the filter material, this will be discussed for all types of filter material.



Fig. 1 Dispersion of fine particles from the solid surface Conceptual diagram^[1]

(Ventilation conditions)

- Passages through which gas streams flow are not present inside the storage container. The differential pressure inside and outside the container gives rise to gas streams inside the container.
- However, the ventilation system inside the building is supposed to perform air intake as well as exhaust, and hence there is no difference in the pressure inside and outside the container.
  - $\Rightarrow$  It is believed that the flow rate conditions that would lead to dispersion of particles would not arise.

## Excluded from the failure mode assuming that the ventilation system does not lead to dispersion of fine particles.

[1] Kuniaki Goto, "A Study on Particle Dispersion Technique and Powder Adhesiveness.", Pulverization No.59 (2016), p51-59



- Investigation of the impact that each degradation factor has on the filter
- Summary
  - The impact of each degradation event on filter degradation (blockage, damage) was evaluated.
  - The following 8 events were selected as degradation events that are likely to have an impact on degradation of the filter during the process of collecting solid waste.

Note that, the process in which the impact becomes apparent is mentioned in [] and the filter material that is affected is mentioned in ().

<ul> <li>Hydrogen embrittlement</li> </ul>	[During storage] (Polymer)
<ul> <li>Radiation deterioration</li> </ul>	[During storage] (Polymer)
Impact	[During collection / during transportation / during storage] (All materials)
<ul> <li>Dust formation due to vibrations</li> </ul>	[During storage] (All materials)
Wall thinning	[During storage] (All materials)
Salt deposition	[During storage] (All materials)
Water film	[During storage] (All materials)
Freezing	[During storage] (All materials)

• Hereinafter, degradation simulation and performance assessment methods will be investigated for the 8 events that have an impact using the performance test method.



# (a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters Study of performance testing methods

- Methods of testing filter materials
- O Hydrogen embrittlement test
- The amount of hydrogen generation is calculated based on the duration of storage of solid waste and the rate of hydrogen generation from waste. Based on that, the maximum, intermediate and minimum amount of hydrogen generation are selected, and each type of filter material is charged with hydrogen. The filter material charged with hydrogen is mounted on the filter performance evaluation equipment and the test is carried out. Hydrogen can be charged either by means of the electrolytic method or the high pressure charging method. The method that is suitable for the material is used.
- The charged filter is checked for the amount of hydrogen charged. This is carried out using the thermal desorption method. (Refer to Fig. 1)



Fig. 1 Example of thermal desorption analyzer



(a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters Study of performance testing methods

- Methods of testing filter materials
- Radiation deterioration test
- The material to be tested is exposed to radiation with the expected radiation dose. The mechanical characteristics before and after irradiation are investigated.

It is investigated whether or not the above-mentioned characteristics satisfy the stress expected when the material is used as a filter.

Exposure dose

It is difficult to estimate the exposure dose of the filter as it depends on the amount of radioactivity present in the contents of the container, the distance, moisture content, etc. Hence it is tentatively set at 30mSv/h as shown in Fig. 1 [1]. The cumulative dose over a period of 50 years is

30 x 10⁻³ (Sv/h) x 24 (h) x 365 (day) x 50 (year) = 1.3 x 10⁴(Sv)

It is believed that the material should be irradiated up to approx. 130000 Sv to evaluate the correlation between the extent of deterioration and the radiation dose.

Testing the effect

The effect of radiation on the mechanical strength can be evaluated by simulating deterioration during use by exposing the filter to radiation beforehand, and then subjecting it to an impact test.



## Fig.1 Example of waste container dose rate [1]

[1] Set using the container surface dose rate mentioned in "Design for slurry stabilization treatment" Special Nuclear Facility Monitoring and Evaluation Committee (88th Session) Document 1-2-1, 2021/1/25, as reference.



# (a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters Study of performance testing methods

Methods of testing filter materials

# Impact test

The filter is subjected to impacts expected to arise during the work undertaken as part of the collection, transfer and storage flow. The integrity of the filter after being subjected to impacts is checked.

<Testing the effect>

- The filter is fixed on to the lid part of the structure simulating the storage container (hereinafter, simulated storage container).
- The simulated storage container is lifted up to the designated height by means of the hoisting accessory.
- The simulated storage container is detached from the hoisting accessory and allowed to fall freely.
- The integrity of the filter is checked after the fall (External appearance, filter performance test, etc.).



Fig. 1 Example of the direction of the fall of the simulated storage container while testing the effect

Note that the results are likely to differ depending on the method of attaching the filter to the lid part, the shape and dimensions of the storage container, the method of attaching the lid part to the storage container, the shape of the inner container, etc., and hence it is desirable to conduct the test after the detailed specifications are finalized.



(a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters
 Study of performance testing methods

Methods of testing filter materials

# **O Test on dust formation due to vibrations (Test on dust formation due to seismic ground motion)**

A vibration test is conducted in which random vibrations simulating actual seismic ground motion are imparted, and fine particles that get airborne as a result are made to adhere to the filter to simulate filter deterioration.



• After imparting vibrations, the amount of matter adhered and the adhesion status is checked based on the weight of the filter and by means of a spectral analysis, and the performance is evaluated separately by means of a performance assessment test.

# Note that the results are likely to differ depending on the method of attaching the filter to the lid part, the shape and dimensions of the storage container, the method of attaching the lid part to the storage container, the shape of the inner container, etc., and hence it is desirable to conduct the test after the detailed specifications are finalized.



(a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters [Study of performance testing methods]

- Methods of testing filter materials
- O Methods of testing wall thinning
- Cyclic corrosion test (JIS K 5600_7_9): A method used for evaluating the paint by means of a corrosion test in which the cycle of salt spray, drying and humidification is repeated. This method is used to evaluate wall thinning and blockage of various types of filters.
- Using the above-mentioned testing method, various filters are exposed to corrosive environment to develop wall thinning and blockage conditions. The filters in which the extent of deterioration changes are placed on the filter performance assessment equipment and tested. Thus the extent of deterioration under which performance can be maintained becomes clear.
- Meanwhile, corrosion tests are conducted on various types of filters by simulating expected environmental conditions, and the occurrence of deterioration is studied through relative comparison with the extent of deterioration under corrosive environment, and in the expected environment.



Fig. 1 Example of the cyclic corrosion test apparatus

The filter is placed inside the equipment illustrated in Fig. 1, combined cycles are conducted and the filter is subject to corrosion.

The tower in the center serves as the spray tower for spraying water, saline water, etc. A humidifier is installed at the bottom, and water in the form of mist blows in through the steam pipe. The temperature is adjusted by means of direct steam heating.



# (a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters Study of performance testing methods

Methods of testing filter materials

# O Simulation test on deterioration due to salt deposition

Deterioration of filters due to salt deposition is simulated by impregnating and drying the filter with saline water.

# (Testing method)

- A simulated aqueous saline solution is prepared (Type of salt and its concentration is simulated).
- The ratio of the solution to the waste (simulated) is adjusted, and the simulated waste is prepared in the container.
- The filter is installed on top of the container.
- (The height is adjusted according to the distance between the waste and filter)
- The lid is closed and a vibration test is conducted by simulating seismic ground motion.
- After imparting vibrations the filter is removed. After drying it, the amount of salt adhered to it and the form of adhesion are evaluated.
- The performance is evaluated by means of a separate performance assessment test.

* Note that the results are likely to differ depending on type of filter, amount of waste filled in the container, moisture content, type and concentration of salt in the solution, its pH, dimensions and shape of the container, etc., and hence it is desirable to conduct the test after the detailed specifications are finalized.



Fig. 1 Schematic diagram of the simulation test on salt deposition



(a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters [Study of performance testing methods]

- Methods of testing filter materials
- O Water film test

Air permeability test of filter that is blocked due to droplets

- If the filter is not completely blocked
  - Frazier Air Permeability Test (JIS L 1096, JIS L 1913)^[1]

The difference in pressure inside and outside the test piece is kept constant, aeration is brought about using a fan and the amount of air (cm³/cm²·s) is measured.

If partial blockage is considered equal to reduction in the area of the filter membrane, the margin in the filter area can be evaluated through this test.

Vertical barometer^{*1}

Test piece

- If the filter is completely blocked
  - Proposed air permeability test of a blocked filter^[2]

The method of testing water resistance of fibers is used for

evaluating the pressure at which water is discharged from

the filter.

The filter is attached, pressure is applied using water and the pressure when water is discharged from the filter is measured.

The pressure required for discharging gas when condensed water is formed can be evaluated with this test.

*1 Indicates the differential pressure between the atmospheric pressure above the test piece and ①.
*2 Indicates differential pressure between ① and ②.



Support plate

Scient

Vertical barometer*2

Hydraulic pressure Fig. 2 Conceptual diagram of the air permeability test of a blocked filter

Sample

(Created based on ^[2])

[1] "Air permeability test of textiles" Aichi Center for Industry and Science Technology News December 2018

(http://www.aichi-inst.jp/other/up_docs/no.201_03.pdf)

[2] Document from W. L. GORE and Associates (https://www.gore.co.jp/sites/g/files/ypyipe116/files/2016-04/PTV-TechInfo-WEP-Testing-US.pdf)



(a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters [Study of performance testing methods]

Method of testing the effect of freezing of filter materials

# **O** Testing the effect of freezing

• The material to be tested is cooled and frozen while it contains water. The mechanical characteristics when the material is frozen and after it is thawed are investigated.

It is investigated whether or not the above-mentioned characteristics can handle the stress expected when the material is used as a filter.

• Freezing conditions

The cooling temperature and cooling period are determined depending on the use conditions of the filter.

# • Testing the effect

The effect on mechanical strength of the filter material can be evaluated by simulating deterioration of the filter material due to freezing and then subjecting it to an impact test.

# (a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters Study of integrity evaluation methods

- Methods of evaluating the particle collection function
  - There are mainly 2 types of air filter testing methods namely the weighing method and the number counting method. The number counting method is used for HEPA and ULPA.
  - In the number counting method, the minimum collection rate (reference number) and the pressure loss when an air current containing aerosol is passed at a rated flow specific to the filter, are evaluated.
- Testing method^[1]
  - The testing environment is prepared with temperature 25±10°C, humidity 55±15°C, flow rate from the fan motor 0.45m/s±20% at the filter portion
  - Aerosol is fed for testing purposes on the upstream side of the filter (1 type (DOP) of the 2 particles for test purposes or an equivalent aerosol is used^[2])
  - The concentration of the aerosol for test purposes is adjusted (The particle measurement instrument on the upstream side indicating 0.3µm or more number of particles at 5 × 10⁶ particles/min or more).
  - The particle measurement instrument^[3] on the downstream side is adjusted (Measured in clean air and let to stand until the reading becomes stable).
  - Aerosol for test purposes is fed to the filter, the numbers of particles on the upstream and downstream side are continuously measured at fixed time intervals, and the collection rate is calculated.
  - The static differential pressure on the upstream and downstream side of the filter is measured using a manometer and the pressure loss is calculated.
- Determination conditions^[4]
  - Minimum collection rate (Reference number): 99.97% or more for particle size 0.3µm in the case of HEPA, and 99.9995% or more for particle size 0.15µm in the case of ULPA
  - Pressure loss: Initial pressure loss 245Pa or less for both HEPA and ULPA



Fig. 1 Performance assessment test Equipment configuration (Example) [1]

For HEPA and ULPA, if the filter performance after the deterioration test satisfies the determination conditions, deterioration effect is considered to be absent.

[1] JIS K 3801 Test Method of HEPA Filters for Microbiological Use

- [2] JIS Z 8901 Test Powders and Test Particles
- [3] JIS B 9921 Light Scattering Airborne Particle Counter
- [4] JIS Z 8122 2000 Contamination Control Terminology



# (a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters <u>Study of integrity evaluation methods</u>

- Method of evaluating the hydrogen permeation function
- The method for evaluating the hydrogen permeation function of filters used in nuclear facilities in the US is described in References [1]. Hydrogen of designated concentration (4%) is sealed inside the container with attached filter and it is let to stand. The hydrogen concentration inside the container is measured 7 times every 5 minutes. The rate of hydrogen permeation (hydrogen diffusivity D) is obtained using the formula listed below.

$$D = \frac{PV}{tRT} Ln\left(\frac{H_0}{H_t}\right)$$

D: Hydrogen permeation rate (Mol/sec/molFraction),

- P: Pressure (atm), V: Container capacity (L), t: Time (seconds),
- R: Ideal gas constant (0.082Latm/molK),
- T: Temperature (K),  $H_0$ : Initial hydrogen concentration,  $H_t$ : Hydrogen concentration at time t
- If being installed inside the container, the required number of designated filters are installed depending on the hydrogen generation rate inside the container.



Fig. 1 Hydrogen permeation function evaluation test Equipment configuration (Example)

[1] Terry Wickland and John Schierloh "HYDROGEN DIFFUSIVITY THROUGH DRUM AND LINER VENTING FILTERS" WM98 (http://archive.wmsym.org/1998/html/sess27/27-37/27-37.htm)



# (a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters

- Results of implementation
  - O Organizing of input conditions
    - ✓ Target waste was selected from the Waste List and the waste characteristics.
    - Storage conditions and storage environment were organized.
  - O Study of deterioration factors
    - ✓ Deterioration modes were defined as blockage and damage of the filter.
    - The causes and events leading to blockage of filters were organized based on the storage environment of solid waste, and 18 deterioration events were identified in all.
  - O Clarification of filter requirements
    - Besides filters used in overseas nuclear facilities, ① manufacturing methods, filter material, ② application, main manufacturers, ③ fine particles collection function, ④ hydrogen permeation function of the filters for general industrial use (metal, ceramic, glass fiber, polymer) were investigated and the results were organized.
    - Filters for general industrial use that satisfy the fine particle performance specifics (equivalent to HEPA) of overseas vent filters can be manufactured. Although the actual value pertaining to the hydrogen permeation function has not been specified in the catalog, etc., in principle, it is presumed to be equivalent to overseas vent filters.
  - O Estimation of failure mode
    - 11 deterioration factors that are likely to have an impact on filter deterioration were identified from 18 deterioration factors based on input conditions.
    - ✓ Failure mode scenarios were developed for each process for the 11 deterioration factors.
    - Occurrence of deterioration, extent of progression of deterioration when each type of filter is used were estimated through literature survey, and 8 affecting events were identified.
  - O Study of performance testing method
    - The performance testing methods required for evaluating the impact of the 8 failure modes on the life of the filter were studied and organized.
  - O Study of integrity evaluation method
    - The filter performance testing methods for evaluating the impact of each failure mode on the integrity of the filter were studied.



(a) Study and evaluation of storage and management methods [2] Study of the requirements for containers and storage facilities : Performance evaluation of vent filters

# Summary

# **Results so far**

- Characteristics that are common to each type of waste were identified based on the organization of the types of solid waste, and in addition, based on the expected treatment processes, the environments in which the filters will be placed were assumed as input conditions and organized.
- ✓ The function of letting hydrogen permeate and the function of collecting fine particles were defined as functions required of a filter, and environmental deterioration factors that are likely to affect these functions were identified. Also, general filter specifications were investigated, and prospects of being able to select filter material depending on environmental requirements were seen.
- Scenarios likely to impair filter performance were studied based on filter functions and solid waste treatment processes, respective failure mode scenarios were investigated and organized, and in addition, proposed testing methods for evaluating the impact that the failure modes have on filter life were studied.

# Issues, etc.

As types of solid waste, treatment methods, storage requirements, environmental criteria, facility constraints, etc. serve as input information for determining filter specifications, items pertaining to which conditions become concrete as production level work progresses need to be compared with the input conditions assumed in this research, and the necessity to reselect the deterioration simulation test conditions and failure modes needs to be studied once again.



(b) Technological development of contamination evaluation for sorting solid waste

- Achievements up to FY2020
- α camera was manufactured and a function verification test of the system was conducted.
- A mock-up test of the α contaminated equipment (flange tank piece) was conducted at the site to assess performance.

It was possible to measure the 220Bq, 740Bq, 2960Bq  $\alpha$  ray sources (Am-241) affixed on the flange tank piece, but the noise generated was from equivalent or higher concentrations.

# Goal

- The factors due to which the flange tank piece generated noise will be specified and noise countermeasures will be taken.
- O Upon undertaking countermeasures  $\alpha$  contamination detection performance will be evaluated once again.
- Details of implementation (See details on next pages)
- Issues in the items that affect measurement will be consolidated based on site investigation.
- The following will be implemented in response to the consolidated issues.
   α camera: Noise countermeasures will be taken for the flange tank piece.

Distance measurement system: The range within which the TOF conditions are satisfied will be indicated based on the results of site investigation.

Means for access: A conceptual drawing of the crawler on which the  $\alpha$  camera is mounted will be provided. (The scope of this Government R&D project is up to consolidation of issues)

- Indicators for the achievements of goal
- Presentation of issues based on the mock-up test results for using the α camera at the site, and corresponding countermeasures.

(Target at the completion: TRL:  $3 \rightarrow 4$ )

Fig. 1 FY2020 mock-up test results



(b) Technological development of contamination evaluation for sorting solid waste Current status of  $\alpha$ -contaminated waste management and issues thereof

# Current status of α-contaminated waste management

When α-nuclide contamination with surface contamination density of 0.4 Bq/cm² or more is detected by means of smear measurement, the waste is sorted and placed in storage containers. Adopting smear measurement for ascertaining the status of contamination in the future when full-fledged decommissioning work will be carried out, is not practical considering the time required for measurement and from the viewpoint of radiation dose reduction.

# • Requirements related to the technology for sorting waste

- > Being able to ascertain surface contamination distribution of target objects from a distance.
- > Being able to ascertain contamination distribution of targets over a wide area in a short time.

# • Purpose of this project

To develop a technology that can **comprehensively and in a short time** measure the surface of the object (or area) to be dismantled. Also, to clearly specify the applicable process depending on performance.



# IRID

(b) Technological development of contamination evaluation for sorting solid waste Principle of an  $\alpha$  camera and its advantages

# Measurement principle

- (1) Reaction takes place with nitrogen within  $\alpha$  radiation range (about several cm) to produce several 100 photonic ultraviolet rays.
- 2 These ultraviolet rays are focused and imaged with a lens, and the distribution of  $\alpha$  rays is measured from the distribution of the number of photons reaching the light detector.
- ③ PMT (Photomultiplier Tube) is shielded from γ rays as it is likely to be affected by Cherenkov light or others when γ rays enter PMT.



Can be measured at a distance from the source of α contamination
A wide area can be measured at once.

#### Examples of main presentations made in connection with $\boldsymbol{\alpha}$ camera

- 1. IEEE2013 NSS、Remote Detector of Alpha-Ray Using Ultraviolet Ray Emitted by Nitrogen in Air
- 2. ICONE-23 REMOTE DETECTION OF ALPHA RADIATION USING UV PHOTONS EMITTED BY NITROGEN
- 3. Spring meeting of Atomic Energy Society of Japan in FY2013, Technology for remote measurement of  $\alpha$  radiation
- 4. Autumn meeting of Atomic Energy Society of Japan in FY2014, Technology for remote measurement of α radiation Application to lighting environment -



Fig. 1 Schematic diagram illustrating the principle



Cited from J.Sand. Remote Optical Detection of Alfa Radiation. IAEA-CN-184/23.



# (b) Technological development of contamination evaluation for sorting solid waste Development history



*Peak other than the radiation source location



(b) Technological development of contamination evaluation for sorting solid waste This year's report

The following was implemented in response to the unanticipated noise generation found as a result of the FY2020 1F mock-up test.

(1) Common items

- Obstructive factors concerning the α camera, distance measurement system, and means for access were identified based on the site photographs.
- Issues in each element were consolidated based on the obstructive factors.
- (2) Measures in response to issues with the  $\alpha$  camera
- Method of implementing measures in response to the site investigation results
- The α rays, β rays emission processes were organized quantitatively (This year the focus was on nitrogen light emission)

A prediction model was developed based on emission tests and simulations, and was evaluated.

• Countermeasures were comprehensively studied based on the characteristics of noise emission, and their effectiveness was evaluated.

(3) Measures in response to issues with the distance measurement system

- Clear specification of the range in which distance measurement is feasible
- Improvement in the distance combination algorithm

(4) Means of access

- Conceptual drawing assuming that an existing decontamination crawler is used
- Issues until application to actual equipment

# (b) Technological development of contamination evaluation for sorting solid waste Consolidation of issues Looking back at the FY2020 mock-up test (1) Details of the test location -

The test was conducted at the building for large component inspection where the flange tank piece decontamination work was being carried out.

 $\beta$   $\gamma$  contamination was spread over a wide area. Hence a flange tank piece with as high a dose rate as possible was selected from that area for measurement.

- Measurement target: 1 flange tank piece meeting the test conditions was selected from flange tank pieces prior to decontamination work.
- Measurement location: Contaminated area (Y zone). Contamination was mainly Sr/Y-90 nuclides emitting β rays.





External appearance of large component decontamination equipment



Fig. 1 External appearance of the location of the large components inspection building Cited from TEPCO HD "Status of installation of large components decontamination equipment April 16, 2018" Fig. 2 Flange tank piece Photographed on October 30, 2020



# (b) Technological development of contamination evaluation for sorting solid waste Consolidation of issues Looking back at the FY2020 mock-up test (2)

Test details	Test method	Results
Evaluation of correlation between $\beta \gamma$ surface contamination density and noise (Test ①)	$\beta$ $\gamma$ surface contamination density (2 - 5k Bq/cm ² ) at different sites was measured by changing the tilt angle to 10°, 40°, 60°, 90°, 120°, to evaluate the correlation between $\beta$ $\gamma$ surface contamination density and amount of noise.	Correlation between $\beta \gamma$ surface contamination density and noise was obtained. The amount of noise increased 20 times than the functional test.
Evaluation of correlation of amount of signals with respect to sources of $\alpha$ rays (Test 2)	Standard sources of $\alpha$ rays of varied strengths (Am-241, 220, 740, 2960Bq) were arranged in the angle of view when the tilt angle was 90° to obtain the correlation between $\alpha$ surface contamination density and the amount of signals ^{*1} .	The sensitivity was $0.31 \pm 0.16[s^{-1}/Bq/cm^2]$ in the performance verification test and it was $0.47[s^{-1}/Bq/cm^2]$ in the laboratory test. Thus it was <b>consistent with a margin of error.</b>
Evaluation of dependency on the measured distance (Test ③)	Standard source of $\alpha$ rays (Am-241, 2960Bq) was set up at locations with varied measured distance to evaluate the dependency of $\alpha$ rays sensitivity on the distance.	It was found that just as in the functional verification test, the sensitivity was inversely proportional to the square of the distance. However, real-time correction was difficult due to the impact of noise.
Evaluation of the accuracy in ascertaining contamination distribution (Test ④)	Based on data from multiple angles $(10^\circ, 40^\circ, 60^\circ, 90^\circ, 120^\circ)$ three dimensional contamination distribution was developed from the difference with the measurement results obtained when there was no radiation source, to evaluate the accuracy of identifying locations with $\alpha$ surface contamination	The accuracy worsened than the position resolution when the test was conducted using standard radiation source due to the impact of noise, but it was possible to identify the location of radiation source at a position resolution of approx. 6 cm which is lower than the target 10 cm.

# Table 1 Test items by evaluation items

*1 Due to the impact of noise, the difference between the value of pixels with radiation source and the value of pixels without radiation source was calculated as the signal originating from the radiation source.





Fig. 3 Distribution after subtracting noise from the measurement result (Test ④)



(b) Technological development of contamination evaluation for sorting solid waste Identification of issues: Investigation of site conditions



# Table 1 Issues and factors identified in Unit 3

Items	Identified issues and factors		
Measurement	Wall	Paint	
of α rays	Floor	Paint (water accumulated)	
	Cables	Black	
	Sheet	None	
	Light shielding property	No light emitting substance	
Distance measurement	No impact		
Accessibility	Unevenness of floor surface		



# Table 2 Issues and factors identified in Unit 2

Items	Identified issues and factors		
Measurement of	Wall	Paint	
α rays	Floor	Paint	
	Cables	None	
	Sheet	Pink covering sheet	
	Light shielding property	External light present	
Distance measurement	Impact of equipment likely		
Accessibility	Unevenness of floor surface, dispersion of equipment and material		



# (b) Technological development of contamination evaluation for sorting solid waste Mapping of site investigation results





(b) Technological development of contamination evaluation for sorting solid waste Consolidation of issues: Items that are likely to have an impact on measurement

Items that are likely to have an impact on measurement are identified from the perspective of measuring the contamination distribution of the building before dismantling.

Target system		Items	Event	Site conditions that must be evaluated
α camera	Emission noise due to the impact of $\beta$ rays, $\gamma$ rays	Scintillation light	Light emission of nitrogen (air)	-
			Light emission besides nitrogen	Paint, type of sheets, etc.
		Cherenkov light	Cerenkov light emission from matter having a refractive index	Paint, type of sheets, etc.
	Impact on amount of α ray signals	Material to be measured	Change in $\alpha$ ray sensitivity due to roughness of the surface of the material	Presence of rubble (More the roughness higher the sensitivity)
			Wetness of the surface	Presence of water accumulation
	Limiting conditions during measurement	Light shielding property	Impact of lighting	Presence of emergency lighting Light leakage from outside the building
Distance measurement		Impact of matter in the vicinity	Shooting angle	Direction of access
			Infrared transmission	Materials
			Infrared absorption	Materials
Access cart		Route for carrying-in	The cart cannot be carried in	Presence of steps, width of the passageway, etc.

# Table 1Items that must be evaluated during site investigation



# (b) Technological development of contamination evaluation for sorting solid waste Issue consolidation drill down: Issues pertaining to the $\alpha$ contamination measurement system



# IRID

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(b Technological development of contamination evaluation for sorting solid waste Initiatives for addressing  $\alpha$  camera issues



Fig. 1 Excerpts from the process of drilling down issues

<Matters that must be considered with respect to issues>

 Amount of light emitted by various material and the signals detected by α camera need to be closely examined
 Actions need to be taken so as to be able to deal with unanticipated light emitting sources at the site
 Measures must be taken in response to items for which measures can be taken in advance

(1), (2): Makes it possible to quantitatively ascertain each light emission and provide estimates based on simulation.
 (3): The possible countermeasures are comprehensively studied based on the characteristics of light emission.

Light emission phenomenon	Light emission source	Characteristics of light emission
Scintillation light	Nitrogen in air	<ul> <li>Light emission by nitrogen due to α rays as well as β rays has the same wavelength distribution</li> <li>The condition of spatial light emission is likely to differ depending on the difference in the range of α rays and β rays</li> <li>Duration of light emission depending on the length of the range</li> </ul>
	Other than nitrogen (Paint, covering sheet)	<ul> <li>The amount of light emitted differs depending on the material</li> <li>The wavelength distribution differs depending on the material</li> <li>The duration of light emission is in the ns order</li> </ul>
Cherenkov light	Paint Sheet Water Optical system	<ul> <li>The wavelength distribution is ∝λ⁻², which is different from the light emission by nitrogen due to α rays</li> <li>Amount of light emission depends on the refractive index n.</li> <li>The direction of light emission is conical at an angle of cosθ=c/nv (v: velocity in a medium) with respect to the incident direction of charged particles.</li> <li>The duration of light emission is in the ps order</li> </ul>

#### Table 1 Light emission sources and their characteristics



# (b) Technological development of contamination evaluation for sorting solid waste Course of action for FY2021



Fig. 1 FY2020 mock-up test results

Status in FY2020 and preconditions for evaluating noise

- Surface dose rate of  $\beta$  rays ( $\beta$ ) is about 20mSv/h
- The noise signal count rate is about 500cps

## Overview of the FY2021 policy

1. Quantitative ascertaining of the light emission process

1-1. Light emission test using sources of  $\beta$  rays

1-2. Development of simulations

1-3. Quantitative ascertaining of the light emission phenomenon

Implemented up to quantitative ascertaining of light emission by nitrogen due to  $\alpha$  rays and  $\beta$  rays

2. Noise countermeasures

1-1. Verification of the light emission measurement characteristics of  $\alpha$  camera and comprehensive study based on the characteristics of each light emission 1-2. Evaluation of the effectiveness of noise countermeasures

Effectiveness of various measures evaluated



(b) Technological development of contamination evaluation for sorting solid waste
 Overview of the method of quantitative organization of the light emission process
 Process of light emission by nitrogen due to α rays -



Fig. 1 Flowchart of light emission by nitrogen due to α rays



(b) Technological development of contamination evaluation for sorting solid waste Overview of the method of quantitative organization of the light emission process - Process of light emission by nitrogen due to  $\beta$  rays -



Fig. 1 Flowchart of light emission by nitrogen due to β rays



(b) Technological development of contamination evaluation for sorting solid waste Purpose of the  $\beta$  ray light emission test and means of implementation

(1) Ascertaining the amount of light emission under high β radiation dose: Straight line system



Fig. 1 Schematic diagram of the set-up for testing light emission from high dose  $\beta$  rays

Source of β rays is equivalent to 1F mock-up environment (Approx. 15mSv/h)
Only scintillation light (nitrogen light emission) when samples are not provided.
Scintillation light as well as Cherenkov light when samples are provided.

(2) Separating the scintillation light and Cherenkov light to ascertain the amount of light emission: Right-angle system



Fig. 2 Schematic diagram of the test set-up in which light emission phenomena are separated

-Considering the direction of emission of Cherenkov light, the source of  $\beta$  rays - sample -  $\alpha$  camera are placed at right angles.

 Light emission is measured in varied conditions of occurrence of Cherenkov light by using high energy as well as low energy^{*} radiation sources.
 *Radiation sources that have very few β rays that exceed critical energy, are selected.



(b) Technological development of contamination evaluation for sorting solid waste Test set-up for the  $\beta$  ray light emission test



Fig. 1 External appearance of a straight line system



Fig. 2 External appearance of a right angled system

# Table 1 List of sources of $\beta$ rays

Radiation source	Dose rate ^{⁺1}	Distance* ²	Evaluation target	External appearance
Sr-90/ Y-90	15.6 mSv/h (98mSv/h)	50cm (20cm)	Nitrogen light emission, scintillation light, Cherenkov light	
Pm-147	0.21 mSv/h	20cm	Scintillation light (excluding Cherenkov light)	βrays

*1 Dose rate at the surface where the sample is placed *2 Distance from the source of  $\beta$  rays to the sample

• Sr-90/Y-90 are measured at a distance where the dose rate is equivalent to the dose rate around the 1F flange tank piece.

• Scintillation light and Cherenkov light are measured when nitrogen light emission + sample are placed.

•As a maximum of approx. 220keV of  $\beta$  rays are produced by Pm-147, the amount of Cherenkov light emitted is so small that the amount detected by  $\alpha$  camera from the direction of occurrence can be ignored.



(b) Technological development of contamination evaluation for sorting solid waste Energy distribution of the  $\beta$  ray source


(b) Technological development of contamination evaluation for sorting solid waste Straight line system for the  $\beta$  ray light emission test Organization of light emission phenomenon



Fig. 1 Light emission phenomena in a Sr-90/Y-90 straight line system

Frame (Aluminum frame)



Fig. 2 External appearance of sample Pink covering sheet

Table 1 List of light emission phenomena in a Sr-90/Y-90 straight line system

No.	Light emission/phenomena	Memo
1	Nitrogen light emission before the sample	Nitrogen light emission in the 50cm between the radiation source and the sample
2	Nitrogen light emission after the sample	Nitrogen light emission from attenuated electrons
3	Shielding of $\textcircled{1}$ due to the sample	Permeability differs depending on the sample
4	Shielding of $\textcircled{1}$ due to the frame	Shielding effect is assumed to be the same regardless of the conditions
(5)	Light emission from the sample Scintillation light	Phenomenon to be clarified 1
6	Light emission from the sample Cherenkov light	Phenomenon to be clarified 2
1	Light emission inside the $\boldsymbol{\alpha}$ camera	Generated by the bremsstrahlung or the electrons that have reached the optical system



# (b) Technological development of contamination evaluation for sorting solid waste $\beta$ ray light emission test : Test conditions of the straight line system and affecting factors

Table 1 Sr-90/Y-90 straight line system: Affecting factors in each condition

No.	Sample	Supplementary information	① Pre N2	2 Post N2	③ Shielded by sample	Shielded by frame	<b>⑤</b> Scintillati on light	6 Cherenk ov light	Contract Contrac	<b>Legend</b> O: There is an impact ∆: Although small there is an impact
1	None (Nitrogen)		0	0	-	-	-	-	0	—: There is no impact
2	White paint	t0.3mm	Δ	0	0	0	0	0	0	Other test conditions
3	White paint	t0.5mm	Δ	0	0	0	0	0	0	•Measurement time 10 min
4	White paint	t1.0mm	Δ	0	0	0	0	0	0	•BG measurement 5min
5	Gray paint	t1.0mm	Δ	0	0	0	0	0	0	·Signal is the difference with BG
6	Black paint	t1.0.mm	Δ	0	0	0	0	0	0	•Room temperature 21.3°C to 21.9°C
7	Pink covering sheet		0	0	Δ	0	0	0	0	1011.2hPa
8	Transparent covering sheet		0	0	Δ	0	0	0	0	•Humidity 26.7% to 41.7%
9	α camera light shielding		-	-	-	-	-	-	0	Processing of detection results
10	Frame only		0	0	-	0	-	-	0	<ul> <li>Measurement is started after</li> </ul>
11	Frame + shielding		Δ	0	0	0	-	-	0	sample to be measured is set up.
12	Transparent covering sheet + shielding		Δ	0	0	0	-	-	0	After completing measurement, the radiation source is removed and BG is measured

1.3	2.3	2.7	3.1	3.2	2.7	2.1	1.0
1.8	3.5	4.9	5.5	5.1	4.3	3.2	1.5
2.4	5.2	7.4	9.9	9.2	6.2	4.2	2.1
2.6	5.9	9.5	37.9	32.1	8.2	4.6	2.2
2.6	5.8	9.7	37.2	32.1	8.3	4.6	2.2
2.1	4.8	6.9	8.8	8.7	6.3	4.1	2.1
1.8	3.2	4.3	4.9	4.5	4.0	2.9	1.6
1.3	2.2	2.7	2.9	2.9	2.6	2.1	1.1

#### Analysis of test results

(1) Sum total of the data in the 2 × 2 cells in the center  $\rightarrow$  Size of view equal to that of the sample

② Sum total of the data in the 4×4 cells in the center

 $\rightarrow$  Light from outside the frame is considered as well

3 Sum total of the data in all the 8 × 8 cells in the center

 $\rightarrow$  Entire light emission is considered

Fig. 1 Example of test results (No. 7) Unit: cps



The difference between BG and

the sample measurement for each ch is obtained and that value is

used as the evaluated test result.

(b) Technological development of contamination evaluation for sorting solid waste  $\beta$  ray light emission test: Straight line system test results

Unit: cps

0 20

40

1.7         3.1         3.9         4.1         3.9         3.5         2.9         1.7           2.5         4.8         5.9         6.8         6.4         5.4         4.1         2.1           3.3         6.4         9.1         11.3         10.5         7.6         5.3         2.7           3.8         7.4         11.1         20.2         19.7         8.8         5.4         2.7           3.5         7.4         11.1         20.1         19.6         9.0         5.5         2.9           3.2         6.0         8.6         10.7         10.2         7.3         4.9         2.7           2.4         4.6         5.5         6.3         5.9         5.1         4.0         2.2           1.9         3.1         3.6         3.8         3.8         3.5         2.8         1.5	<u>No. 1</u> No sample (Nitrogen)	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No. 5</u> Gray paint t1.0mm	0.8         1.1         1.1         1.2         1.3         1.2         1.1         0.7           0.9         1.0         1.1         1.1         1.1         1.1         0.9         0.8           1.0         1.2         1.2         1.1         1.1         1.1         0.9         0.8           1.0         1.2         1.2         1.2         1.1         1.1         1.0         0.9         0.8           1.2         1.2         1.2         1.1         1.1         1.0         1.1         0.9           1.2         1.2         1.0         1.1         1.0         1.0         0.9           1.0         1.2         1.2         1.2         1.2         1.1         1.0         0.9           1.1         1.2         1.2         1.2         1.2         1.1         1.0         0.9           1.1         1.2         1.3         1.1         1.2         1.1         1.2         1.0           0.9         0.9         1.2         1.0         1.1         1.0         1.0         0.8           0.7         1.0         1.3         1.3         1.2         1.1         1.1         0.6 <th><u>No.9</u> α camera light Shielding</th>	<u>No.9</u> α camera light Shielding
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No. 2</u> White paint t0.3mm	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No. 6</u> Black paint t1.0mm		<u>No. 10</u> Frame only
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No. 3</u> White paint t0.5mm		<u>No. 7</u> Pink Covering sheet	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No.11</u> Frame + shielding
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No. 4</u> White paint t1.0mm	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	<u>No.8</u> Transparent Covering sheet	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	No. 12 Transparent covering sheet + shielding

Fig. 1 Test results under all conditions



# Legend Results of obtaining the differential

O: There is an impact  $\Delta$ : Although small there is an impact —: There is no impact

# (b) Technological development of contamination evaluation for sorting solid waste $\beta$ ray light emission test: Analysis of straight line system test results

Table 1 Results of difference processing and remaining effect

Differential No.* ¹	Sample	Supplementar y information	① Pre N2	② Post N2	③ Shielded by sample	④ Shielded by frame	5 Scintillati on light	⑥ Cherenk ov light	⑦ Light emission inside the camera	2 × 2'2	4 × 4°2	8×8'2
2-11	White paint	t0.3mm	-	Δ	Δ	-	0	0	_	-3.9	-15.2	-41.1
3-11	White paint	t0.5mm	-	Δ	Δ	-	0	0	-	-0.1	-10.0	-31.4
4-11	White paint	t1.0mm	-	Δ	Δ	_	0	0	_	-7.1	-23.7	-56.1
5-11	Gray paint	t1.0mm	-	Δ	Δ	-	0	0	-	-0.6	-12.9	-39.9
6-11	Black paint	t1.0mm	-	Δ	Δ	-	0	0	-	1.6	-14.2	-46.3
8-11	Transparent covering sheet		Δ	Δ	Δ	-	0	0	-	33.7	43.8	55.7
8-12	Transparent covering sheet		-	Δ	Δ	-	0	0	-	12.2	19.7	32.4
7-11	Pink covering sheet		-	Δ	Δ	-	0	0	-	98.2	119.2	134.1
1-9	No sample	Nitrogen light emission	0	0	-	-	-	-	-	75.3	176.9	322.3

*1 Difference with the test results on P.146, *2 Refer to Fig. 1 on P.145



Fig. 1 Results of difference processing and remaining effect (including statistical error)



(b) Technological development of contamination evaluation for sorting solid waste Right-angled system for the β ray light emission test Organization of light emission phenomena



Table 1 List of light emission phenomena in a right-angled system

No.	Light emission/phenomena	Memo
1	Nitrogen light emission before the sample	Nitrogen light emission in the 20cm between the radiation source and the sample
2	Nitrogen light emission after the sample	Nitrogen light emission from scattered electrons
3	Reflection of $\textcircled{1}$ by the sample	Reflectivity differs depending on the sample
4	Light emission from the sample Scintillation light	Phenomenon to be clarified 1
5	Light emission from the sample Cherenkov light	Phenomenon to be clarified2, level that can be ignored in the caseof Pm-147
6	Light emission inside the $\boldsymbol{\alpha}$ camera	Generated by the bremsstrahlung or the electrons that have reached the optical system



# (b) Technological development of contamination evaluation for sorting solid waste $\beta$ ray light emission test: Test conditions of the right-angled system and affecting factors

Table 1 PM-147 straight line system : Affecting factors in each condition

No.	Sample	Supplementary information	① Pre N2	2 Post N2	3 Reflected by sample	Generation light	<b>⑤</b> Cherenkov light	Eight emission inside the camera
1	None (Nitrogen)		0	-	-	_	_	-
2	Pink covering sheet		0	-	Δ	0	-	-
3	Transparent covering sheet		0	-	Δ	0	-	-
4	White	Paint	0	-	Δ	0	-	-
5	Light gray (Gray)	Paint (Same as the flange tank piece)	0	-	Δ	0	-	-

#### Table 2 Sr-90/Y-90 straight line system : Affecting factors in each condition

No.	Sample	Supplementary information	① Pre N2	2 Post N2	3 Reflected by sample	A Scintillation light	5 Cherenkov light	6 Light emission inside the camera
6	None (Nitrogen)		0	-	-	-	-	0
7	Pink covering sheet		0	0	Δ	0	-	0
8	Transparent covering sheet		0	0	Δ	0	-	0
9	White	Paint	0	0	Δ	0	-	0
10	Light gray (Gray)	Paint (Same as the flange tank piece)	0	0	Δ	0	-	0
11	α camera light shielding		-	-	-	-	-	0

- Unit: cps

Fig. 1 Data processing position in a rightangled system



(1) Sum total of the data in the  $2 \times 2$  cells in the center

 $\rightarrow$  Size of view equal to that of the sample

2 Sum total of the data in the 4 × 4 cells in the center

 $\rightarrow$  Light from outside the frame is considered as well

③ Sum total of the data in all the 8×8 cells in the center

 $\rightarrow$  Entire light emission is considered

#### Legend

O: There is an impact

 $\Delta$ : Although small there is an impact

-: There is no impact

#### Other test conditions

- Measurement time 10 min
- BG measurement 5min
- ·Signal is the difference with BG
- •Room temperature 20.0°C to 20.4°C
- Pressure 1003.8hPa 1022.3hPa
- •Humidity 31.2% to 43.0%

#### **Processing of detection results**

•Measurement is started after sample to be measured is set up.

• After completing measurement, the radiation source is removed and BG is measured.

• The difference between BG and the sample measurement for each ch is obtained and that value is used as the evaluated test result. (b) Technological development of contamination evaluation for sorting solid waste  $\beta$  ray light emission test: Right-angled system test results

### Results of Pm-147

### Results of Sr-90/Y-90 (Reference)



Fig. 1 Test results under all conditions



# (b) Technological development of contamination evaluation for sorting solid waste $\beta$ ray light emission test: Analysis of right-angled system test results

O: There is an impact  $\Delta$ : Although small there is an impact —: There is no impact

Legend

#### Table 1 Results of difference processing and remaining effect



Fig. 1 Results of difference processing and remaining effect (including statistical error)



(b) Technological development of contamination evaluation for sorting solid waste Purpose of simulation and details of implementation

<Purpose of simulation>

To secure  $\alpha$  contamination detection performance range (lower  $\alpha$  contamination detection limit depending on the on-site environmental conditions) for  $\beta \gamma$  surface contamination that gets spatially distributed.

- 1. Simulation of  $\alpha$  contamination density and spatial distribution
- 2. Simulation of  $\beta$  contamination density and spatial distribution
- 3. Provision as a tool for predicting factors that do not have a measured value or it is difficult to measure their value

<Details of FY2021 implementations>

The following was implemented with a system similar to the  $\beta$  rays light emission test as the basic model.

- 1. Matching the actual nitrogen light emission counting rate in the system in which Am-241 is placed in space.
- 2. Matching the actual nitrogen light emission counting rate in the system in which Sr-90/Y-90 are placed in space.



(b) Technological development of contamination evaluation for sorting solid waste Simulation Geant4 basic model Straight line system





(b) Technological development of contamination evaluation for sorting solid waste Simulation:  $\alpha$  source system

Number of events: 1 × 10⁶[Alpha] Statistical amount: equivalent to

xxx[sec]

0.1	0.1	0.3	0.3	0.1	0.1	0.1	0.1
0.2	0.4	0.7	0.9	0.5	0.2	0.1	0.0
0.2	0.9	3.2	4.8	2.5	0.7	0.1	0.1
0.4	1.1	5.3	21.4	3.8	0.8	0.2	0.1
0.2	0.5	3.0	4.5	3.0	0.6	0.2	0.2
0.3	0.3	0.6	1.0	0.6	0.3	0.1	0.1
0.2	0.2	0.2	0.2	0.2	0.2	0.1	0.1
0.1	0.1	0.1	0.1	0.1	0.1	0.0	0.1

<b>Irradiation conditions</b> Source: Am-241 (5.4Mev, α) Source size: φ3.6cm Direction of emission: 2π	Measured value of the visual fiel source (Am-241)	ld of the
	Simulation:	21
Analytical conditions	Actual measured value :	16

21.4cps 16.8cps

Fig. 1 α rays nitrogen light emission simulation results

It was verified that simulation is possible with an accuracy of 0.5≦ (calculated value)/(actual measured value)≦2 (=Factor2) for radiation source locations in multiple cases



0.3	-0.2	0.2	0.8	1.2	0.0	0.4	0.4
-0.3	0.1	0.3	0.7	0.6	0.0	0.1	-0.1
-0.3	0.3	0.7	1.1	1.0	0.3	-0.3	0.3
-0.3	0.1	1.6	16.8	6.6	0.3	0.5	0.1
-0.1	0.4	0.8	4.4	2.2	0.0	-0.2	-0.2
0.0	-0.2	0.3	0.8	0.6	-0.1	0.0	0.3
-0.2	0.4	0.0	0.0	0.2	-0.3	-0.2	0.2
0.2 0.4		0.1	0.2	0.4	-0.6	-0.2	0.1
0		20		40	Unit:	cps	

Fig. 2  $\alpha$  rays nitrogen light emission simulation results (previously reported in FY2020)



# (b) Technological development of contamination evaluation for sorting solid waste Simulation: $\beta$ source straight line system

Re	esults o	of eval	luating	the n	umber	of det	tection	s of $\beta$	rays	<u>C</u>	ompai	rative e	evalua	tion of	i the si	mulati	on an	d actual
	nitro	igen i	gnt en	lission	трут	eans o	or simu	liation	_				<u>r</u>	neasu	remen	<u>t</u>		
	1.9	3.7	4.4	4.3	4.3	4.4	3.5	2.1			1.1	1.2	1.1	1.1	1.1	1.3	1.2	1.2
	3.9	5.7	6.6	7.9	8.0	7.4	6.0	3.1			1.5	1.2	1.1	1.2	1.3	1.4	1.5	1.4
	4.0	6.0	8.5	9.1	9.1	8.5	7.3	3.3			1.2	0.9	0.9	0.8	0.9	1.1	1.4	1.2
	3.6	7.0	9.1	11.1	11.2	8.2	6.9	3.4	F	Percentage	0.9	0.9	0.8	0.5	0.6	0.9	1.3	1.3
	4.5	7.0	9.2	11.7	11.0	9.2	7.1	3.2			1.3	1.0	0.8	0.6	0.6	1.0	1.3	1.1
	4.1	6.3	7.3	9.2	8.5	9.0	6.7	3.1			1.3	1.0	0.8	0.9	0.8	1.2	1.4	1.2
	3.1	5.7	6.6	7.8	7.8	7.7	6.5	2.6			1.3	1.2	1.2	1.2	1.3	1.5	1.6	1.1
	1.7	3.3	4.5	4.7	5.1	4.0	3.6	1.9			0.9	1.1	1.2	1.2	1.4	1.1	1.3	1.3
Re	sults o	f evalu	uating	the nu	imber	of dete	ections	s of β	rays		0.5		1	2	U	nit: -		
<u>n</u>			emiss		mean	sora			<u>=a)</u>		0.9	0.0	0.5	0.9	0.5	0.0	0.0	0.4
	1.7	3.1	3.9	4.1	3.9	3.5	2.9	1.7			0.2	0.6	0.5	0.2	0.5	0.9	0.6	0.4
	2.5	4.8	5.9	6.8	6.4	5.4	4.1	2.1			1.4	0.9	0.8	1.1	1.6	2.0	1.9	0.9
	3.3	6.4	9.1	11.3	10.5	7.6	5.3	2.7			0.8	-0.5	-0.6	-2.2	-1.4	1.0	2.0	0.6
	3.8	7.4	11.1	20.2	19.7	8.8	5.4	2.7	C	Differential	-0.2	-0.4	-2.0	-9.1	-8.5	-0.6	1.4	0.7
	3.5	7.4	11.1	20.1	19.6	9.0	5.5	2.9			1.0	-0.4	-1.9	-8.5	-8.6	0.2	1.6	0.3
	3.2	6.0	8.6	10.7	10.2	7.3	4.9	2.7			0.9	0.3	-1.3	-1.4	-1.7	1.8	1.8	0.5
	2.4	4.6	5.5	6.3	5.9	5.1	4.0	2.2			0.7	1.1	1.1	1.5	1.9	2.6	2.5	0.3
	1.9	3.1	3.6	3.8	3.8	3.5	2.8	1.5			-0.2	0.2	0.9	0.9	1.3	0.5	0.8	0.4
	0	20	)	40	Un	it: cps	3				-10		0	1(	) Ui	nit: 🛆	cps	

Fig. 1 Results of comparative evaluation of  $\beta$  rays nitrogen light emission (Straight line system)

It was verified that simulation is possible with an accuracy of  $0.5 \le$  (calculated value)/(measured value)  $\le 2$  (=Factor2)



(b) Technological development of contamination evaluation for sorting solid waste Organization of the light emission process: Light emission by nitrogen due to  $\alpha$  rays





(b) Technological development of contamination evaluation for sorting solid waste Organization of the light emission process: Light emission by nitrogen due to  $\beta$  rays





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## (b) Technological development of contamination evaluation for sorting solid waste Appendix. 1 Quantum efficiency of PMT



WAVELENGTH (nm)



(b) Technological development of contamination evaluation for sorting solid waste Appendix. 2 Stopping Power of  $\beta$  rays



"Stopping-Power & Range Tables for Electrons, Protons, and Helium Ions" Source: National Institute of Standards and Technology (NIST) ESTAR Database NIST Standard Reference Database 124 DOI: https://dx.doi.org/10.18434/T4NC7P



# (b) Technological development of contamination evaluation for sorting solid waste Quantitative organization of the light emission process

- <β rays light emission test>
- ·Light emission test with Sr-90/Y-90, Pm-147 as the radiation source was completed.
- ·It was verified that paint emits little light.
- It was verified that the covering sheet emits lot of light.

It was verified that scintillation light is generated. Meanwhile, it was found that whether or not Cherenkov light is generated needs to be studied in further detail.

·It was verified that there is lot of nitrogen light emission in air.

• It was confirmed that the noise in FY20202 was likely to be mainly due to covering sheet (pink) and nitrogen light emission.

<Summary of simulation>

•As a result of developing a model simulating the phenomenon of light emission by nitrogen due to  $\alpha$  radiation source and evaluating the amount of light detected by the  $\alpha$  camera, it was found that it had a Factor2 accuracy (in comparison with the actual measured value).

•As a result of developing a model simulating the phenomenon of light emission by nitrogen due to  $\beta$  radiation source and evaluating the amount of light detected by the  $\alpha$  camera, it was found that it had a Factor2 accuracy (in comparison with the actual measured value).

<Quantitative organization of the light emission process Important items>

•2 to 3 Photon were detected around the  $\alpha$  rays, and 3.6 × 10⁻⁸Photon were detected around the  $\beta$  rays.

•The number of photons detected around  $\alpha$  rays as well as  $\beta$  rays largely change depending on the contamination density and the air dose rate.

•In the light emission test conducted this year, as against a maximum of approx. 17cps at 1ch for Am-241 of 900Bq, in the case of Sr-90/Y-90 it was a maximum of approx. 20cps for a ch with an air dose rate of 15.6mSv/h.

•The model needs to be extended to include a spatial spread of contamination and change in density in its simulation.



## (b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Overview of the policy

Table 1 Possible countermeasures for each light emission phenomenon

Light emission phenomenon	Items that must be considered	Possible countermeasures
Scintillation light	<nitrogen emission="" light=""> <ul> <li>Light emission by nitrogen due to α rays as well as β rays has the same wavelength distribution</li> <li>The state of light emission is likely to differ depending on the distribution and density of α contamination and β contamination.</li> <li>The duration of light emission is longer than that from the sample.</li> <li>Light emission other than from nitrogen&gt;</li> <li>The wavelength range differs depending on the material of the source of light emission.</li> <li>The amount of light emitted differs depending on the material of the source of light emission.</li> <li>Light spreads from where the source of light emission is present.</li> <li>The duration of light emission is in the ns order</li> </ul></nitrogen>	<nitrogen emission="" light=""> <ul> <li>Distinction using the difference in the state of light emission</li> <li>Indication of the range within which α contamination can be detected with respect to β contamination (lower detection limit of α contamination depending on the on-site environmental conditions).</li> <li>Light emission other than from nitrogen&gt;</li> <li>Filter that uses the difference in wavelength range</li> <li>Distinction using the difference in duration of light emission</li> <li>Prevention by taking measures such as not using samples that become a source of light emission, elimination in advance, etc.</li> <li>Distinction using the concurrence of light emission</li> </ul></nitrogen>
Cherenkov light	<ul> <li>The wavelength distribution is ∝λ⁻²</li> <li>Amount of light emission depends on the refractive index n.</li> <li>The direction of light emission is conical at an angle of cosθ=c/nv (v: velocity in a medium) with respect to the incident direction of charged particles.</li> <li>The duration of light emission is in the ps order</li> </ul>	<ul> <li>Filter that uses the difference in wavelength distribution</li> <li>Distinction using the difference in duration of light emission</li> <li>Distinction using the concurrence of light emission</li> </ul>

<Noise countermeasures implementation items>

- 1. Measuring the scintillation light and Cherenkov light by means of the  $\beta$  rays light emission test
- 2. Checking the current α camera signal processing circuit, and verifying the possibility of temporal discrimination of each type of light emission by means of the above-mentioned test
- 3. Comprehensively determining the countermeasures that must be taken based on the above

(b) Technological development of contamination evaluation for sorting solid waste Study of noise countermeasures: Current  $\alpha$  camera signal processing circuit

The possibility of temporal discrimination based on the light emission phenomenon and the circuit time constant is studied as a means for noise discrimination.



### IRID

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(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Signal processing time constant and count

 Table 1
 Signal processing from photon incidence to signal

Circuit	Impact
Charge sensitive Gain (x500, 1MHz max)	Signal amplification
Baseline restorer Pole-zero cancellation	Waveform shaping



Fig. 1 Example of signals before and after signal processing (1 event)

Table 2 Signal counts

Circuit	Impact
Window discriminator	Cut at the upper and lower
Schmitt trigger	

Threshold range that sufficiently includes light emission due to  $\alpha$ ,  $\beta$ 



Fig. 2 Example of conversion to the count

Current  $\alpha$  camera circuit has a time constant in the  $\mu$ s order

ightarrow Distinction cannot be made based on duration of light emission by the Cherenkov light or the scintillation light



(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures : Results of evaluating the time constant of the current circuit (Verification of the actual signal)



Fig. 1 Signal waveform when  $\alpha$  ray source is measured

When 1 photon is incident, the output is 1.6mV, and by Gain × 500 the peak height is around 800mV.
The width of the output waveform is approx. 2µs between 10% and 90%.

 $\rightarrow$  The current  $\alpha$  camera circuit does not have a temporal resolution of less than  $\mu$ s.



## (b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Study of the likelihood of speeding up

Table 1	Possible cou	Intermeasures	for each l	light emis	ssion phe	nomenon
---------	--------------	---------------	------------	------------	-----------	---------

Light emission phenomenon	Items that must be considered	Possible countermeasures
Scintillation light	<nitrogen emission="" light=""> <ul> <li>Light emission by nitrogen due to α rays as well as β rays has the same wavelength distribution</li> <li>The state of light emission is likely to differ depending on the distribution and density of α contamination and β contamination.</li> <li>The duration of light emission is longer than that from the sample.</li> <li>Light emission other than from nitrogen&gt;</li> <li>The wavelength range differs depending on the material of the source of light emission.</li> <li>The amount of light emitted differs depending on the material of the source of light emission.</li> <li>Light spreads from where the source of light emission is present.</li> </ul></nitrogen>	<nitrogen emission="" light=""> <ul> <li>Distinction using the difference in the state of light emission</li> <li>Indication of the range within which α contamination can be detected with respect to β contamination (lower detection limit of α contamination depending on the on-site environmental conditions).</li> <li>Light emission other than from nitrogen&gt;</li> <li>Filter that uses the difference in wavelength range</li> <li>Distinction using the difference in duration of light emission</li> <li>Prevention by taking measures such as not using samples that become a source of light emission, elimination in advance, etc.</li> </ul></nitrogen>
Cherenkov light	<ul> <li>The wavelength distribution is ∝λ⁻²</li> <li>Amount of light emission depends on the refractive index n.</li> <li>The direction of light emission is conical at an angle of cosθ=c/nv (v: velocity in a medium) with respect to the incident direction of charged particles.</li> <li>The duration of light emission is in the ps order</li> </ul>	<ul> <li>Filter that uses the difference in wavelength distribution</li> <li>Distinction using the difference in duration of light emission</li> <li>Distinction using the concurrence of light emission</li> </ul>

1) Use of a high-speed amplifier with a band frequency of 100Ghz or more

 $\rightarrow$  Not realistic considering the cost of improving the circuit system, compatibility with 64ch, etc.

2) Replacement of the I/V amplifier of the ns order with a charge sensitive amplifier

 $\rightarrow$  Since the peak height of the I/V amplifier depends on the number of incident photons, if the number of incident photons changes depending on the duration of light emission, the peak height changes as well.

(Expected phenomenon)

In the case of scintillation light that is emitted for ns and Cherenkov light that is emitted for ps, if light is detected continuously throughout the duration of light emission, the peak height of the scintillation light rises.

Verification by means of the light emission test in which scintillation light and Cherenkov light are separated



(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: I/V amplifier verification by means of the  $\beta$  rays light emission test



Fig. 1 Light emission measurement system using the I/V amplifier

Table 1 Layout and light emission phenomenon to be measured

PMT Layout	Perceived light emission phenomenon
1	Nitrogen light emission (scintillation light) was measured at the location where incidence of the beams of electrons was avoided.
2	Nitrogen light emission was shielded and Cherenkov light was measured by allowing incidence of electron beams.



(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Results of I/V amplifier verification by means of the  $\beta$  rays light emission test



Fig. 1 Waveform in which light emission is detected in layout ①

 Incidence of nitrogen light emission was 3.6 × 10⁻⁸Photon/Beta (P.157)

•1Photon to several Photon were detected in this system as well



Fig. 2 Waveform in which light emission is detected in layout ②

•Number of incident photons was extremely numerous in Cherenkov light from the window

• There were waveforms in which large quantities of photons were detected in this system as well

•As it is assumed that 1 photon onwards per event is detected at the site, for example in a low air dose rate environment it is difficult to identify based on peak height.

•In a high air dose rate environment where photons from multiple events are detected over a certain duration, the light emission duration and number of incident photons are likely to be correlated, and in such a case it is possible to identify them.

 $\rightarrow$  The possibility needs to be ascertained considering multiple factors such as spatial distribution of contamination, its density, placement of the source of light emission, etc.



## (b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Simultaneous counting method

#### *Principle of the simultaneous counting method and its effect

- This is a technique for distinguishing between signal and noise using the concurrence of light emission detection. Hence the same view is measured in 2 PMTs.
- If nitrogen light emission is considered as the target example, as against β rays, since the number of incident photons per event in the case of α rays are larger, the decrease in β rays nitrogen light emission counting rate can be expected to be far greater than the decrease in α rays nitrogen light emission counting rate. (S/N improvement can be expected)
- This effect is presumed to be a decreasing effect common to noise generated at random times such as thermal noise, electrical noise, noise generated by light, noise generated by γ rays, etc. regardless of the light emission principle.
- However, when the frequency of noise generation increases, noise gets accidentally counted simultaneously leading to a false detection.
- ⇒ The sensitivity^{*} of the signal from an ancillary detector that optimizes S/N should be evaluated.

*Generally, if the sensitivity to  $\alpha$  rays are enhanced, noise sensitivity such as sensitivity to  $\beta$  rays increases as well.



IRID

(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Study of the composition of the ancillary detector



#### Table 1 List of prospective elements

RI

(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Preliminary evaluation of the simultaneous counting method

Table 1Evaluation system

Classifi cation	Composition
А	Current system 8 × 8 array + Short focus lens
В	φ10mm Single PMT + Long focus lens
С	φ100mm Single PMT

The combination simulating the simultaneous counting method

A, A-B, A-C, B-B (Reference)

Transmission rate (%) No. **Filter conditions** Remarks Nitrogen light Cherenkov emission 275nm to 375nm 100 100 Past 1 Entire nitrogen light 2 315nm to 375nm 100 52.4 emission Part of the nitrogen 3 337nm to 375 79 31 light emission 18.2 4 337nm±10nm 42 Part of the nitrogen light emission



Fig. 1 Results of evaluating the lower detection limit



#### Table 2 Analytical conditions

(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Test manufactured ancillary detector and the test set-up





## (b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures

Evaluation of the effectiveness of the simultaneous counting method

Testing Measu tw (time	rement e gate v	ons time: 9 vidth):	720sec 1µs		) i	Placeme	ent of A	∖m-24	11							
			0		1	5 Un	it: cps					0		8x10 ⁻²	Unit:	cps
0.78	0.97	0.82	1.45	3.43	2.16	0.98	1.69		1.5E-02	2.9E-03	2.8E-03	2.4E-03	1.6E-03	1.1E-03	1.5E-03	1.5E-02
0.51	0.80	1.07	1.93	1.89	1.25	0.65	0.72		4.0E-03	1.0E-03	6.2E-04	8.2E-04	8.2E-04	7.2E-04	6.2E-04	4.5E-03
0.53	1.13	2.67	2.93	1.91	1.28	0.72	0.62		4.3E-03	1.0E-03	1.3E-03	3.9E-03	3.6E-03	2.0E-03	6.2E-04	3.2E-03
0.64	1.25	7.86	5.81	5.99	1.36	0.69	0.57		9.5E-03	1.9E-03	2.3E-03	3.0E-02	4.3E-02	3.4E-03	2.4E-03	7.2E-03
0.52	1.00	3.98	10.68	15.21	1.59	0.91	1.05		5.9E-03	1.0E-03	3.8E-03	4.5E- <b>0</b> 2	8.2E-02	5.0E-03	2.3E-03	9.3E-03
0.46	0.72	1.42	3.48	2.90	1.59	0.91	0.57		3.0E-03	5.1E-04	1.9E-03	4.1E-03	5.9E-03	2.0E-03	1.2E-03	3.2E-03
0.45	0.83	0.76	0.86	1.31	0.91	0.58	0.41		4.8E-03	1.1E-03	3.1E-04	2.2E-03	2.8E-03	1.3E-03	9.3E-04	2.5E-03
2.32	0.60	0.84	0.85	0.85	0.85	0.62	0.46		1.6E-02	5.2E-03	2.9E-03	4.6E-03	6.9E-03	4.1E-03	3.9E-03	1.3E-02

Fig. 1 Before adopting the simultaneous counting method

Fig. 2 After adopting the simultaneous counting method

Noise such as BG, etc. besides the Am-241 view is reduced

• The counting rate is lowered to 0.5%, measurement time is approx. 16 times (Measurement time in this test / normally 10 minutes)

 $\rightarrow$  It is essential to narrow down the process to be applied (Example: high radiation dose region, etc.)



(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Noise reduction by means of the band-pass filter



Fig. 1 Spectrum of each type of light emission and filter characteristics of the conventional band-pass filter

(*) H.Nakamura et.al., "Evidence of deep-blue photon emission at high efficiency by common plastic", EPL, 95 (2011)



(b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Frequency characteristics of the prospective band-pass filter



Fig. 1 Filter characteristics of the prospective band-pass filter

Each band-pass filter is evaluated for sensitivity after it is applied



## (b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures

Evaluation of S/N ratio after the application of the prospective band-pass filter

Band-pass filter	Amount of nitrogen light emission (S) (*)	Amount of Cherenkov light (N) (*)	S/N	Priority
Past	1.00	1.00	1.00	5
Option 1	1.15	0.75	1.54	4
Option 2	0.96	0.59	1.62	3
Option 3	0.96	0.57	1.68	1
Option 4	1.08	0.66	1.62	2

Table 1 Results of evaluating the S/N for each type of prospective band-pass filter

*Standardized with numerical values of conventional band-pass filters

The following prospects were seen based on the evaluation of prospective band-pass filter 3 that had its peak at a wavelength longer than 337nm which was the maximum peak of nitrogen light emission.

• The amount of Cherenkov light reduced to approx 1/2 as compared to the conventional band-pass filter

S/N improved to 1.7 times.



## (b) Technological development of contamination evaluation for sorting solid waste Noise countermeasures: Conclusion

Proposed countermeasures were selected based on the characteristics of the Cherenkov light and scintillation light and their effectiveness was evaluated.

Feasibility of distinction using the difference in duration of light emission

· It was verified that the current  $\alpha$  camera signal processing circuit has a temporal resolution of  $\mu$ s.

•As a result of studying the applicability of the I/V amplifier having a temporal resolution of ns, it was found that the number of detected photons has a correlation with the peak height, however, it was confirmed that the relation between the time taken to detect photons with the Cherenkov light and scintillation light depending on the site environment and the number of photons detected needs to be sorted out.

Feasibility of distinction using the difference in wavelength distribution

•Several band-pass filters were selected in which the transmission characteristics were shifted to the longer side of the wavelength for Cherenkov light that is distributed on the shorter side of the wavelength as compared to nitrogen light emission.

• It was confirmed that the S/N ratio improved to approx. 1.7 times by using band-pass filters that bring about the most improvement in S/N.

Noise reduction using the simultaneity of light emission detection

• The composition that provides approx. 4 times the sensitivity was selected as the ancillary detector for using the simultaneous counting method.

• The counting rate and measurement time were evaluated by means of the verification test using the test manufactured ancillary detector.

- •Noise components such as BG, etc. reduced, but it was found that the counting rate decreased to 0.5%.
- It was found that the required measurement time was approx. 16 times.

• It was found that the process to be used for simultaneous counting needs to be narrowed down.

## (b) Technological development of contamination evaluation for sorting solid waste Consolidation of issues: Issues in distance measurement

### Table 1 Issues based on site investigation

Items	Details of the issues	Study plan
Issues related to distance measurement	Since distance is measured based on the time-of- flight of infrared rays (859nm), and unless the infrared rays return their time-of-flight cannot be measured, the conditions are such that distance cannot be measured.	Whether or not distance can be measured with respect to the material found during the site investigation, using the angle of incidence of the infrared rays as the variable, will be evaluated.
Issues related to synthesizing the distance measurement results	Since the results are synthesized based on the shape characteristics, it may not be possible to synthesize results in the case of wall surfaces, etc. where there is absolutely no change in shape.	Whether or not it is possible to synthesize results pertaining to objects that do not change their shape, by using an image of the outer appearance, and combining the characteristics of the image as well while synthesizing the results, will be clarified.

#### Table 2 Test conditions for verifying the range in which distance measurement is feasible

Assumed target	Material	Shape	Typical dimensions	Shooting distance	Shooting angle
Wall surface, floor surface	Epoxy coating (Gray)	Flat plate			0° (Front facing) to 90°
Covering sheet	Polyethylene	Flat plate		Selected from the α amera angle of view and the actual dimensions of the target object	0° (Front facing) to 90°
Cables	Vinyl chloride (Black)	Cylindrical	Selected from the $\alpha$ camera angle of view		-
Instrument window	Acrylic	Flat plate	dimensions of the		0° (Front facing) to 90°
Instrument window	Glass	Flat plate	larger object		0° (Front facing) to 90°
Water accumulation	Water	-			0° (Front facing) to 90°



(b) Technological development of contamination evaluation for sorting solid waste Evaluation of the range in which distance measurement is feasible: Test set-up (1)



Table 1	Test conditions for verifying the range in which distance measurement is feasible							
Assumed target	Material	Shape	Typical dimensions	Shooting distance	Shooting angle			
Wall surface, floor surface	Epoxy coating (Gray)	Flat plate			0° (Front facing) to 90°			
Covering sheet	Polyethylene	Flat plate			0° (Front facing) to 90°			
Cables	Vinyl chloride (Black)	Cylindrical	Selected from the α camera angle of	Uniformly spaced from 1000mm to	-			
Instrument window	Acrylic	Flat plate	dimensions of the target object	3000mm	0° (Front facing) to 90°			
Instrument window	Glass	Flat plate			0° (Front facing) to 90°			
Water accumulation	Water	-			0° (Front facing) to 90°			



Unit: mm



(b) Technological development of contamination evaluation for sorting solid waste Evaluation of the range in which distance measurement is feasible: Evaluation results

Table 1 Results of verification with concrete as the target of measurement and L=1000mm



Red portion: Distance measurement results not available

Results of verifying the difference in the surface of concrete

(1) The middle section of the front facing part could not be measured because of regular reflection. Can be imputed.

(2) Epoxy resin cannot be measured at  $60^{\circ}$ , but it becomes impossible to measure concrete (solid) at  $75^{\circ}$ .

This is presumed to be an impact of the reflectivity or unevenness of the object to be measured. (3) Concrete (chipped) can be measured up to  $75^{\circ}$ .




Results of verification with cable as the target of measurement

- (1) A cable of  $\varphi$ 20 cannot be measured
  - (i) A cable of  $\varphi$ 20 cannot be measured at 15° or less (data missing)

(ii) It is not possible to measure the reflection from the cable in the case of a cable of  $\varphi 20$  at  $30^{\circ}$  or more and a cable that is  $\varphi 7$  or smaller. Light reflected from the wall surface in the background is measured in that case.





Table 1 Results of verification with transparent materials as the target of measurement and L=1000mm



Red portion: Distance measurement results not available



Fig. 1 Distance image Color is the same as the background (same distance), and hence it has not been possible to measure the distance of the target



Fig. 2 To be measured (Left: acrylic, right: glass)

Results of verification with transparent materials as the target of measurement (1) The middle section at 0° could not be measured. This is presumed to be because of the high reflection intensity caused by regular reflection.

(2) As the distance to the background is included in the measurement result of the target portion, it is not possible at any angle. In the case of transparent material, the reflection intensity of the target is weak, and it is presumed that the reflection of the transmitted background is measured.



Table 1 Results of evaluating the impact of the distance when the covering sheet is the target of measurement



Red portion: Distance measurement results not available

Results based on the difference in distance (covering sheet as the material, with infrared permeability) (1) At a distance of 1m, at  $60^{\circ}$  or more, distance cannot be measured.

(2) At a distance of 2m or more, distance cannot be measured at any angle. In the case of transparent material, the intensity of reflected light is weak, and it is presumed that it gets weaker as the distance increases.

#### IRID





Fig. 1 Test set-up with water surface as the target of measurement

Verification of the impact of water accumulation found on the floor surface on distance measurement

•The depth of the water accumulation is assumed to be approx. 1mm to 20mm

•Assuming that a TOF camera is installed on a remotely operated cart, it is fixed at a height of approx. 900mm. •Since the tilt angle range of the remotely operated cart is 0° to 55° ( $\theta = 35^{\circ}$  to 90°),  $\theta$  is considered to be 30° onwards

•Since the distance becomes greater than 3000mm when  $\theta = 75^{\circ}$ , measurement is carried out with  $\theta$  in the range of 30° to 60°.







Table 3 Evaluation results when d = 20mm

Material	<b>30°</b>	45°	60°
Epoxy resin (100mm square)			
Concrete (Solid, 100mm square)			
Only water			and a second

Results based on the difference in depth of water and angle

•Measurement was possible regardless of the depth of water and angle in the scope of this evaluation.





Table 2 Evaluation results when d = 10mm

Red portion: Distance measurement results not available

	rable i Summary of t		test for verifying the	ange in which	uistance measuren	ilent is leasib	
Assumed target	Material	Shooting distance	Range in which shooting is feasible (Angle)	Assumed target	Material	Depth of water	Range in which shooting is feasible (Angle)
		1000mm	0° (Front facing) to 45°	Water accumulation		Approx. 1mm	30° to 60°
	Epoxy coating (Grav)	2000mm	0° (Front facing) to 45°		vvater + Epoxy coating (Gray)	10mm	30° to 60°
		3000mm	0° (Front facing) only			20mm	30° to 60°
		1000mm	$0^{\circ}$ (Front facing) to $60^{\circ}$			Approx. 1mm	30° to 60°
	Concrete (Calid)	2000mm	$0^{\circ}$ (Front facing) to $00^{\circ}$		Water + Concrete (Solid)	10mm	30° to 60°
vali surface, floor surface	Concrete (Solid)	2000mm	0 (Front facing) to 60			20mm	30° to 60°
		3000mm	0° (Front facing) to 45°				
		1000mm	0° (Front facing) to 75°				
	Concrete (Chipped)	2000mm	0° (Front facing) to 60°				
		3000mm	0° (Front facing) to 45°				
	Covering sheet + Epoxy coating (Gray)	1000mm	$0^{\circ}$ (Front facing) to $60^{\circ}$				
Floor surface		2000mm	0° (Front facing) to 60°				
(covered)	Covering sheet + Concrete (Solid)	1000mm	0° (Front facing) to 60°				
		2000mm	0° (Front facing) to 60°				
		1000mm	$0^\circ~$ (Front facing) to $45^\circ$				
Covering sheet	Polyethylene	2000mm	Measurement not possible				
		3000mm	Measurement not possible				
Instrumente	Glass	1000mm to 3000mm	Measurement not possible				
Instruments	Acrylic	1000mm to 3000mm	Measurement not possible				
	Ethylene propylene rubber (Black, φ20)	1000mm to 3000mm	Measurement not possible				
Wires, cables	Polyvinyl chloride (Black, $\phi$ 7)	1000mm to 3000mm	Measurement not possible				
	Polyvinyl chloride (Black, φ3)	1000mm to 3000mm	Measurement not possible				

Table 1 Summary of the results of the test for verifying the range in which distance measurement is feasible

The direction of access for each material shall be based on input from the site



(b) Technological development of contamination evaluation for sorting solid waste Evaluation of the synthesis of distance measurement results: Configuration study

The offset (error) is minimized by comparing 3 dimensional point clouds using ICP (Iterative Closest Point) algorithms.

 $\rightarrow$  This is repeatedly carried out for all measurement data to minimize overall offset.

•An ICP algorithm involves the following:



Fig. 1 Past algorithms for synthesizing distances

When there is no change in the shape of the target object, or when the shape of the target object is repeated, the position at which the difference in shape (error) is the minimum is uniquely not asked.

#### Countermeasures

Using the color information (texture, pattern) of the images that are simultaneously photographed, the difference in shape and difference in color information is evaluated, and the point cloud is moved to the position where this difference is the least.



#### (b) Technological development of contamination evaluation for sorting solid waste Evaluation of the synthesis of distance measurement results: Test set-up

Test conditions for evaluating the synthesis of distance measurement results

- The target object does not have any characteristic shape, it is either a plane surface or the shape is repeated.
- The target object has texture (pattern)

The synthesis of distance measurement results when images are captured under the above-mentioned conditions is verified.

•Assuming that a TOF camera is installed on a remotely operated cart, the camera is installed at a height of approx. 900mm.

 Assuming the measured distance expected at the time of measurement, the camera is installed 1000mm from the front facing wall surface.

• The tilt angle is in the range of -60° to 60° , and the pan angle is in the rage of 0° to 30° .





Measurement target (floor)



## Measurement target (wall surface)

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Fig. 1 Distance synthesis evaluation test set-up and external appearance of the object to be evaluated





Fig. 1 Results of distance synthesis of each target object

The effectiveness of the distance synthesis algorithm that uses color information is verified.



#### (b) Technological development of contamination evaluation for sorting solid waste Distance measurement system: Summary

<Issues concerning the distance measurement system> Materials found during the site investigation were subjected to a measurement test. It was found that those materials can be measured within the following ranges. Epoxy resin : 0° to 45° Concrete coat: 0° to 45° Solid concrete: 0° to 60° Chipped concrete: 0° to 75° Φ3-20mm cable: Cannot be measured Acrylic, glass: Cannot be measured Covering sheet: 0° to 45° @1m, Cannot be measured @ 2m or more Concrete coating under water: Entire area

Issues related to synthesizing the distance measurement results>

The following was implemented for wall surfaces and floor surfaces in which the shape does not change or in which the same shape is repeated.

•Modified to an algorithm for evaluating the difference in color information (texture, pattern)

• It was verified that distance synthesis is possible even though it could not be performed for the target object using conventional algorithms.



#### (b) Technological development of contamination evaluation for sorting solid waste Reference material: TOF camera and stereo camera

#### Table 1 TOF camera and stereo camera comparative chart

Items	TOF Camera	Stereo camera
Measurement principle	Active distance measurement using the TOF of infrared rays	Passive distance measurement using trigonometry
Characteristics	<ul> <li>Measurement is possible if the target object reflects infrared rays</li> <li>Ambient light has little impact (Measurement is possible even in low intensity light)</li> <li>Not suitable for measuring long distances as infrared rays get diffused and their intensity reduces</li> </ul>	<ul> <li>The target object needs to have a pattern on the surface (Not suitable for uniform planes such as walls, floor, etc.)</li> <li>Sufficient ambient light is required</li> <li>The range within which measurement is feasible can be adjusted based on the distance between the angle of view and the camera</li> </ul>



The distance is calculated based on the timeof-flight of infrared rays

Fig. 1 Schematic diagram of TOF camera

Measurement target

Fig. 2 Schematic diagram of stereo camera

A TOF camera is used for this system considering the range of distances measured by  $\alpha$  camera, and the likelihood of not being able to provide sufficient lighting as dismantling is underway,



#### (b) Technological development of contamination evaluation for sorting solid waste Consolidation of issues: Issues concerning access

#### Table 1 Issues based on site investigation results

Items	Details of the issues	Study plan	On-site action plan
Issues related to movement	Cannot pass due to level difference on the floor surface.	The location of level difference that hinders movement will be clearly specified based on the architectural drawings of the building and photographs.*	Temporary slope will be provided where the level difference hinders movement.
	Cannot pass due to narrow passageways, or the range within which measurement is possible gets restricted as the direction of the cart cannot be changed due to narrow passageways.	The locations where passageways are narrow will be identified based on architectural drawings of the building and photographs, and the range within which measurement is possible will be clarified based on the position of the cart while passing.	
Issues related to measurement	Due to its combination with the remotely operated cart, the $\alpha$ camera is installed at a higher location. Hence the area in the vicinity of the floor surface where measurement is feasible is likely to get restricted.	The area where measurement is possible will be clarified by studying the arrangement when the camera is combined with the remotely controlled cart.	-

*To be studied: Unit 3 - 1F, Unit 2 - 5F

α camera

Pan tilt mechanism

Hoisting mechanism

Image when combined with a remotely operated cart (decontamination system for elevated location)





Crawler

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(b) Technological development of contamination evaluation for sorting solid waste Schematic diagram of the  $\alpha$  camera measurement system mounted on the cart



Proposed structure when the camera is mounted on the existing crawler will be developed



#### (b) Technological development of contamination evaluation for sorting solid waste Conception picture of the camera mounted on the cart and the results of consolidating the issues



Fig. 1 Conception drawing of mounting the camera on the decontamination crawler

lssues concerning the access cart>

- · Avoiding the risk of turning over (center of gravity of the crawler)
- The height of the floor surface exceeds the height required for the  $\alpha$  camera (Additional installation of hoisting mechanism, etc.)
- Power shortage for the pan tilt mechanism (due to increase in weight as a result of mounting the ancillary detector)

•The decontamination crawler can handle a level difference of approx. 40mm. For larger level differences slopes need to be additionally installed.



#### (b) Technological development of contamination evaluation for sorting solid waste Summary

### **Results so far**

- The light emission processes from light emission due to α rays and β rays to signal detection were quantitatively organized, and a simulation model was developed. Further, the accuracy of simulation was evaluated by verifying the bare minimum simulation model consisting of the radiation source and α camera built based on the light emission test.
- ✓ Nitrogen light emission was evaluated using the basic model and it was confirmed that accuracy was 0.5 ≤ (calculated value)/(measured value) ≤2 (=Factor2).
- ✓ The physical quantity of each process of nitrogen light emission was quantitatively organized based on the simulation results.
- Temporal discrimination using high speed circuit as a noise countermeasure, simultaneous counting method using the concurrence of light emission, filters that use the difference in wavelength range, were studied, and their respective effectiveness was verified.
- Based on site investigation results, the range within which measurement is feasible was clearly specified focusing on materials that could become a hindrance in the distance measurement system. Also, it was confirmed that the distance measurement results can be synthesized for surfaces that do not undergo any change in shape (wall, etc.).
- $\checkmark$  The conception drawing of the access cart on which the improved α measurement system is mounted was created. And, the issues anticipated until application to actual equipment were clearly specified.

#### Issues, etc.

- $\checkmark$  Development of a simulation model simulating the spatial distribution of  $\beta$  contamination
- ✓ Development of a simulation model simulating light emission sources such as paint, sheets, etc.
- Indication of the α contamination detection performance range (lower α contamination detection limit depending on the on-site environmental conditions) when the air dose rate (+ distribution) and material expected on the site are simulated.
- Applicability assessment of α camera based on the α contamination detection performance range and clear specification of the incidental on-site environmental conditions



### 2. Project details (No. 2)

- b. Establishment of concepts of treatment and disposal, and development of safety assessment methods
  - (a) Establishment of selecting advance treatment methods
    - [1] Low-temperature treatment technique
    - [2] Study on approach to assessing applicability for treatment technology
  - (b) Provision of disposal methods and development of safety assessment methods
    - Information organization to study the disposal concepts and to establish safety assessment methods
    - [2] Development of techniques for assessing impact of affecting substances, etc. on disposal



#### (a) Establishment of selecting advance treatment methods

#### Achievements up to FY2020

- Techniques (approaches) that enable multifaceted evaluation of technology were presented by identifying the evaluation axes for evaluating both waste characteristics and treatment technology.
- Of the evaluation axes that were considered in connection with low-temperature treatment technologies (cement solidification and Alkali Activated Materials (AAM) solidification), where data on characteristics of solidified substances was insufficient, it was acquired through simulated mixed waste system.
- Goal
- O To acquire and evaluate data on high temperature treatment technologies and low temperature treatment technologies, required for identifying stabilization and solidification technologies expected to be applicable to actual treatment, so as to contribute to the establishment of techniques for selecting advance treatment methods.
- Implementation items and overview

Implementation items		Overview	Indicators for the achievements of goal	
① Low temperature	i. Study on techniques for verifying the feasibility of low-temperature	Study of quantification by means of image determination	Screening technique incorporating the image determination method is presented.	
treatment technologies	treatment solidification	<ul> <li>Study of applicability while taking the extensive waste generated at the Fukushima Daiichi Nuclear Power Station into consideration, and in addition, acquisition of required data</li> </ul>		
	ii. Investigation of the change in properties of solidified material due to heating, etc.	<ul> <li>Acquisition of data on cyclical conditions, etc. simulating the storage environment</li> </ul>	Data concerning the constituent phase of solidified material under storage environmental (cyclic) conditions are investigated and investigation results are presented.	
	iii. Evaluation of the relationship between the inventory of Cs, etc. and temperature of solidified material	<ul> <li>Analysis related to the impact during accumulated storage of square shaped containers</li> </ul>	The temperature attained when square shaped containers are accumulated and stored is evaluated and the method of calculating it is presented.	
	iv. Investigation and evaluation of factors affecting long-term degradation	<ul> <li>Investigation of the method of accelerated testing of degradation</li> <li>Data acquisition through accelerated testing and study of applicability</li> </ul>	The method of accelerated testing of degradation is investigated and the investigation results are presented.	
② Study on approach towards evaluating applicability of	i. Organization of acquired data and evaluation axes used for comparing technologies	Summarization of the data acquired through tests along with existing data, and organization of evaluation axes	Techniques (approach methods) for evaluating solidification technologies that are likely to be applicable to various solid waste are presented.	
treatment technologies	ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control	<ul> <li>Acquisition of data related to Cs volatilization control based on tests</li> </ul>	Results of investigation of the volatilization control effect of various events is presented.	



(a) Establishment of selecting advance treatment methods

- Relationship between the developed techniques and the technology selection process -





#### (a) Establishment of selecting advance treatment methods

- Flow of the study for establishing the approach (techniques) towards evaluating applicability of treatment technologies -
- ✓ During low temperature solidification treatment, the relationship between change in properties of solidification material due to the physical and chemical properties of waste, and the performance required of the solidified material needs to be considered.
- The required time period differs depending on the performance (Fig. 1)
- Ascertaining performance during and after treatment
- Confirming changes in the constituent phases focusing on longterm stability (heating, drying (radiation), duration)

Performance	Treatment	Storage	Disposal
① Fluidity			
② Hardening rate			
③ Compressive strength		I	
④ Hydrogen generation rate			
5 Nuclide leaching rate		l	

Fig. 1 The time period during which performance factors and properties of major solidified substances determining the limit values (waste filling rate) determined by waste properties during the solidification treatment are required





# (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique i. Study on techniques for verifying the possibility of low-temperature solidification treatment - Details of implementation -

- Achievements up to FY2020
- Techniques for determining the feasibility of low-temperature solidification treatment (cement, AAM) were studied based on properties of various types of solid waste, and a technique consisting of primary screening (evaluation of possibility of solidification) and secondary screening (composition study and verification of performance of the solidified substances) was developed. Also, quantification was carried out on a trial basis using image determination during the primary screening, to verify applicability.
- The following issues were identified with respect to on-site application and further refinement of data.
  - ✓ Creation of quantitative criteria for determination during primary screening.
  - Extensive verification using chemical reagents, etc. assuming various effects on the solidification material, in order to generalize the examination method.
- Objective

To study quantification by means of image determination for the low-temperature solidification treatment technologies of cement solidification and AAM solidification, and make proposals. Also, to study applicability of the proposed technique taking the extensive waste generated at Fukushima Daiichi Nuclear Power Station into consideration, and in addition, to acquire and compile the required data.

#### **Details of implementation**

#### Verification of the effect of fluidity improvement due to chemical admixtures

- Chemical admixtures (water reducing agents) that are expected to have a fluidity improvement effect with respect to cement and AAM will be investigated, and selected.
- As secondary screening (composition study and verification of performance of solidified substances), the effect of adding the selected chemical admixtures expected when actual solidification conditions are set up will be studied.

Study of quantification by means of image determination

- Addition of inspection items and the method of acquiring data on solidification properties will be studied.
- Quantification by means of image determination equipment will be studied and will be reflected in the screening technique.



#### Indicators for the achievements of goal

Screening technique incorporating the image determination method is presented.



## (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique i. Study on techniques for verifying the feasibility of low-temperature solidification treatment - Selection of chemical admixtures -

- This study focused on 4 types of typical chemical components from amongst water-reducing agents and high-performance water-reducing agents (hereinafter described as chemical admixtures) used for improving the fluidity of cement-based material.
- From the investigation results, prospective chemical admixtures were selected which had varied chemical compositions that played a role in water reduction (Table 1).
- The name was selected considering experience or track record of past use or information obtained from the manufacturer.

Туре	Molecular structure (Schematic diagram)	Effect in the presence of cement	Effect in the presence of AAM	Name
Lignin sulfonic acid based	A Contraction	Adheres to cement particles, because of which the particles get negatively charged and an electrostatic repulsion effect is achieved. Advantage: Cheap Disadvantage: Cement hydration is delayed. Water reduction effect is low.	There are only a few examples of research using AAM. Some of the research reports indicate water reduction effect but the views are not consistent. Tolerant to highly alkaline substances (expected).	Pearllex DP Pearllex NP
Naphthalene sulfonic acid based	H-8-0H-8-0H-800	Adheres to cement particles, because of which the particles get negatively charged and an electrostatic repulsion effect is achieved. Advantage: It is difficult to delay hydration reaction Disadvantage: Fluidity reduces easily with to passage of time.	There are comparatively many examples of research using AAM. Some of the research reports indicate water reduction effect but the views are not consistent. Tolerant to highly alkaline substances (expected).	Mighty 100
Melamine sulfonic acid based		Adheres to cement particles, because of which the particles get negatively charged and an electrostatic repulsion effect is achieved. Advantage: It is difficult to delay hydration reaction Disadvantage: Fluidity reduces easily with to passage of time.	There are very few examples of research using AAM. Water reduction effect is unknown. Tolerant to highly alkaline substances (expected).	SikamentFF86
Polycarboxylic acid based	CH, -C, -C, -p, -p, -C, -, -, -, -, -, -, -, -, -, -, -, -, -,	Adhered to cement particles. Charging effect is weak, but steric repulsion effect is achieved for particles belonging to the same side chain. Advantage: Fluidity is easily maintained even with passage of time. Water reduction effect is high. Disadvantage: Expensive. Cement hydration is delayed. *The largest number of use examples are found in the civil engineering and architectural domain.	There are comparatively many examples of research using AAM. Some of the research reports indicate water reduction effect but the views are not consistent. There are concerns about tolerance to highly alkaline substances.	Sikament 1200N

#### Table 1 Selected chemical admixtures



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

i. Study on techniques for verifying the feasibility of low-temperature solidification treatment

- Verification of the effect of fluidity improvement due to chemical admixtures -

- > Verification of the effect of fluidity improvement due to chemical admixtures
  - Tests were conducted using the selected chemical admixtures (Table 1) to find out the kind of impact that the chemical admixture has on physical properties of solidification material (cement, AAM).
  - As there are differences in the usage results of and research on chemical admixtures with respect to cement and AAM, implementation details were established for each solidification material.
    - Cement: As there are ample findings indicating fluidity improvement effect in general, the tests will be conducted focusing on studies related to simulated mixed waste systems (carbonate slurry: CS, iron co-precipitation slurry: IS).
      - Verification of the effect of the selected chemical admixture (base material, simulated mixed waste system)
      - Acquisition of data pertaining to properties of solidified substances in the simulated mixed waste system when the chemical admixture is added (Adjustment of the chemical admixture addition rate and the waste filling rate (Fig. 1))
    - AAM: As there are only a few research examples concerning the addition of chemical admixtures, whether or not the chemical admixture has an effect will be investigated.
      - · Verification of the effect of the selected chemical admixture (base material, simulated mixed waste system)
      - · Investigation of the composition of AAM that has a fluidity improvement effect

woro	Class	Туре	Product name	Manufacturer
were		the tension of the state of the second	Pearllex NP (OPC)	Nippon Paper Industries Co., Ltd.
tures	Chemical	Lignin suifonic acid based	Pearllex DP (AAM)	Nippon Paper Industries Co., Ltd.
ied	admixture	Naphthalene sulfonic acid based	Mighty 100	Kao Chemicals
eu	Melamine sulfonic acid based	SikamentFF86	Sika Japan Ltd.	
		Polycarboxylic acid based	Sikament 1200N	Sika Japan Ltd.
	]			
Pulvulerant b				

 Table 1
 Selected chemical admixtures

Besides acquisition of data on each solidification material, the
changes in properties of chemical admixtures due to radiation were
investigated as well.

- Investigation of the impact of radiation on chemical admixtures
  - Investigation of the change in properties of chemical admixtures due to irradiation
  - Investigation of the irradiation characteristics of solidified cement containing chemical admixtures



Fig. 1 Definition of waste filling rate



- (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
  - i. Study on techniques for verifying the feasibility of low-temperature solidification treatment_
    - Verification of the effect of fluidity improvement due to chemical admixtures (Cement) -
- Impact of chemical admixtures on cement
- A paste of base material and simulated mixed waste was made and fluidity data was acquired through a small flow test (Fig. 1) using a small ring (φ25 × 50 mm) to understand the fluidity improvement effect of the selected chemical admixtures.
- There were 5 types of simulated waste, namely, light calcium carbonate, silica powder, heavy calcium carbonate, simulated carbonate slurry, and simulated iron co-precipitation slurry.
- The obtained small flow values were compiled in terms of the rate of increase in fluidity as against systems that do not contain chemical admixtures (Table 1).



Fig. 1 Small flow test

Туре		Evaluation based on the rate of increase in small flow values*1					
		Base material	se material Simulated mixed waste system				
		Cement	Light calcium carbonate	Silica powder	Heavy calcium carbonate	Simulated carbonate slurry	Simulated iron co-precipitation slurry
Lignin sulfonic acid based	Pearllex NP	O	0	0	0	0	0
Naphthalene sulfonic acid based	Mighty 100	Ø	0	0	0	-	0
Melamine sulfonic acid based	Sikament FF86	O	0	0	0	—	0
Polycarboxylic acid based	Sikament 1200N	O	Ø	O	Ø	_	Ø

Table 1 Results of verifying the fluidity of the selected chemical admixtures

*1 Explanation of the evaluation symbols O: Large improvement (+30% or more), O: Improvement (0 to +30%), -: No improvement

✓ All chemical admixtures showed fluidity improvement effect except in simulated carbonate slurry.

- ✓ Only lignin sulfonic acid based admixture showed an improvement effect in simulated carbonate slurry.
- All chemical admixtures showed fluidity improvement effect in simulated iron co-precipitation slurry, but the effect was particularly high with polycarboxylic acid based admixture.
- ✓ In the case of cement it was found that there is a chemical admixture having a fluidity improvement effect with respect to all simulated waste.





(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

i. Study on techniques for verifying the feasibility of low-temperature solidification treatment

- Acquisition of data on properties of solidified substances when chemical admixtures are added (Cement) -
- Data on properties of solidified substances containing chemical admixtures was acquired for simulated carbonate slurry and simulated iron coprecipitation slurry which are simulated secondary wastes generated from contaminated water treatment.
- Test method
- The chemical admixture that showed the maximum fluidity improvement effect on each simulated slurry was used based on the results of verifying the fluidity improvement effect of chemical admixtures.
  - Simulated carbonate slurry: Lignin sulfonic acid based admixture (Pearllex NP)
  - Simulated iron co-precipitation slurry: Polycarboxylic acid based admixture (Sikament 1200N)
- Data was acquired by changing the waste filling rate (Table 2) based on the evaluation criteria (Table 1) and the method for testing properties of the solidified substances.
- The amount of chemical admixture to be added was indicated as weight % of the effective amount of admixture for the weight of the pulverulent material (cement + simulated waste) (P).

	Fluidity	Condensation	Compressive strength	Material segregation (Cement)
Test method	Small flow test Small ring of Φ25 x 50 [mm]	Condensation test using the vicat needle	Measurement of compressive strength Small solidified body of 2 x 2 x 3 [cm]	Verification of the number of days until bleeding disappears
Evaluation criteria	Small flow test = 66 to 81 [mm] (Cement) = Unit 110 to 130[mm] (AAM)	No flash setting, and solidifies without bleeding within 24 hours	1.47 [N/mm ² ] or more (Material age 28 days)	In principle disappears within 24 hours Not observed after 3 days in the case of waste for which it is difficult to attain the criteria.

#### Table 1 Method of testing the properties of solidified substances and the evaluation criteria

#### Table 2 Composition of the mixture

	Ratio of water and powder (W/P) [mass%]	Waste filling rate [mass %]
Simulated carbonate slurry mixture system	53.8	30 to 40
Simulated iron co-precipitation slurry mixture system	43.0	20 to 50



(a) Establishment of selecting advance treatment [1] Low-temperature treatment technique

- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
- Acquisition of data on properties of solidified substances when chemical admixtures are added: Simulated carbonate slurry (Cement) -
  - Properties of the simulated mixed waste system containing chemical admixtures
  - Data was acquired on properties of solidified substances when chemical admixture (lignin sulfonic acid based) that shows fluidity improvement effect in simulated carbonate slurry (CS) mixture system is added (Fig. 1, Fig. 2 and Fig. 3).





✓ Even if the filling rate is increased, by adjusting the amount of chemical admixture added, fluidity can be secured.





Fig. 2 Relation between filling rate and strength

- ✓ As filling rate increases the strength decreases
- ✓ Waste can be mixed up to a 40mass% filling rate.

#### Condensation



Effective addition rate (Px%)

Fig. 3 Relation between the rate of addition of chemical admixtures and number of days required for hardening

✓ The hardening is completed within 24 hours at all levels with the considered composition of up to 40mass% filling rate and about Px4% addition rate of chemical admixtures.

#### Ease of material segregation

- ✓ Bleeding water disappears within 24 hours at all levels with the considered composition of up to 40mass% filling rate and about Px4% addition rate of chemical admixtures.
- The filling rate (30mass%) of simulated carbonate slurry into solidified cement in the typical composition determined last year can be improved to up to 40mass% in the scope of investigation this time, by addition of chemical admixtures.



Fluidity

- Properties of the simulated mixed waste system containing chemical admixtures
  - Data was acquired on properties of solidified substances when chemical admixture (polycarboxylic acid based: Sikament 1200N) that shows fluidity improvement effect in simulated iron co-precipitation slurry (IS) mixture system is added.





✓ Even if the filling rate is increased, by adjusting the amount of chemical admixture added, fluidity can be secured.

#### Ease of material segregation





Fig. 2 Relation between filling rate and strength

- ✓ As filling rate increases, strength decreases.
- ✓ The evaluation criteria of 1.47N/mm² is met even with a filling rate of 50mass%.

#### Condensation



Fig. 3 Relation between the rate of addition of chemical admixtures and number of days required for hardening

- ✓ With the increase in filling rate the number of days required for hardening increases as well.
- ✓ There is a possibility of being able to reduce the number of days required for hardening by using hardening accelerators.
- $\checkmark$  Bleeding disappeared within 24 hours at all levels even with a filling rate of 50mass%.
- ✓ In the case of simulated iron co-precipitation slurry mixture system, even though fluidity can be secured by adding chemical admixtures, as condensation (number of days required for hardening) gets delayed, the waste filling rate is not expected to improve.



Fluidity

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

i. Study on techniques for verifying the feasibility of low-temperature solidification treatment

- Verification of the effect of fluidity improvement due to chemical admixtures (AAM) -

- Impact of chemical admixtures on AAM
- A paste of base material (M: only Metakaolin, MB40: M + slag 40mass%) and a system containing simulated waste was made and fluidity data was acquired through a small flow test using a small ring (φ25x50mm) to understand the fluidity improvement effect of the selected chemical admixtures.
- There were 2 types of simulated wastes, namely, simulated carbonate slurry, and simulated iron co-precipitation slurry.
- The obtained small flow values are shown in Fig. 1, and their compilation in terms of the rate of increase in fluidity as against systems that do not include chemical admixtures is shown in Table 1.



Fig. 1 Small flow values of base materials in which chemical admixtures were added (Top: AAM (M), Bottom: AAM (MB40)

Precipitation slurry mixture system.
 ✓ None of the chemical admixtures showed an effect throughout all cases under AAM, but it was found

✓ None of the chemical admixtures showed a fluidity improvement effect in simulated iron co-

that the effect is seen depending on the waste mixed.

admixtures except polycarboxylic acid based admixture.

- (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
  - i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
    - Verification of the effect of fluidity improvement due to chemical admixtures (AAM) -
- Background of verification
  - In the case of AAM, the chemical admixture that didn't have an effect on the base material, showed different tendencies such as some fluidity improvement effect in the simulated mixed waste system.
  - It is believed that this difference in fluidity improvement effect depends on the composition of the solution, and in the case of AAM, the composition of the alkaline solution is expected to have an impact on the effect of the chemical admixture.
  - Hence, the effect of chemical admixture when the concentration of Na and Si in the alkaline mixing water is changed in a mixed waste system was investigated.
- Testing conditions
  - AAM (M+CS (filling rate 30%)), AAM (M+IS (filling rate 20%)), AAM (MB40+CS (filling rate 30%)), AAM (MB40+IS (filling rate 20%))
  - Composition of the alkaline mixing water: Concentration of Na and Si in the solution is changed
  - The changes in small flow values when the composition of the alkaline solution is changed are shown in Fig. 1.



- Test results
  - ✓ Water reduction effect largely changed depending on the composition of mixing water in the case of all chemical admixtures.
  - ✓ Polycarboxylic acid based admixtures did not have a water reduction effect in the scope of any of the combinations.
  - It was found that with a decrease in the concentration of Na in the alkaline mixing water, water reduction effect increases. In particular, it was found that naphthalene sulfonic acid based admixtures and melamine sulfonic acid based admixtures tend to have a larger water reduction effect.
  - ✓ When chemical admixtures are added to AAM, it is necessary to adjust the composition once again for each chemical admixture used.



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
- Study on the scope of application of solidification treatment when chemical admixtures are added (Cement) -
- Data acquired on the properties of solidified cement mixed in simulated waste in which chemical admixtures are added, was reflected in the triangular diagram presented in FY2020. (Portion enclosed by the O was added).
- Solidification characteristics of solidified cement in the simulated carbonate slurry + lignin sulfonic acid based admixture (Pearllex NP)



Fig. 1 Scope of feasibility of solidification treatment of solidified cement in the simulated carbonate slurry when chemical admixture (Pearllex NP) is added

✓ The scope of feasibility of solidification of the carbonate slurry was increased by adding the chemical admixture.



- (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
- Study on the scope of application of solidification treatment when chemical admixtures are added (Cement) -
- Solidification characteristics of solidified cement in simulated iron co-precipitation slurry + polycarboxylic acid based admixture (Sikament 1200N)



Fig. 1 Scope of feasibility of solidification treatment of solidified cement in the simulated iron coprecipitation slurry when chemical admixture (Sikament 1200N) is added

✓ When fluidity improves, condensation gets delayed. Hence ultimately the scope of application did not increase.

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
  - Irradiation test: Irradiation characteristics of the chemical admixtures -

> Investigation of the change in properties of chemical admixtures due to irradiation

- Changes in the properties of chemical components that show water reduction effect of the chemical admixture, due to radiation, were confirmed through γ irradiation test.
- Samples of 5 types of chemical admixtures indicated in Table 1 were irradiated, they were diluted so that the effective amount of admixture was 20mass% and the moisture content was about 80mass%, and placed in a glass vial.
- γ irradiation was carried out under the conditions mentioned in Table 2.
- The samples before and after irradiation were compared by means of FT-IR analysis (Attenuated Total Reflection method) (Fig. 1).
- After irradiation, the hydrogen gas inside the container was analyzed, and the G value was calculated (Fig. 2).

#### Table 1 Material to be irradiated

0.6

#### **Chemical admixture** Form of sample Test details y irradiation of chemical admixtures Powder (aqueous 1 Lignin sulfonic acid based (Pearllex NP) Takasaki Advanced Radiation Research solution) Testing site Institute Powder (aqueous 2 Lignin sulfonic acid based (Pearllex DP) solution) Radiation source Co-60 Powder (aqueous Naphthalene sulfonic acid based (Mighty 100) solution) Approx. 3 [kGy/h] Dose rate Melamine sulfonic acid based (Sikament FF86) Liquid Integral dose 3 [kGy] 5 Polycarboxylic acid based (Sikament 1200N) Liquid (Planned)



4000 3600 3200 2800 2400 2000 1600 1200 800 400 Wavenumber (cm⁻¹)

Pearllex NP 3kGv



 During FT-IR, no differences were seen in the shape of peaks before and after irradiation, in the case of all chemical admixtures.

Table 2 Irradiation conditions

- ✓ Except Sikament 1200N which is a polycarboxylic acid based admixture, the G value of all chemical admixtures was lower than the G value of water (0.45).
- ✓ The G value of Sikament 1200N was high, but it is presumed based on the results of FT-IR that this was not due to breakdown of admixtures.
- ✓ Changes in properties of chemical admixtures due to radiation were not found.

Fig. 1 Results of FT- IR analysis of chemical admixtures





(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
- Irradiation test: Irradiation characteristics of the solidified cement containing chemical admixtures -
- Irradiation characteristics of solidified cement containing chemical admixtures
  - Investigation was carried out to check whether the properties of the chemical admixtures in solidified substances change due to the impact of ٠ radiation leading to leaching of organic components.
  - Solidified cement indicated in Table 1 was used as the sample for irradiation. ٠
  - y irradiation was carried out under the conditions mentioned in Table 2.
  - Leaching test was conducted for the samples before and after irradiation. Further, the changes in organic components in the solid phase after the leaching test were verified by CHN analysis.



#### Table 1 Material to be irradiated

Results of the CHN analysis of the solid phase are indicated in Fig. 1.



Fig. 1 Results of the CHN analysis of the solid phase after the leaching test

✓ During the leaching test, there was no leaching of organic matter before and after irradiation.

Integral dose

(Planned)

- ✓ Data showing an increase in the proportion of carbon in all samples after irradiation was obtained, but the leaching characteristics were not affected.
- ✓ The possibility of uptake of carbon from outside (for example the gaseous component in the container, etc.) can be considered.
- ✓ Leaching of the chemical admixture component due to radiation could not be verified, and hence it is believed that it had only a small effect on leaching characteristics.



3 [kGy]



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
  - Determination of fluidity improvement effect based on primary screening -
- > Determination of fluidity improvement effect based on primary screening
  - If the effect of chemical admixtures can be determined during primary screening (determined using a combination of waste and inspection liquid), the effect can be determined more easily than performing secondary screening (determined using a paste made by mixing waste and solidification material).
  - It was verified that the effect of the chemical admixture can be determined during primary screening, by measuring the dispersibility of waste using inspection liquid (cement equilibrium water, 40g) in which chemical admixture (0.1g) is added.
  - The results of primary screening (verification of dispersibility of waste and inspection liquid with a centrifuge tube) (Table 1)) and the results of secondary screening (fluidity verification test using the paste) (Table 2) were compared.

#### **D** Cement

Type of simulated	Results of primary screening (solidification feasibility evaluation) (Evaluation based on the state of sedimentation of particles*1)				ition) s* ¹ )	
Inspection liquid (Basis of comparison Cement equilibrium water)	Cement for research	Light calcium carbonate	Silica powder	Heavy calcium carbonate	Simulated carbonate slurry	Simulated Iron coprecipitati on slurry
Cement equilibrium water + Pearllex NP	0	0	0	0	Ι	0
Cement equilibrium water + Sikament FF86	0	0	0	Ø	I	0
Cement equilibrium water +Mighty 100	0	0	O	Ø	-	0
Cement equilibrium water + Sikament 1200N	0	0	Ø	Ø	-	0

Туре	Evaluation based on the rate of increase in small flow values *2					
Type of simulated waste Type of chemical admixture	Cement	Light calcium carbonate	Silica powder	Heavy calcium carbonate	Simulated carbonate slurry	Simulated Iron coprecipitati on slurry
Pearllex NP	0	0	0	0	0	0
Sikament FF86	0	0	0	0	_	0
Mighty 100	0	0	0	0	-	0
Sikament 1200N	0	0	0	0	-	0

Table 2 Results of fluidity verification by means of secondary screening method

*2 Explanation of the evaluation symbols **():** Large improvement (+30% or more), **():** Improvement (0 to +30%), -: No improvement

*1 Explanation of the evaluation symbols **()**: **Dispersibility improvement (All particles)**, **()**: **Dispersibility improvement (Only fine particles)**, **(–)**: **No change** 

- The dispersibility of particles improved due to addition of chemical admixture with cement alone and simulated waste (total 5 types) except simulated carbonate slurry (Table 1).
- ✓ The fluidity improvement effect was consistent for all simulated waste except simulated carbonate slurry (Table 2).

- ✓ The trend seen in the primary screening (change in dispersibility) and the trend seen in secondary screening (change in fluidity) were not consistent.
- In the case of cement, the results are consistent for all simulated waste other than simulated carbonate slurry, and hence the effect can be determined by means of the screening method even for systems containing chemical admixtures.





- i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
- Study on addition of inspection items and the method of acquiring data on solidification characteristics: Condensation verification test
  - > Study of the method of verifying condensation of solidified substances by means of the untrasonic method
  - Condensation, which is an evaluation item of solidification characteristics, is supposed to be tested and evaluated using the vicat needle in secondary screening, but from the perspective of reducing exposure, etc. it is desirable to complete the operation in a sealed system such as the centrifuge tube.
  - The method of obtaining condensation from the outer surface of the centrifuge tube by means of ultrasonic waves was studied.
  - Cement paste (Table 1) was mixed, and the Ultrasonic Propagation Time (UPT) from right after it was filled in a centrifuge tube up to 24 hours was obtained.



Fig. 1 Relation between elapsed time and the ultrasonic propagation time

Table 1 Compounding conditions				
Sample solidified substance	Filling factor [mass%]	W/P [mass%]		
Cement	-	30		
Cement + CS	30	53.8		
Cement + IS	20	48		



Fig. 2 Image illustrating the relation between the change in characteristics at the time of cement hardening and the acquired data

- ✓ All samples indicated that there was an inflection point at which the ultrasonic propagation time largely decreases (Fig. 1).
- As the propagation velocity of ultrasonic waves changes in the paste and in solidified substances, it is believed that it was possible to ascertain how the paste changes to solidified substance with passage of time.
- It is believed that the start and end time of condensation obtained using the vicat needle, and the condition of the solidification material that indicates the propagation time obtained by means of the ultrasonic method is as shown in Figure 2.
- It is thus implied that the time required for the paste to solidify can be obtained without making contact with the sample by inserting the vicat needle.



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

i. Study on techniques for verifying the feasibility of low-temperature solidification treatment

- Study on addition of inspection items and the method of acquiring data on solidification characteristics: American Nuclear Society leaching test -

- > Study of the method of evaluating leaching of solidified substances
- While evaluating solidification characteristics since understanding leaching is important as well besides the current evaluation criteria, the method for easily evaluating leaching was studied.
- From the perspective of reducing exposure, etc. it is desirable to complete the operation in a sealed system such as the centrifuge tube.
- The applicability of the centrifuge tube method was studied by conducting a leaching test using a 50ml centrifuge tube (hereinafter, centrifuge tube method), and the results were compared with the results of the ANS leaching test (hereinafter, ANS method) (Table 1).
- About 15 cc sample was filled in a centrifuge tube and hardened, and it was sealed and cured up to a material age of 28 days. Thereafter, 30cc ultrapure water was poured on the sample in the centrifuge tube, it was allowed to stand, and then solution was collected and refilled. The element concentration of the collected solution was measured.



Fig. 1 Comparison of the leaching rate obtained by means of the ANS method and the centrifuge tube method Cement + CS (Left: Real axis, right: Logarithmic axis)

Fig. 2 Comparison of the leaching rate obtained by means of the ANS method and the centrifuge tube method AAM + CS) (Left: Real axis, right: Logarithmic axis)

- The easily leachable and not so easily leachable elements obtained as a result of the centrifuge tube method for both the cement system as well as the AAM system were consistent with those obtained by means of the ANS method (Fig. 1, Fig. 2).
- The relationship of magnitude of the leaching rate for each element was consistent in the cement system. In the AAM system, the relationship of magnitude of the leaching rate for the not so easily leachable elements was not consistent.
- ✓ The centrifuge tube method is likely to be applicable for determining the ease of leaching of the elements.
- If the relationship of the ANS method and the wet surface area can be controlled, there is the possibility of being able to evaluate leaching (obtain the L value) using the centrifuge tube.
- It thus implied that there is the possibility of being able to ascertain leaching behavior of elements by means of a small scale leaching test (centrifuge tube method).



 Table 1 Comparison of the testing conditions of the ANS leaching test and the simple test

Leaching period

[h]

2, 7, 24, 48,

72, 96, 120

2, 7, 24, 48,

72. 96. 120

Quantity of

solution collected

Around 1.2 [L] /

round

30 [cc] / round

Quantity of solution (cm³)

Sample contact

surface ratio

10

2.62

Testing conditions

ANS leaching test

(Implemented in

FY2020)

Centrifuge tube

method

(Implemented this

vear)

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

i. Study on techniques for verifying the feasibility of low-temperature solidification treatment

- Study of quantification using the image determination test equipment -

- > Study of quantification by means of the image determination test equipment
- In order to establish the determination criteria for the verification technique, each determination criteria is quantified using the image determination equipment (Fig. 1).
- For establishing the determination criteria, data needs to be accumulated on waste indicating both "appropriateness" and "inappropriateness" with respect to the determination criteria.
- So far, it has been found that solidifiable simulated waste can be determined by means of the image determination equipment similar to visual inspection.
- In this study, data was obtained by means of the image determination equipment on the 3 types of simulated waste (Table 1) that were determined as inappropriate as a result of visual determination, from among 12 types of simulated waste studied during the FY2020 investigation.

Shutter box for shielding light





Table 1 3 types of simulated waste determined as inappropriate as a result of visual determination

Simulated waste	Properties determined as "inappropriate"	Inspection liquid
Aluminum powder	Foaming after dripping of inspection liquid	Simulated cement kneading water
Montmorillonite	Swelling after dripping of inspection liquid	Pure water
Magnesium sulfate	Sudden heating after dripping of inspection liquid	Pure water

Fig. 1 Configuration, specifications and external appearance of the image determination test equipment

- > Acquisition of images indicating inappropriate properties
  - ✓ 3 types of simulated wastes were loaded on to the equipment. The obtained images are shown Table 2.



#### Table 2 Images of each simulated waste

- □ Aluminum powder
  - ✓ Foaming was confirmed from visible imaging
- □ Montmorillonite
  - ✓ The state of slight swelling was confirmed from visible imaging
- Magnesium sulfate
  - ✓ The state of heating was confirmed by means of thermal imaging
  - ✓ Sudden heating after dripping of inspection liquid was obtained as numerical values along with passage of time.
- ✓ Inappropriate properties can be determined similar to visual inspection using the image determination equipment.


- (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
  - i. Study on techniques for verifying the feasibility of low-temperature solidification treatment
    - Study of quantification using the image determination test equipment -
- > Study on quantification of inappropriate properties
  - It was verified whether or not inappropriate properties can be quantified with respect to foaming and swelling.
  - It was verified whether the images where foaming can be seen can be adjusted using an analytical software so that selecting the portion with the foaming and obtaining the area of that section can be automated (Fig. 1).



Fig. 1 Perception of foaming by image adjustment (area of the red portion to be calculated)

- The portion with foaming was perceived as smaller than the size of the portion with actual foaming. Hence the area of only the portion with foaming could not be calculated.
- The portion with the shadow of the centrifuge tube, etc. was perceived as an area with the same contrast and thus sections besides the portion with foaming got included as well.
- ✓ In order to automate the process of obtaining the amount of foaming from the image, the study needs to include conditions during shooting as well.
- The determination criteria of heating was established on a trial basis focusing on the values of rise in temperature (Δ Temperature = [Temperature of sample] — [Surrounding temperature])
   Table 1 Maximum Δ temperature of simulated waster
- ✓ Besides simulated waste, the  $\Delta$  temperature of the kneaded cement paste was obtained by means of the image determination equipment, and it was compared with the  $\Delta$  temperature of the simulated waste that was obtained in a similar manner by means of the image determination equipment (Table 1).
- ✓ As long as the range of  $\Delta$  temperature of simulated waste does not exceed the  $\Delta$  temperature (1.2°C) of the cement base material, it can be determined as solidifiable.
- ✓ The ∆ temperature of simulated iron co-precipitation slurry is 3.4°C which is higher than the cement paste, but it has been confirmed to be solidifiable in the studies conducted up to last year.
- $\checkmark$  The determination criteria of heating is estimated to be between 3.4 and 24.1 °C.
- ✓ In order to derive a clear determination criteria, data needs to be accumulated by means of tests using samples that offer a variety of heat generation.

	Maximum Δ temperature [°C]	Determination of solidification
Cement	1.2	-
Magnesium sulfate	24.1	Inappropriate
Aluminum powder	1.2	Appropriate
Montmorillonite	1.3	Appropriate
Heavy calcium carbonate	1.7	Appropriate
Simulated carbonate slurry	1.3	Appropriate
Simulated iron co- precipitation slurry	3.4	Appropriate





(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique i. Study on techniques for verifying the feasibility of low-temperature solidification treatment

## **Results so far**

- ✓ Chemical admixtures were investigated and 5 types of chemical admixtures that are likely to have an effect on cement and AAM were identified.
- The effects that the identified chemical admixtures have on change in characteristics of solidified substances such as fluidity, condensation, strength and ease of material segregation were verified and data was obtained.
- Cement solidification: Lignin sulfonic acid for simulated carbonate slurry and polycarboxylic acid for simulated iron co-precipitation slurry were selected as the chemical admixtures with a powerful improvement effect. It became possible to improve the maximum filling rate of simulated carbonate slurry from 40 mass% to 45 mass% by adding the chemical admixture.
- AAM solidification: Chemical admixtures did not have an effect on AAM In order to use chemical admixtures for AAM, the composition needs to be adjusted once again.
- ✓ As an improvement in the screening technique, the evaluation technique that enables verification of condensation without making contact with the sample by using a centrifuge tube is expected to have potential. It was found that there is the possibility of being able to ascertain leaching behavior of elements by means of a leaching test using a centrifuge tube.

## Issues, etc.

- The simple measurement method that can be used as a verification technique needs to studied further.
- In order to generalize the verification techniques, verifications assuming various solid wastes need to be carried out.
- In order to develop a technique for quantification of foaming and swelling based on images and to derive a criteria for determining feasibility of solidification, data needs to be accumulated by means of tests using samples that offer a variety of heat generation.



 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique ii. Investigation of the change in properties of solidified substances due to heating, etc.
 Details of implementation -

- Achievements up to FY2020
- Investigations and tests related to the change in properties of solidified substances due to heating and drying were conducted, that made it clear that heating and drying contributed to the decrease in the strength of solidified substances.
- The following issue was identified with respect to on-site application and further refinement of data.
  - Acquisition of data on change in properties of solidified substances in actual storage environment, and evaluation of the attained temperature.
- Goal

To conduct tests simulating heating, drying, etc. assuming the temperature and humidity in actual environment for investigating the impact that storage environment has on the change in properties of solidified substances, in order to investigate the impact on solidified substances during the period from treatment to storage.

## **Details of implementation**

Acquisition of data on cyclic conditions, etc. simulating the storage environment

- Setting of conditions to which the solidified substances are exposed assuming the temperature and humidity in the storage environment
- · Evaluation of change in properties of solidified substances after introducing cyclic conditions

### Indicators for the achievements of goal

Data concerning the constituent phase of solidified material under storage environmental (cyclic) conditions are studied and investigation results are presented.





(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
 ii. Investigation of the change in properties of solidified substances due to heating, etc.
 Test conditions 1. Item to be tested and method of exposure -

- Test conditions
  - Item to be tested: Prepared solidified substances (Table 1)
  - Method of preparing the solidified substances: A 1 cm space was created at the top of a cylindrical mold (φ5 x 10cm) (Fig. 1)

substances

• Exposure testing method: Testing material of material age 7 days (φ5 x 9 cm) was exposed inside temperature and humidity control equipment that was set to various exposure conditions (Fig. 2).

Sample n	ame Label*	Waste filling rate	Water / Solid ratio	Si / Al ratio	Na / Si ratio
		[mass%]	[mass%]	[mol/mol]	[mol/mol]
	Base material	-	45	-	-
Cement	Cement + CS	30	54	-	-
	Cement + IS	20	48	-	-
	Base material	-	150	1.8	1.2
AAM (M)	M+CS	30	92	1.8	1.2
(,/	M+IS	20	100	1.9	1.0
	Base material	-	100	1.8	0.8
AAM (MB40)	MB40+CS	30	80	1.8	0.8
(	MB40+IS	20	87	1.9	0.95

*Explanation of abbreviations CS: Simulated carbonate slurry IS: Simulated iron co-precipitation slurry Cement: Ordinary Portland Cement (OPC) AAM(M): Metakaolin 100mass% AAM(MB40): Metakaolin 60mass% + blast furnace slag 40mass%



Fig. 1 Solidified substance placed in the mold *By using a translucent mold, the status inside the mold (amount of bleeding, etc.) can be roughly observed without removing the contents of the mold.



Fig. 2 The inside of the temperature and humidity control equipment





- ii. Investigation of the change in properties of solidified substances due to heating, etc.
- Test conditions 2. Setting the cyclic conditions simulating the storage environment -
- > Setting of cyclic conditions simulating the storage environment

[Environmental load conditions to which solidified substances are expected to get exposed during storage]

- Stored in sealed solidification containers (No contact with outside air → conditions not conducive to drying)
- Cyclic changes in temperature during the day and at night and changes in temperature due to the seasons occur in the storage environment (indoors)
- Range of change in temperature: about -5 to 45°C

Set up by adding the increase in temperature in a system wherein multiple drums are arranged (Cs-137, 1E+9 Bq/cm³, natural convection current  $20^{\circ}C \rightarrow 27^{\circ}C$ ) to the Namie town AMeDAS Olympic data (Summer: 37.3°C in 2018, Winter: -12.4°C in 2014).



Cyclic conditions: In order to accelerate the change in solidified substances, a cycle of maximum temperature  $45^{\circ}C \rightarrow Minimum$  temperature  $-5^{\circ}C \rightarrow Maximum$  temperature  $45^{\circ}C$  was set as 1 cycle of 6 hours. (4 cycles in 24 hours, 20 minutes each of rise and fall in temperature, maximum and minimum temperature maintained for 160 minutes) (Fig. 1, Table 1)

- Method of evaluating the change in properties of solidified substances
  - The phenomena that are likely to occur including results of studies until FY2020 were investigated, and property data (Table 2) and sites to be analyzed (Fig. 2) were determined.

#### Table 2Property data to be evaluated

No.	Property	Analysis methods	Analysis sites
1	Compressive strength	Ultrasonic waves (Non-destructive)	-
2	Density	Non-destructive (Calculated based on dimensions and weight)	-
3	Amount of moisture (Free water)	Coarse crushing $\Rightarrow$ Method of drying at 105°C	Top and bottom
4	Crystalline structure (XRD)	Pulverizing $\Rightarrow$ XRD	Top and bottom
5	Pore size distribution	Coarse crushing ⇒ Pretreatment ⇒ MIP	Top and bottom



Fig. 1 Set temperature conditions to which solidified substances get exposed

Table 1 Temperature conditions set for 1 cycle (6h)

Time [min]	Temperature set in the constant temperature reservoir
10	20°C→45°C
160	45°C
20	45°C→-5°C
160	-5°C
10	-5°C→20°C



Fig. 2 Image of the sites to be analyzed on the sample

 In order to verify the internal temperature of the solidified substances during exposure, a thermocouple was embedded into 1 solidified substance sample of each composition.



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 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique ii. Investigation of the change in properties of solidified substances due to heating, etc.
 Change in external appearance 1. Photographs -

> Change in external appearance after exposure (Table 1)

Table 1 Photograph of the solidified substances after exposure to cyclic conditions

			Cement	AAM	AAM	AAM	AAM	AAM	AAM
	Cement	Cement + CS	+IS	(M)	(M+CS)	(M+IS)	(MB40)	(MB40+CS)	(MB40+IS)
100 th cycle				Hat	6				HB40-TE
200 th cycle	23456789			10 2 4 5 6 7 8 9 II	1 634 6 75 C 19 5 10 C 2 Kint	BRESTERNE CAN			
280 th cycle									

✓ 100th cycle: Cracks developed at the top and bottom in all test specimens of only AAM (M + CS). Other solidified substances had no abnormalities.

✓ 200th cycle: Cracks developed at the top and bottom in all test specimens of only AAM (M + CS). Other solidified substances had no abnormalities.

✓ 280th cycle: Multiple cracks developed in all test specimens of only AAM (M + CS). Other solidified substances had no abnormalities.

✓ (Summary) Only the AAM (M + CS) solidified substance developed cracks due to cyclic temperature load.



- (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
  - ii. Investigation of the change in properties of solidified substances due to heating, etc.
  - External appearance 2. Investigation of the solidified substances that were damaged
- Investigation related to solidified substances in which damage was seen
  - Data from the thermocouple embedded into 1 solidified substance sample of each composition was confirmed for verifying internal temperature of the solidified substances during exposure (Fig. 1, Fig. 2).



 $\checkmark$ 

It was found that the internal  $\checkmark$ temperature of the solidified substances was different only in the case of the AAM (M + CS) solidified substances that developed cracks.

AAM (M + CS) solidified substance of the same composition was additionally prepared for cause investigation, and temperature load test was conducted under cyclic conditions (Fig. 3).



Fig. 3 Changes in the internal temperature of AAM (M + CS) solidified substances

(The portion that was initially prepared and the portion that was prepared additionally)

- The internal temperature of the additionally prepared AAM (M + CS) solidified substance  $\checkmark$ showed a similar tendency. Thus it was verified that this tendency is peculiar to AAM (M + CS) solidified substance.
- This implies that the occurrence of the phenomenon of intermittent change in the internal  $\checkmark$ temperature during rise in temperature and during fall in temperature is associated with changes in thermal conductivity and thermal capacity of the solidified substance caused by temperature, and absorption and generation of heat.
- Since a tendency of internal temperature that is different from that of other solidified  $\checkmark$ substances was verified in the first cycle of the cyclic conditions, it is presumed that the cracks developed in the first cycle.
  - The cause for the correlation between this peculiar tendency of internal temperature and the cracks needs to be studied in detail in the future.

ii. Investigation of the change in properties of solidified substances due to heating, etc.

- Test results 1. (1) Compressive strength, (2) Density, (3) Amount of free water -





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(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique ii. Investigation of the change in properties of solidified substances due to heating, etc.

- Test results 2. ④ Crystalline structure (XRD)

(4) Change in the crystalline structure of solidified substances due to cyclic temperature load

• As there was no difference in the top and bottom portion of the solidified substance, the results of

the bottom portion were used for comparison (Fig. 1, Table 1).



				e analysis re	ouno
Cycle r	number	0	100	200	280
	Base material	-	-	-	-
Cement	CS	-	-	-	-
	IS	-	-	-	-
	Base material	-	-	✓ Formed Faujasite	✓ Increase in amount formed Faujasite
AAM (M)	CS	-	-	✓ Formed Faujasite	✓ Increase in amount formed Faujasite
	IS	-	-	✓ Formed Faujasite	✓ Increase in amount formed Faujasite
	Base material	-	-	-	-
AAM (MB40)	CS	-	-	✓ Formed Faujasite	✓ Formed Faujasite
	IS	-	-	✓ Formed Faujasite	✓ Formed Faujasite

Table 1 Crystalline structure analysis results

(The name of the mineral listed represents the mineral facies that is formed)

Change in constituent phase —: Absent, ✓: Present

✓ Cement solidified substance: There was no change in mineral facies.

Legend

- ✓ AAM (M) solidified substance: With the increase in the number of cycles, there was an increase in the quantity of Faujasite formed in the base material and CS, IS mixed solidified substances. (Faujasite is a type of zeolite.)
- ✓ AAM (MB40) solidified substance: CS and IS mixed solidified substances formed Faujisite from 200th cycle onwards.

Fig. 1 Crystalline structure analysis results (AAM(M), AAM(MB40))



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique ii. Investigation of the change in properties of solidified substances due to heating, etc.

 5 Pore size distribution due to cyclic temperature load
 6 Pore size distribution due to cyclic temperature load As there was no difference in the top and bottom part of the solidified substances, the results of the top part were used for comparison (Fig. 1).

*As the sample taken after subjecting to cyclic load broke down from the time it was removed from the mold, it was not able to be measured.



: Along with the increase in number of cycles, the pore size became compact, but it is believed that this was because of crystal growth due to the progress in hydration reaction. Hence it is presumed that this was not an impact of the cyclic conditions.

: No change in base material. The CS mixed sample was unable to be measured as it had broken down. The pore size of the IS mixed sample increased from 0th → 100th cycle

✓ AAM(MB40) : No change in base material. The porosity of the CS mixed sample somewhat increased and the pore size became slightly larger as well. The pore size of the IS mixed

✓ AAM (M)

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- ii. Investigation of the change in properties of solidified substances due to heating, etc.
- Test results_4. At a glance (Changes observed due to the load of cyclic conditions) -
- Each analysis was conducted 4 times at 0th, 100th, 200th, and 280th cycle. The changes seen between 2 milestone cycles are listed in the table below.

Cycle nu	mber	Change	es between $0^{th} \rightarrow 100$	0 th cycle	Changes	between $100^{th} \rightarrow 2$	00 th cycle	Changes	between 200 th $\rightarrow$ 2	30 th cycle
		Cracks	XRD	MIP	Cracks	XRD	MIP	Cracks	XRD	MIP
	Base material	-	_	✓ Compactness	-	_	✓ Compactness	_	_	_
Cement based	CS	-	-	✓ Compactness	-	_	✓ Compactness	-	-	-
	IS	—	_	✓ Compactness	-	_	✓ Compactness	—	-	_
	Base material	-	-	-	-	✓ Formed	-	-	✓ Increase in amount formed	✓ Increase in size
AAM (M) based	CS	H	-	-		✓ Formed	-		✓ Increase in amount formed	-
	IS	-	-	✓ Increase in size	-	✓ Formed	✓ Compactness	-	✓ Increase in amount formed	✓ Compactness
	Base material	_	_	_	_	_	_	—	_	_
AAM (MB40) based	CS	-	-	✓ Increase in size	-	✓ Formed	✓ Increase in size	-	✓ Formed	✓ Compactness
	IS	-	_	✓ Increase in size	-	✓ Formed	✓ Compactness	-	✓ Formed	_
			Legend	Cracks -	–: Absent, Pho	tos: present	Change in cons Char	stituent phase ( nge in porosity	XRD) —: Abse (MIP) —: Abse	nt, ✓: Present nt, ✓: Present

- ✓ There was no impact of cyclic conditions on cement solidified substances.
- ✓ Cracks developed or there were changes in the constituent phase of AAM (M) solidified substances due to load of cyclic conditions.
- ✓ There were changes in the constituent phase of AAM (MB40) solidified substances due to load of cyclic conditions.

 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique ii. Investigation of the change in properties of solidified substances due to heating, etc.
 Summary -

## **Results so far**

- ✓ Tests were conducted to investigate the changes in properties of solidified substances due to change in temperature within the range of -5°C to 45°C that is determined by considering the amount of heat generated by solidified substances in addition to the maximum and minimum temperatures in Namie town.
- Results of observing the changes in properties of solidified substances exposed to cyclic conditions simulating storage environment are given below.
- There were no cracks or changes in mineral facies in the cement solidification substances.
   Hence it was found that the properties do not change even in storage environment.
- It was found that cracks are likely to develop in AAM (M) solidified substances depending on the type of waste mixed.
- Although cracks were not seen in the AAM (MB40) solidified substances, as change in the constituent phase (change from amorphous to mineral facies) was observed, it was found that the solidification performance such as leaching characteristics of waste, etc. is likely to change (deteriorate).

### Issues, etc.

✓ As the solidification performance of AAM (M) and AAM (MB40) solidified substances is likely to deteriorate due to the impact of storage environment depending on their combination with waste, methods for checking the combination with waste in advance are necessary.



# (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances

- As the performance of solidified substances is affected by temperature in the case of low-temperature treatment technology (cement, AAM), the upper temperature limit depends on the material.
- As heat transfer depends on the system, the limit value of waste filling rate is expected to change depending on the shape and size of the container.



By analyzing the maximum temperature and clarifying the changes due to shape and dimensions, the solidifiable inventory limit value studied based on decay heat is acquired as data.

- Achievements up to FY2020
- Heat generation by the inventory contained in the system in which stand-alone solidified substances and cylindrical solidified substances are arranged in an accumulated state was analyzed with the shape of the container, solidification material and radioactive nuclides as parameters, and a relational expression for easily estimating the temperature of the solidified substance was derived.
- The following issue was identified with respect to on-site application and further refinement of data.
  - ✓ Increasing knowledge pertaining to accumulation type storage form (square containers) and deriving a formula for evaluating the highest temperature attained taking that knowledge into consideration.



- Goal
  - The temperature of the solidified substance when radioactive waste is solidified will be analyzed, and the relationship between nuclides such as Cs, etc. contained in the solidified substance and the temperature of the solidified substance will be evaluated.
  - The inventory that can be contained while maintaining integrity of the solidified substance will be studied based on the evaluation results, thereby contributing to the estimation of heat impact.

#### **Details of implementation**

Investigation of thermal effect when square containers are stored in an accumulated state

• Creating the system model • Setting the analysis conditions • Analyzing thermal effect of Cs and other nuclides

Derivation of the formula for estimating thermal effect

• Derivation of formula for obtaining the temperature and amount of heat generated by containers that are arranged in a stand-alone state • Derivation of formula for obtaining the temperature and amount of heat generated by containers that are arranged in an accumulated state

#### Indicators for the achievements of goal

The temperature attained when square shaped containers are accumulated and stored is evaluated and the method of calculating it is presented.





(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
 iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances
 Analysis of the effect of arranging square containers in an accumulated state on heat generation 1. Model and analysis conditions -

- Creation of accumulation type arrangement model for square containers, and setting of analysis conditions
  - Solidified substance containers were stored in an accumulated state by arranging multiple containers horizontally and vertically, and the increase in temperature due to the thermal effect amongst containers was estimated.
  - A model was created for analyzing the amount of heat generated and the temperature with respect to square containers that have excellent container storage efficiency as compared to drum cans, and which have undergone analysis when arranged in an accumulated state, for on-site application of the analytical data.
  - Analysis conditions were set up assuming secondary waste generated from contaminated water treatment as waste that has undergone cement solidification.
     X1 X2 X3



Fig. 1 Image of accumulation type arrangement

Analysis conditions

Analysis conditions	Details
Container	27 square 1m ³ containers (Thickness 0.2 [cm])
Solidification material	Cement
Solidified substance density[g/cm ³ ]	2.69
Activity concentration [Bq/ cm ³ ]	1.0E+05, 06, 07, 08, 09
Software used	PHITS (Analysis of amount of heat generated) COMSOL (Temperature analysis)



Information on the accumulation type arrangement model for square containers

- Cement with uniform radiation is filled in 1m³ square containers up to 80 % of their height and 27 such containers were accumulated.
- The containers were stacked so that the length, breadth and height was 3 x 3 x 3 containers such that it was possible to observe the thermal impact of the surrounding on the container at the center. (Fig. 1)
- The portion with solidified substance in a single container was divided into 108 cells of length breadth and height of 6 x 6 x 3 (Fig. 2) (For the sake of convenience while setting up the software used for analyzing the amount of heat generated, the division of the containers in the X3 column was roughly set up. (Fig. 3))
- An accumulated type arrangement model with 1m³ square containers which have a good storage efficiency was created for on-site applicability of the analytical data.





iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances

- Analysis of the effect of arranging square containers in an accumulated state on heat generation 2. Analysis of amount of heat

denerated -

- Analysis of amount of heat generated when square containers are arranged  $\geq$ in an accumulated state
  - The amount of heat generated in the case of accumulation type arrangement in which heat generation is expected to increase due to radioactive interaction between containers, was analyzed in order to obtain the values used for temperature analysis.
- Analysis conditions ≻
  - · The change in the amount of heat generated by solidified substances due to accumulation is anticipated to be different for different nuclides depending on the type of radiation.
  - Sr-90. Cs-137, Co-60 which emit radiation of different intensities were analyzed.
  - In order to evaluate the impact of accumulation, the amount of heat generated by each stacked container was arranged against the amount of heat generated by a stand-alone container assuming that heat generated by the stand-alone container is 1.00.
- Results of analyzing the amount of heat  $\geq$ generated
  - ✓ In the case of Co-60 and Cs-137 the amount of heat generated increased as compared to the stand-alone type arrangement (Fig. 1, Fig. 2), and the magnitude relationship of the rate of increase was consistent with magnitude relationship of the energy from gamma rays. (Table 1)
  - ✓ The impact of surrounding containers depends on the energy from the emitted gamma rays.
  - ✓ In the case of Sr-90, the amount of heat generated from the stand-alone type arrangement did not increase. (Fig. 3)
  - ✓ In the case of nuclides that do not produce gamma rays, the impact of accumulation was only due to heat transfer.
  - √ It was verified that the increase in amount of heat generated due to accumulation depends on the type of radiation released by the nuclide and the energy of the gamma rays.

1.05	1.07	1.05	1.07	1.09	1.07	1.05	1.06	1.05
1.06	1.08	1.07	1.09	1.11	1.09	1.06	1.08	1.06
1.05	1.07	1.05	1.07	1.09	1.07	1.05	1.07	1.05
	Lower	level		Mid I	evel		Uppe	r level

Fig. 1 Rate of increase in amount of heat generated considering that the amount of heat generated by a 1m³ container is 1.00 (Co-60)

1.02	1.03	1.02	1.03	1.04	1.03	1.02	1.03	1.02
1.03	1.04	1.03	1.04	1.05	1.04	1.03	1.04	1.03
1.02	1.03	1.02	1.03	1.04	1.03	1.02	1.03	1.02
	Lower	level		Mid I	evel		Uppe	r level



1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Lower level

Fig. 3 Rate of increase in amount of heat generated considering that the amount of heat generated by a 1m³ container is 1.00 (Sr-90)





Table 1 Decay energy of nuclides

Gamma rays [MeV]
1.173 1.332
0.662 0.0321 0.0365
None

- (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
- iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances
- Analysis of the effect of arranging square containers in an accumulated state on heat generation 3. Temperature analysis -
- > Temperature analysis when square containers are arranged in an accumulated state
  - The temperature in the case of accumulation type arrangement in which heat generation is expected to increase due to thermal interaction between containers, was analyzed under 2 conditions namely, when there is no convection current and when natural convection is taken into consideration.
  - The initial temperature of the ambient air and the solidified substance was set at 20°C, and the temperature was analyzed using the results of the analysis of amount of heat generated (Cs-137).
  - During actual storage, the heat removal (cooling effect) due to the natural convection current resulting from the difference in the temperature of the solidified substance and the ambient air needs to be considered.
  - First, on the safer side, the condition wherein convection current because of which the temperature result is expected to be on the higher side, is not considered (gravity was not set), was analyzed.
  - Next, the gravity was added to the same model, so that the condition wherein natural convection current because of which a more realistic result is expected, is considered, was analyzed.



Fig. 1 Results of temperature analysis under conditions without convection current



✓ The maximum temperature of solidified substances was 38.70°C under natural convection current conditions.

Fig. 2 Results of temperature analysis under natural convection current conditions

- Estimation of the upper limit value of inventory using the analysis results
  - In order to evaluate the inventory that can be contained while maintaining integrity of the solidified substance, the results of analyzing conditions considering natural convection current were plotted on a graph with the activity concentration of the solidified substance on the x axis and the maximum temperature of the solidified substance on the y axis (Fig. 1), and the inventory limit value was obtained using the approximation formula.



- ✓ The relational expression between activity concentration and maximum temperature of the solidified substance was derived from the plotted analysis results.
  - $y = (-6.35 \times 10^{-19})x^2 + (1.94 \times 10^{-8})x + 20$  (Formula 1)
    - x: Activity concentration [Bq/ cm³]
    - y: Maximum temperature of solidified substance [°C]
- ✓ Since y = 60, the inventory limit value of Cs-137 is estimated to as  $2.22 \times 10^9$  Bq/cm³
- ✓ The inventory limit value of Cs-137 obtained using the relational expression between activity concentration and maximum temperature of solidified substance derived using the temperature analysis results considering natural convection current was 2.22 × 10⁹ Bq/cm³.

Fig. 1 Relation between activity concentration and maximum temperature of the solidified substance



- iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances
  - Estimation of the thermal impact based on analysis results 1. Stand-alone type arrangement 1/2 -
- > Estimation of thermal impact in a stand-alone type arrangement
  - The formula for obtaining temperature under arbitrary conditions was derived from the analysis results to facilitate estimation of the temperature of solidified substances containing radioactive waste.
  - The relational expression between the amount of heat generated and the temperature was derived with the shape of the containers arranged in a stand-alone state as the parameter. (e)
  - The relational expression between activity concentration and amount of heat generated was derived with the nuclide, shape of container and solidification material as the parameters. (a, b. c)
  - The 2 formulas were used in combination to obtain the temperature of solidified substances with arbitrary parameters.
- Derivation of the relational expression between the amount of heat generated and the temperature (e)
  - The formula (Formula 2) for obtaining the maximum temperature from the amount of heat generated was derived based on the results of analysis using each system model. (Table 1)

 $T = oW^2 + pW + q$  (Formula 2)

- T : Maximum temperature [°C]
- W : Amount of heat generated [W/m³]

(Stand-alone type arrangement Cell with maximum temperature)



Table 1	List of relational expressions pertaining to the
	maximum temperature in each system

Sy	rstem model	Relational expression between the amount of heat generated and the maximum temperature
ent	1m ³ square	$T = -2.16 \times 10^{-4} W^2 + 0.70 W + 20$
igem	6m ³ square	$T = -3.03 \times 10^{-3} W^2 + 1.70 W + 20$
type arran	1m ³ square with shield	$T = -1.33 \times 10^{-4} W^2 + 0.43 W + 20$
one .	Pails	$T = -6.40 \times 10^{-5} W^2 + 0.10W + 20$
nd-a	Drum cans	$T = 3.06 \times 10^{-5} W^2 + 0.35 W + 20$
Sta	HIC	$T = -2.76 \times 10^{-3} W^2 + 1.28 W + 20$



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances

- Estimation of the thermal impact based on analysis results 2. Stand-alone type arrangement 2/2 -
- Derivation of relational expression between activity concentration and amount of heat generated (a, b.
   c)

The formula for obtaining the amount of heat generated from the activity concentration was derived using the results of analyzing the nuclides, shape of container and solidification material.

 First, basic conditions were established, and the relational expression between activity concentration and amount of heat generated under those basic conditions was derived. (Formula 3)

 $W = (1.21 \times 10^{-13}) \times x$  (Formula 3)

- W: Amount of heat generated [W/m³]
  x: Activity concentration [Bq/ cm³]
- Next, the results under basic conditions and other conditions were multiplied with the formula derived from the basic conditions as coefficients of each parameter and the formula (Formula 4) for obtaining the amount of heat generated under arbitrary conditions was derived.

 $W = \{(1.21 \times 10^{-13}) \times (a_{Nuclide}/a_{Cs-137})\} \times (a_{Container}/a_{drum}) \times (a_{Solidification material}/a_{Cement (Literature data)}) \times x$ (Formula 4)

- · Coefficient of nuclides:  $a_{Nuclide}/a_{Cs-137}$
- · Coefficient of the shape of container:  $a_{Container}/a_{drum}$
- · Coefficient of solidification material:  $a_{Solidification material}/a_{Cement (Literature data)}$
- Combined use of the formulas for estimating the temperature and amount of heat generated



For example, if the nuclide is configured as Co-60, shape as a 1m³ square, solidification material as AAM(M) and activity concentration as 1.0E+08 [Bq/cm³] (Table 2), based on
 W = {(1.21 × 10⁻¹³) × (2.96)} × (1.01) × (0.92) × (1.00 × 10⁸)

the amount of heat generated by the solidified substance was estimated to be  $3.31 \times 10^{1}$  [W/m³], and based on

 $T = -2.16 \times 10^{-4} W^2 + 7.04 \times 10^{-1} W + 20$ 

the temperature of the solidified substance was estimated to be 43.05°C.

- The formula for estimating the amount of heat generated was derived using the formula for estimating the maximum temperature of the solidified substance having an arbitrary shape and the coefficients of nuclides, shape of container, and solidification material respectively.
- By using the 2 formulas in combination, it became possible to estimate the maximum temperature of solidified substances under arbitrary conditions and activity concentration.



#### Table 1 Basic conditions

Nuclides	Cs-137
Shape of container	Drum cans
Solidification material	Cement

I. Analysis conditions (selected)		<ol> <li>Estimated amount of he</li> <li>(Coll with maximum tomp)</li> </ol>	eat generated
Container Square 1m^3		alone type arrangement)	
Solidification material	М	3.31E-05	[W/cm3]
Nuclides	Co-60	3.31E+01	[W/m3]
. Activity concentration	n (entered)	5. Estimated maximum tem (stand-alone type arran	iperature gement)
Activity concentration	1.005.09	43.05	[dedC]
[Ba/cm3]		L	1 –

Fig. 1 Example of a calculation sheet for estimating maximum temperature

Table 2 Values	used as	coefficients
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Coefficient	Conditions	Values
a _{Nuclide} /a _{CS-137}	Co-60	2.96
$a_{Container}/a_{drum}$	1m ³ square container	1.01
$a_{Solidificationmaterial}/a_{Cement(Literature)}$	AAM (M)	0.92

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- iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances
  - Estimation of the thermal impact based on analysis results 3. Accumulation type arrangement 1/2 -
- Estimation of thermal impact in an accumulation type arrangement
- The results of the stand-alone type arrangement that has sufficient analysis results (a, b, c) with nuclides and solidification material as parameters, were extended to the accumulation type arrangement to facilitate estimation of the temperature of solidified substances in an accumulation type arrangement. (d)
- The relational expression between the amount of heat generated and the temperature was derived. (e)
- The relational expression between the amount of heat generated in a stand-alone type arrangement and in an accumulation type arrangement was derived. (d)
- The 2 formulas were used in combination to obtain the temperature of solidified substances with arbitrary parameters.
- The scope of reflection of the formulas derived for the accumulation type arrangement is as follows.
  - Nuclide: Cs-137
  - Solidification material: Cement
  - Accumulated containers: Drum can, 1m³ square container
- > Derivation of the relational expression between the amount of heat generated and the temperature (e)
  - The relational expression (Formula 5) for obtaining the maximum temperature from the amount of heat generated in an accumulation type arrangement when there was no convection current was derived based on the results of analysis using each system model. (Table 1)

$$T = oW_{accumulation}^2 + pW_{accumulation} + q$$
 (Formula 5)

- · T : Maximum temperature [°C]
- *W_{accumulation}*: Amount of heat generated [W/m³] (Container with maximum temperature in accumulation type arrangement)

Table 1 List of relational expressions pertaining to the maximum temperature in each system

	System model	Relational expression between the amount of heat generated and the maximum temperature
ion type ment	108 Drum cans Accumulation type arrangement	$T = -1.70 \times 10^{-2} W_{accumulation}^{2} + 6.25 W_{accumulation} + 20$
Accumulat arrange	27 square 1m ³ containers Accumulation type arrangement	$T = -9.6 \times 10^{-3} W_{accumulation}^2 + 5.07 W_{accumulation} + 20$



- iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances
  - Estimation of the thermal impact based on analysis results 4. Accumulation type arrangement 2/2 -
- Derivation of the relational expression between the amount of heat generated in a stand-alone type arrangement and in an accumulation type arrangement (d)
- The relational expression between the amount of heat generated in a stand-alone type arrangement and in an accumulation type arrangement was derived based on the analysis results (Formula 6)

 $W_{accumulated} = b \times W_{Sum}$  (Equation 6)

- . W_{Sum}: Amount of heat generated [W/m³] (Sum of single units arrangement)
- . b : Coefficient (Depends on system model, nuclide)
- *W_{accumulated}*: Amount of heat generated [W/m³] (Sum of the containers with maximum temperature in accumulation type arrangement)
- The coefficient b of each system can be expressed by the following equations (Equation 7, Equation 8) based on the decay energy of the nuclides to be analyzed.
  - Drum cans:  $b = 0.18 \times l + 1$  (Equation 7)
  - $1m^3$  square container: $b = 0.06 \times l + 1$  (Equation 8)
- Largest proportion of photon energy emitted during decay of each nuclide
- W_{sum} was obtained with the following relational expression (Equation 9) based on the total amount of heat generated in the stand-alone type arrangement.

$$W_{sum} = k \times W$$
 (Equation 9) .

- W: Amount of heat generated [W/m³] (Cell with maximum temperature in stand-alone type arrangement)
  - k : Coefficient (Depends on nuclide)
  - W_{Sum}: Amount of heat generated [W/m³] (Sum of the single units arrangement)
- > Combined use of the formulas for estimating the temperature and amount of heat generated

By using Formula 6 and Formula 5 from the previous page in combination, it became possible in this section to estimate the maximum temperature of solidified substances under arbitrary conditions and activity concentration extended from the results of the stand-alone type arrangement, in the range of analysis results. (Fig. 1)

For example, if the nuclide is configured as Co-60, shape as a 1m³ square, solidification material as AAM(M) and activity concentration as 1.0E+08 [Bq/cm³] (Table 1),

based on

 $W_{accumulated} = (0.06 \times l + 1) \times k \times W$ 

the amount of heat generated by the solidified substance is estimated to be  $4.00 \times 10^{1}$  [W/m³], and based on

 $T = -9.6 \times 10^{-3} W_{accumulated}^2 + 5.07 W_{accumulated} + 20$ 

the temperature of the solidified substance is estimated to be 207.31°C.

- In this section, the equation for estimating the maximum temperature of solidified substances in an arbitrary accumulation type arrangement and the equation for estimating the amount of heat generated in an accumulation type arrangement from the amount of heat generated in a stand-alone type arrangement, in the range of the analysis results, were derived.
- By using the relational expressions in combination, it became possible to estimate the temperature of solidified substances in the range of accumulation extended from the results of the stand-alone type arrangement.



1. Analysis conditions (selected)

Container	Square1m^3
Solidification material	м
Nuclides	Co-60

2. Activity concentration (entered)

Activity concentration [Bq/cm3]	1.00E+08
---------------------------------------	----------

#### 3. Estimated amount of heat generated

(Cell with maximum temperature in the stand-alone type arrangement)

3.31E-05	[W/cm3]
3.31E+01	[W/m3]

### 4. Estimated amount of heat generated (Containers with maximum temperature in the accumulation type arrangement)

4.00E-05	[W/cm3] (27 containers_X2Y2Z3 container
4.00E+01	[W/m3] (27 containers_X2Y2Z3 container)

5. Estimated maximum temperature (stand-alone type arrangement)

- 4	3.05	10	ded(

6. Estimated maximum temperature (accumulation type arrangement)

207.3057129 [dedC] (27 containers_X2Y2Z3 container)

Fig. 1 Example of a calculation sheet for estimating maximum temperature

#### Table 1 Values used as coefficients

Coefficient	Conditions	Values
w	Co-60 1m ³ square container AAM (M)	$3.31 \times 10^{-5}$
l	Co-60	1.3320
k	Co-60	0.97

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(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances - Summary -

### **Results so far**

- ✓ An accumulation type arrangement model for 1m³ square containers which have a good storage efficiency was created for on-site application of the analytical data.
- It was found that the increase in amount of heat generated due to accumulation of containers depends on the energy of gamma rays released by the nuclide.
- ✓ The inventory limit value of Cs-137 obtained using the relational expression between activity concentration and maximum temperature of solidified substance derived using the temperature analysis results was 2.22 × 10⁹ Bq/cm³.
- The equation for estimating the maximum temperature of the solidified substances having an arbitrary shape and the equation for estimating the amount of heat generated using coefficients of nuclides, shape of container, and solidification material respectively, in the range of analysis results, were derived. By using equations in combination, it became possible to estimate the maximum temperature of solidified substances under arbitrary conditions and activity concentration.
- The equation for estimating the maximum temperature of solidified substances in an arbitrary accumulation type arrangement format and the equation for estimating the amount of heat generated in an accumulation type arrangement from the amount of heat generated in a stand-alone type arrangement, in the range of the analysis results, were derived. By using the equations in combination, it became possible to estimate the temperature of solidified substances in the range of accumulation by extending the results of the stand-alone type arrangement.

### Issues, etc.

✓ For more detailed study such as for the investigation of heat removal effect due to air convection, etc., it is necessary to conduct analysis in which conditions that take into account actual storage environment are set up.



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
 iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances
 Estimation of the thermal impact based on analysis results
 2. Stand-alone type arrangement 2/2 Supplementary materials

The following coefficients are listed as supplementary information for - Estimation of the thermal impact based on analysis results 2. Stand-alone type arrangement 2/2 -

 $a_{nuclide}/a_{CS-137}$ 

Table 1 List of coefficients  $(a_{nuclide}/a_{Cs-137})$ of nuclides in Equation 4

 $a_{nuclide}/a_{Cs-137}$ 

1 49

Nuclides

Pm-146

0,00	1.10	1 111 1 10	1.01
Y-90	1.23	Pm-147	0.08
Tc-99	0.14	Sm-151	0.03
Ru-106	1.87	Eu-152	1.53
Rh-106	2.12	Eu-154	1.80
Ag-110m	3.31	Eu-155	1.63
Cd-113m	0.25	Pu-238	9.56
Sn-119m	0.02	Pu-239	8.69
Sn-123	0.71	Pu-240	8.49
Sn-126	3.41	Pu-241	1.91
Sb-125	0.71	Am-241	8.61
Te-123m	0.19	Am-242m	25.87
Te-125m	0.05	Am-243	374.13
Te-127	0.31	Cm-243	16.14
Te-127m	0.42	Cm-244	26.53
I-129	0.12	Mn-54	0.98
Cs-134	2.04	Co-60	2.96
Cs-137	1.00	Ni-63	0.02
Ce-144	1.76	Zn-65	0.66
Pr-144	1.64	H-3	0.01
Pr-144m	1.31		

Table 2 List of coefficients  $(a_{container}/a_{drum})$  of the shape of containers in Equation 4

Supplementary materials

Shape of container	$a_{\text{container}}/a_{\text{drum } can}$
Pails	0.871
Drum cylinders	1.000
HIC	1.045
1m ³ square container	1.010
6m ³ square container	0.979
1m ³ square container with shield	0.973

Table 3 List of coefficients

 $(a_{Solidification material}/a_{Cement (Literature data)})$  of solidification material in Equation 4)

Solidification material	$a_{\text{Solidification material}}/a_{OPC}$ (Literature data)	
Cement (Literature	1.00	
data)	1.00	
Cement	0.05	
(Measurements)	0.95	
AAM (M)	0.92	
AAM(MB20)	0.92	
AAM(MB40)	0.93	

Nuclides

Sr-90



#### Supplementary materials

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique

iii. Evaluation of the relationship between the inventory of Cs, etc. and the temperature of solidified substances

- Estimation of the thermal impact based on analysis results 4. Accumulation type arrangement 2/2 Supplementary materials -
- The following coefficients are listed as supplementary information for Estimation of the thermal impact based on analysis results 2. Accumulation type arrangement 2/2 -

Nuclides	l	b	k	Nuclides	l	b	k
Sr-90	0.0000	1.0000	1.01	Pm-146	0.4530	1.0831	0.91
Y-90	0.0000	1.0000	1.01	Pm-147	0.1210	1.0222	1.01
Tc-99	0.0000	1.0000	1.01	Sm-151	0.0215	1.0039	1.01
Ru-106	0.0000	1.0000	1.00	Eu-152	0.0400	1.0073	0.91
Rh-106	0.5120	1.0939	0.98	Eu-154	0.1230	1.0226	0.91
Ag-110m	0.6580	1.1207	0.89	Eu-155	0.0865	1.0159	1.00
Cd-113m	0.0000	1.0000	1.00	Pu-238	0.0159	1.0029	1.01
Sn-119m	0.0252	1.0046	1.00	Pu-239	0.0159	1.0029	1.01
Sn-123	1.0890	1.1997	0.99	Pu-240	0.0159	1.0029	1.01
Sn-126	0.0876	1.0161	0.94	Pu-241	0.0156	1.0029	1.00
Sb-125	0.0274	1.0050	0.94	Am-241	0.0161	1.0030	1.02
Te-123m	0.1590	1.0292	0.93	Am-242m	0.0179	1.0033	1.01
Te-125m	0.0274	1.0050	1.00	Am-243	0.0747	1.0137	1.00
Te-127	0.4180	1.0767	0.99	Cm-243	0.2776	1.0509	1.01
Te-127m	0.0274	1.0050	1.02	Cm-244	0.0169	1.0031	1.01
I-129	0.0297	1.0054	1.01	Mn-54	0.8350	1.1531	0.88
Cs-134	0.6050	1.1110	0.90	Co-60	1.3320	1.2443	0.89
Cs-137	0.6620	1.1214	0.93	Ni-63	0.0000	1.0000	1.01
Ce-144	0.1340	1.0246	1.02	Zn-65	1.1160	1.2046	0.88
Pr-144	0.6970	1.1278	0.99	H-3	0.0000	1.0000	1.01
Pr-144m	0.0359	1.0066	0.88				

Table 1 Coefficients for each nuclide in Equations 6 to 9 (Accumulation type arrangement of drum cans)

Table 2 Coefficients for each nuclide in the Equations 6 to 9 (Accumulation type arrangement of  $1m^3$  square containers)

Nuclides	l	b	k
Sr-90	0.0000	1.0000	1.00
Cs-137	0.6620	1.1214	0.98
Co-60	1.3320	1.2443	0.97
Pu-238	0.0159	1.0029	1.01
Am-241	0.0161	1.0030	1.02
H-3	0.0000	1.0000	1.00



 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

 Details of implementation 

- Achievements up to FY2020
- While a certain applicability has been confirmed through verification of the feasibility of estimating long-term changes in properties of solidified substances, using thermodynamic equilibrium calculations, it was found that knowledge pertaining to long term mineral facies changes needs to be acquired.
- The following issue was identified with respect to on-site application and further refinement of data.
  - Acquisition of tentative knowledge for making it possible to estimate long-term changes in properties of solidified substances
- Goal

In order to investigate long term property change behavior of solidified substances, the method of accelerated testing of long-term property change in solidified substances will be investigated, and the applicability of the investigated method of accelerated testing to evaluating longer term property change behavior of solidified substances will be studied.

### **Details of implementation**

Investigation of the method of accelerated testing related to change in properties

Investigation and organization of the factors affecting change in properties of cement, concrete, materials of the engineered barriers for geological disposal, glass, etc. which have been used in the past, the change in their properties and methods for evaluating the change in properties (including accelerated testing), etc.

#### Data acquisition through accelerated testing and study of applicability

Implementation of actual accelerated testing based on the methods of accelerated testing that were investigated, and acquisition of data.



Indicators for the achievements of goal

The method of accelerated testing of degradation is studied and the investigation results are presented.





(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique
 (b) iv. Investigation and evaluation of factors affecting long-term change in properties
 Investigation of the methods of accelerated testing -

- > Investigation of the methods of accelerated testing related to change in properties
- The factors affecting change in properties of cement, concrete, materials of the engineered barriers for geological disposal, glass, etc. which have been used in the past, the change in their properties and methods for evaluating the change in properties (including accelerated testing), etc. were investigated and organized (Table 1).
  - Table 1 Results of investigation and organization related to accelerated testing

Target	Factors leading to change in properties	Effect of the factors leading to change in properties	Phenomenon that could occur	Performance degradation	Method of evaluating the change in properties (an example)	Accelerated testing (an example)	Testing standards, etc. for reference
		Shrinking associated with drying	Cracks	Strength reduction	Visual confirmation, measurement of change in length, strength measurement, etc.	Test of shrinkage due to drying	JIS A 1151(2002)
		Change in capacity due to change in temperature	Cracks	Strength reduction	Visual confirmation, strength measurement, etc.	Heating-cooling cyclic test	JIS A 6909(2014)
	Temperature	Change in capacity due to freezing and thawing	Cracks Peeling Popping out	Strength reduction	Visual confirmation, elastodynamic coefficient measurement, etc.	Freezing-thawing test	JIS A 1148 (2001)
		Change in volume due to heat	Cracks	Strength reduction	Visual confirmation, measurement of change in length, strength measurement, etc.	Heat curing test	
		Change in generation phase	Cracks	Strength reduction	Visual confirmation, measurement of change in length, strength measurement, XRD, etc.	Heat curing test	
Concrete Carbon Acid, Wate	Humidity	Change in capacity due to change in humidity	Cracks	Strength reduction Visual confirmation, measurement of change in length, strength measurement, etc.		Dry-wet cyclic test	
	Salt content	Expansion due to steel corrosion	Cracks	Strength reduction	Visual confirmation, spontaneous potential measurement, strength measurement, etc.	Salt water immersion test Salt spray test	JCI(2014)
		Change in capacity due to carbonation	Cracks	Strength reduction		Accelerated carbonation test	
	Carbon dioxide gas	Steel corrosion due to decrease in pH of concrete	Cracks	Strength reduction	Visual contirmation, carbonation depth measurement, strength measurement, etc.		JIS A 1153(2003)
	Acid, alkali, etc.	Decomposition of cement hydrates	Cracks Peeling	Strength reduction	Visual confirmation, determination of the quantity of hydrates, strength measurement, etc.	Immersion test	
	Water (Rainfall)	Decomposition and leaching of cement hydrates	Cracks Peeling	Strength reduction	Visual confirmation, determination of the quantity of hydrates (TG/DTA), strength measurement, etc.	Immersion test Water flow test Electrical enhancement test Chemical enhancement test	
		Movement of water soluble components	Efflorescence	Deterioration of view Strength reduction	Visual confirmation, color tone measurement (color difference meter), etc.	Efflorescence test	
	Load	Deformation of components due to tensile stress	Cracks	Strength reduction	Measurement of change in dimensions, strength measurement, etc.	Point load test	
Polymer	Temperature	Change in capacity due to change in temperature	Cracks Peeling	Reduction in adhesive ability	Heating-cooling cycle, etc.	Heating-cooling cyclic test	JIS A 6909(2014)
cement mortar	Ultraviolet rays	Polymer degradation	Chalking		Visual confirmation, color tone measurement (color difference meter), etc.	Accelerated weathering test	JIS A 6909(2014)
	Heat	Change in capacity due to change in temperature	Cracks	Strength reduction	Visual confirmation, strength measurement, etc.	Heating-cooling cyclic test	JIS R 3223(2017)
Glass	Water	Leaching of components	Weight reduction	Strength reduction	Liquid phase composition analysis, etc.	Immersion test Flowing water solubility test	JIS R 3503(2007)
	Acid, alkali, etc.	Erosion	Weight reduction	Strength reduction	Liquid phase composition analysis, etc.	Immersion test Flowing water solubility test	JIS R 3503(2007)

R

- The accelerated tests specified in JIS were only for evaluating performance related to strength reduction.
- ✓ However, evaluating only strength reduction is not sufficient for evaluating the performance of waste bodies (deterioration).
- ✓ It is believed that an evaluation method is required for verifying the impact of change in properties that affects changes in the mineral facies that contributes to the confinement function of solidified substances.
- A testing method believed to be likely to affect changes in mineral facies was identified by focusing on the effects of factors leading to change in properties.



✓ Heat curing test✓ Efflorescence test

• Further, the following testing methods that enable quantification of acceleration based on JIS standards were identified for evaluating the impact on strength reduction.

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## (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties Selection of the methods of accelerated testing -

- > The following 4 tests were identified and implemented as tests for verifying the long-term property change behavior of solidified substances (Table 1)
  - In addition, the anticipated factors leading to change in properties were reviewed considering the properties of 1F waste, the treatment methods, etc.

Type of test	Details of anticipated change in properties	Anticipated factors leading to change in properties	Evaluation indicator	Method of quantifying acceleration
Freezing-thawing test	Mixing using comparatively greater amount of water is required for the low temperature solidification treatment of waste resulting from water treatment (mainly slurry). The integrity (strength, etc.) of the solidified substances is likely to largely reduce in response to the effect of freezing and thawing when the storage environment of the solidified substances is inland, in a cold region, in indoor areas where air-conditioning doesn't work, in outdoor areas.	Temperature and water	Strength reduction	Available (JIS A 1148)
Accelerated carbonation test	If the integrity of the container in which solidified substances are stored is lost, the properties of solidified substances are likely to change in response to the impact of carbon dioxide gas in the atmosphere.	Carbon dioxide gas	Strength reduction	Available (JIS A 1153)
Heat curing test	The amorphous and crystalline phases that are formed as a result of the impact of water supplied from outside the solidified substance or the change in temperature during long-term storage, undergo change in phase, due to which the properties of the solidified substances are likely to differ largely.	Temperature and water	Change in mineral facies phase	Not available⁺
Efflorescence test	If the containers in which solidified substances are stored lose their integrity, the moisture content in the solidified substances gradually evaporates due to which the elements in the solidified substance are likely to move to the surface of the solidified substance and in addition a change in phase is likely to occur.	Water	Mass transfer Change in mineral facies phase	Not available [*]
References Equilibrium solubility test (Implementation in FY2020)	It is believed that leaching from the solidified substance depends on the amount of moisture content that comes in contact per unit of time. Leaching of water soluble components is expected to be accelerated when the amount of moisture content that comes in contact with respect to weight of solidified substance (solid to liquid ratio) is changed.	Water	Leaching of water soluble components	Not available (There are some ideas)

Table 1	Selected	testing	method
	00100100	County	mounou

*Although it has not been established as a method of accelerated testing of degradation (at this point in time there is no equation for evaluating and calculating the acceleration), it is presumed to be similar to the property change mechanism when cement solidified substances are stored, and is believed to be necessary for evaluating the performance of solidified substances (change in mineral facies).





 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties
 Freezing-thawing test 1. Overview and test conditions -

Freezing-thawing test: Test for evaluating the internal damage of concrete caused due to frost damage. (JIS A 1148) Table 1 Overview of the freezing-thawing test

Factors leading to change in properties	Evaluation items	Criteria	Dimensions of solidified substance [mm]	Material age at the beginnin g	Measurement items during each cycle	End period	Measurement items pertaining to samples after the tests are concluded
Temperature	Frost damage resistance (Frost deterioration)	Mutual elastodynamic coefficient 60% or more (Refer to Table 2)	40 × 40 × 160	28 days	First resonance frequency Ultrasonic propagation time (Every 30 cycles*)	300 cycles	Ultrasonic propagation time Measurement of compressive strength MIP Measurement

Freezing-thawing test conditions

*Measured at the 5th cycle and 15th cycle only immediately after the test is started, and thereafter measured every 30 cycles.



MIP Measurement: Number of tests 1

#### Table 2 Placement of mutual elastodynamic coefficient in academic conferences

Name of literature	Mutual elastodynamic coefficient	Additional Notes
Architectural Institute of Japan Recommendations for Durability Design and Construction Practice of Reinforced Concrete Buildings	60% when cracks are at a critical stage	The concrete deterioration indicator when cracks are formed that give rise to safety problems associated with use, is called as mutual elastodynamic coefficient.
Japan Society of Civil Engineers Standard Specifications for Concrete Structures	60% at the minimum limit value	Cross-sectional thickness about 20[cm] or more. (When freezing and thawing is frequently repeated, and the structures are exposed as usual)





Fig. 1 Freezing-thawing test equipment and test status



## (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties Freezing-thawing test 2. Calculation of acceleration -

> Investigation of acceleration in the freezing-thawing test

The extent to which test conditions specified in JIS A 1148 "Method of test for resistance of concrete to freezing and thawing" are accelerated as compared to the conditions in actual environment was calculated based on the "Recommendations for Durability Design and Construction Practice of Reinforced Concrete Buildings" of the Architectural Institute of Japan.

#### Results of investigating acceleration

 $SL_d = C_{\nu n} / C_{\nu p}$ 

In the "Recommendations for Durability Design and Construction Practice of Reinforced Concrete Buildings, 2016 edition" the following method has been described for verifying performance against frost damage.

- The limit state of the cracks due to frost damage is set in terms of the mutual elastodynamic coefficient.
- When the mutual elastodynamic coefficient reaches 60% that state is set as the design limit state. (Design limit: The state when there are safety problems associated with use)
- The number of years in which cracks reach the limit is obtained using the equation below.



 $C_{yn}$  Number of test cycles when the mutual elastodynamic coefficient of the limit state is reached according to the ASTM C 666 (JIS A 1148) A standard [cycles]

Cvp: Number of annual ASTM*1 equivalent cycles *2 [cycles/year]

*1: ASTM: American Society for Testing and Materials





## (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties Freezing-thawing test 3. Test results -

Results of the freezing-thawing test

Table 1 Results of the freezing-thawing test

	Measurement items									Number of	
Sample name		External o	observation		First resonance frequency	Ultrasonic propagation time	Changes and prope	rty changes from the initial sample	Frost damage	years in which cracks reach	
	Precipitate	ipitate Cracks Scaling Damaged		Mutual elastodynamic coefficient	Ultrasonic propagation rate	Strength	MIP	resistance	the limit		
Cement base material	-	-	-	-	<ul> <li>Image: A second s</li></ul>	✓ (100%)	No change (107%)	No major change	+++	48 years or more	
Cement + CS	+	+	+	+	1	✓ (86%)	No change (86%)	No major change	+++	48 years or more	
Cement + IS	-	+	-	-	×	✓ (94%)	No change (102%)	No major change	+++	48 years or more	
AAM(M) base material	+	+	-	+	×	× (59%)	- (14%)	Increase in number of pores of size 10 ⁻² to 10 ⁰	+	3 years	
AAM (M) + CS	+	-	+	-	1	✓ (91%)	+ (245%)	Decrease in number of pores of size 10 ⁻² to 10 ⁰	++	48 years or more	
AAM (M) + IS	+	-	-	-	1	✓ (77%)	- (33%)	Increase in number of pores of size 10 ⁻¹ to 10 ⁰	++	48 years or more	
AAM(MB40) base material	+	-	-	-	×	✓ (97%)	- (34%)	Increase in number of pores of size 10 ⁻² to 10 ⁰	+	43 years	
AAM(MB40) + CS	+	-	-	-	1	✓ (71%)	△ (74%)	Increase in number of pores of size 10 ⁻¹ to 10 ⁰	++	48 years or more	
AAM(MB40) + IS	+	-	-	-	×	✓ (71%)	- (25%)	Increase in number of pores of size 10 ⁻² to 10 ⁰	++	48 years or more	
Description of the signs and symbols		+: F —: No	ormed t formed		<ul> <li>✓: 60% or more</li> <li>×: 60% or less</li> </ul>	<ul> <li>:60% or more</li> <li>:60% or less</li> <li>Value in () indicates lowest</li> <li>value of the ultrasonic</li> <li>propagation speed rate during</li> <li>the period</li> </ul>	<ul> <li>+: Increase</li> <li>△: Reduction of less than 50%</li> <li>-: Reduction of 50% or more</li> </ul>	_	Number of +: Extent of frost damage resistance	-	

*Scaling · · · A frost damage phenomenon because of which concrete deteriorates, and in which there is peeling on the surface due to the effect of freezing and thawing.

- Cement based solidified substance : There was little change in properties (strength reduction and change in porosity) due to freezing and thawing in all cases
- AAM(M) based solidified substance: Strength of the base material reduced. However, it is presumed that AAM(M+CS) and AAM(M+IS) are unlikely to be affected as compared to the base material.
- AAM(MB40) based solidified
   Strength of the base material reduced. However, it is presumed that AAM(MB40+CS) and AAM(MB40+IS) are unlikely to be affected as compared to the base
   material.

Freezing-thawing test : Change in properties of solidified substances due to freezing and thawing can be accelerated quantitatively (equivalent to 48 years (288 times) in the Fukushima region, by means of the 300 cycle test where the mutual elastodynamic coefficient is 60%)) and the cracks in the solidified substance and strength reduction can be evaluated.



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties - Accelerated carbonation test 1. Overview and test conditions -

Accelerated carbonation test: Test for evaluating degradation of material by accelerating carbonation of concrete due to carbon dioxide gas in atmosphere (JIS A 1153) Table 1 Overview of the accelerated carbonation test

Factors leading to change in properties	Evaluation items	Dimensions of solidified substance [mm]	Material age at the beginning	Measurement items	Acceleration period [weeks]	Measurement items pertaining to samples after the test is completed ^{*2}	
Carbon dioxide gas	Evaluation of the impact of carbon dioxide gas in atmosphere	20 × 20 × 50	28 days old (Including 20°C, 60%RH	Carbonation depth	0 ^{*1} , 1, 4, 8	MIP measurement (Measured at 0, 1, 4, 8 weeks) XRD measurement (Measured at 0, 1, 4, 8 weeks) EPMA measurement (Measured at 8 weeks)	
	(Carbonation, drying)	20 × 20 × 30	arying)	Ultrasonic propagation time ^{*2} Compressive strength ^{*2}		_	
*1 Initial sample before accelerated carbonation *2 Measured in addition to							

Accelerated carbonation test conditions



Fig. 1 Status of formation of cracks in the carbonated cross section

Fig. 2 Status of cracked surface before and after spraying phenolphthalein solution (AAM(MB40) base material Acceleration period 4 weeks)



## (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties Accelerated carbonation test 2. Calculation of acceleration -

> Investigation of the extent of acceleration under the accelerated carbonation test conditions

The extent to which test conditions under the accelerated environment specified in JIS A 1153 "Method of accelerated carbonation test for concrete" are accelerated as compared to the atmosphere was calculated based on the "Recommendations for Durability Design and Construction Practice of High Durability Reinforced Concrete Structures" of the Architectural Institute of Japan and past research (Abe and others, 1990) that formed the basis of the test method.



Conversion of the accelerated carbonation test conditions

Using the above method of calculation as reference, the period required for the carbonation depth in atmosphere to change to accelerated carbonation depth at 8 weeks of acceleration under the JIS conditions was calculated. The carbon dioxide gas concentration in atmosphere was considered to be 0.03% indoors and about 0.1% outdoors (assuming that all other conditions remain constant)



If the carbonation depth at 8 weeks of acceleration period is converted to carbonation depth in atmosphere, it is equivalent to about 26 years (outdoors) and about 8 years (indoors).

167 times 50 times



 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties
 Accelerated carbonation test 3. Test results (Cement based solidified substances) -

Results of the accelerated carbonation test

Table 1 Results of accelerated carbonation test of cement based solidified substances (up to 8 weeks of acceleration period)

External obs	servation Cracks	Carbonation Depth (8 weeks)	Changes and change in	properties from the init	al sample (Accelerate	ed carbonation period: 0 weeks)		under actual		
Precipitate	Cracks	Carbonation Depth (8 weeks)				Changes and change in properties from the initial sample (Accelerated carbonation period: 0 weeks)				
-			Strength	XRD	MIP	ЕРМА	Carbonation resistance	equivalent to 8 weeks of acceleration period		
	+	4.9mm	+ (172%)	Calcite formation	No major change	Ca, SI, AI: Decrease in concentration up to 5 mm from the surface	++			
-	-	1.3mm	+ (137%)	Belite peak reduction	No major change	Na: Increase in surface concentration CI: Decrease in surface concentration, increase in concentration from 5mm and deeper	+++	Outdoors: 26 years (Accelerated 167 times) Indoors: 8 years		
-	-	6.8mm	+ (139%)	Belite peak reduction Disappearance of Halite	No major change	Na: Increase in concentration up to 3 mm from the surface CI: Decrease in concentration up to 3 mm from the surface, increase in concentration from 3mm and deeper	++	(Accelerated 50 times)		
Formed Not formed	+: Formed -: Not formed	-	+: Increase △: Decrease of less than 50% -: Decrease of 50% or more Rate of change from the initial sample is indicated by the numerical value in ()	-	-	_	Number of +: Extent of carbonation resistance	-		
6.00 4.00 2.00 6.00 5.00 5.00 4.00 2.00 5.00 <b>itial</b>	16, 60 14, 60 17, 60 10, 60 6, 60 4, 60 2, 60 9, 60 9, 60	5 OPC+IS 4 Increase to about 3 1 Carbona 0 5 10 15	Initial Carbonation B mm from the surface	BW CI 5 50	0.00 6.00 1.00 2.00 1.00 0.60 0.10 0.20 0.00 0.00 0.10 0.00 0.10	6.00 6.00 2.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.00	rease in Cl concentration amm or deeper ase in Cl concentration ase in Cl concentration amm from the surfact rbonation depth 6 0 15 20 25 30	Initial Carbonation 8w Adduction ation on up to ce .8 mm 0 35 40 45 50		
+: F No 6.0 8.0 8.0 8.0 1112	Formed tr formed	iormed +: Formed t formed -: Not formed	1.3mm 1.3mm 6.8mm :ormed t formed -: Not formed - Not formed 	- + 4.9mm + (172%) 1.3mm + (137%) 6.8mm + (139%) 6.8mm + (139%) - +: Increase $\Delta$ : Decrease of less than 50% -: Decrease of less than 50% -: Decrease of 50% or more Rate of change from the initial sample is indicated by the numerical value in () Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial Initial 	-       +       4.9mm       + (172%)       Calcite formation         -       -       1.3mm       + (137%)       Belite peak reduction         -       -       6.8mm       + (139%)       Belite peak reduction         -       -       6.8mm       + (139%)       Belite peak reduction         :ormed       +: Formed       -       -       -         :ormed       -: Decrease of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -         Image: Stress of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -       -         Image: Stress of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -       -         Image: Stress of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -       -         Image: Stress of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -       -         Image: Stress of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -       -         Image: Stress of S0% or more Rate of change from the initial sample is indicated by the numerical value in ()       -       -         Image: Stress of S0% or more S0 = S0 =	+       4.9mm       + (172%)       Calcite formation       No major change         -       -       1.3mm       + (137%)       Belite peak reduction       No major change         -       -       6.8mm       + (139%)       Belite peak reduction       No major change         -       -       6.8mm       + (139%)       Belite peak reduction       No major change         -       -       6.8mm       + (139%)       Belite peak reduction       No major change         -       -       6.8mm       + (139%)       Belite peak reduction       No major change         -       -       6.8mm       -       -       -       -         -       -       -       -       -       -       -         -       -       -       -       -       -       -         -       -       -       -       -       -       -         *: torrease it tormed       -: Not formed       -       -       -       -         *: torrease it to about 3 mm from the surface       -       -       -       -         *: torrease if to about 3 mm from the surface       -       -       -       -         *: torease if to about 3 mm from th	+       4.9mm       + (122%)       Calcite formation       No major change       Ca, S, A: Decrease in concentration up to 5 mm from the surface         -       -       1.3mm       + (137%)       Beilte peak reduction       No major change       Na: Increase in concentration increase in concentration increase in concentration increase in concentration from 5mm and deeper         -       -       6.8mm       + (139%)       Beilte peak reduction       No major change       Na: Increase in concentration up to 3 mm from the surface         -       -       6.8mm       + (139%)       Beilte peak reduction       No major change       Na: Increase in concentration up to 3 mm deeper         ormed       +: Formed       -       -       6.8mm       + (139%)       Beilte peak reduction       No major change       Na: Increase in concentration up to 3 mm from the surface         ormed       +: Formed       -       -       -       -       -       -         it formed       -       -       -       -       -       -       -         it formed       -       -       -       -       -       -       -         it formed       -       -       -       -       -       -       -         it formed       -       -       -       <	+       4.9mm       + (172%)       Calcite formation       No major change       CG, SJ, Al: Decrease in concentration       + + +         -       -       1.3mm       + (137%)       Belite peak reduction       No major change       No: increase in surface concentration       + + +         -       -       6.8mm       + (137%)       Belite peak reduction       No major change       No: increase in concentration from 5mm       + + +         -       -       6.8mm       + (139%)       Belite peak reduction       No major change       No: increase in concentration up to 3 mm from the surface. Increase in concentration up to 3 mm from the surface. Increase in concentration up to 3 mm from the surface. Increase in concentration up to 3 mm from the surface. Increase in concentration up to 3 mm from the surface. Increase in concentration from 5mm and deeper       + + +         iormed       -: Formed       -       -       -       -       -       -       -       Number of +:       Extend of change from the initial sample is indicated by me numerical value in ()       -       -       -       -       -       -       -       -       -       -       -       Number of +:       Extend of change from the initial sample is indicated by me numerical value in ()       -       -       -       -       -       -       -       -       -       -       - <t< td=""></t<>		

Fig. 1 Some of the EPMA mapping images and concentration distribution (Cement + IS: Samples with the deepest carbonation and element mapping images with significant fluctuations are selected)

✓ Cement based solidified substance: Strength increased and changes in the mineral facies phase were observed in the case of base material, CS mixture as well as IS mixture.



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

- Accelerated carbonation test 4. Test results (AAM (M) based solidified substances) -

#### Results of the accelerated carbonation test

Table 1 Results of accelerated carbonation test of AAM(M) based solidified substances (up to 8 weeks of acceleration period)

		Evaluation items	Period anticipated						
Sample name	External obs	ervation		Changes and change in pr	Changes and change in properties from the initial sample (Accelerated carbonation period: 0 weeks)				
	Precipitate	Cracks	Carbonation Depth (8 weeks)	Strength	XRD	MIP	ЕРМА	Carbonation resistance	equivalent to 8 weeks of acceleration perioc
AAM (M) base material	-	-	4.1mm	No change (88%)	No major change	Increase in number of pores of size 10 ⁻² to 10 ⁻⁰	Si: Increase in concentration up to 5mm from the surface Na: Increase in concentration up to 5mm from the surface	++	
AAM (M+CS)	+ (Nahcolite)	-	0.92mm	+ (201%)	Northupite formed	Decrease in porosity Decrease in number of pores of size 10 ⁻² to 10 ⁰	Ca, SI, AI, Mg: Decrease in concentration up to 2mm from the surface Na, CI: Increase in concentration up to 2mm from the surface (Formation of precipitate)	+++	Outdoors: 25 years (Accelerated 167 times) Indoors: 8 years (Accelerated 50 times
AAM (M+IS)	+ (Halite)	-	1.2mm	No change (105%)	No major change	No major change	Na, CI: Increase in concentration up to 1mm from the surface (Formation of precipitate)	+++	
Description of the signs and symbols	+: Formed -: Not formed	+: Formed -: Not formed	-	+: Increase △: Decrease of less than 50% -: Decrease of 50% or more Rate of change from the initial sample is indicated by the numerical value in ()	-	-	-	Number of +: Extent of carbonation resistance	-
St St	14, 98 14, 08 12, 09 18, 09 6, 09 6, 09 0, 09 Nitial	25 M 12, 80 20 10, 80 20 4, 80 (% \$\$20 4, 80 (% \$\$20 10, 80 20 4, 80 (% \$\$20 10, 10 5 80 0	Increase in concentration up about Snam from surface Carbonation depth	Linitial Carbonation Bw Si o to to the 4.1mm		24. 60 13. 00 15. 00 9. 00 6. 00 9. 00 6. 00 9. 00 6. 00	25 15.00 20 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.00 7.0	crease in N oncentration up t seut 5mm from th inface	nitial Carbonation 8w
[S		Gen	Distance from expo	osed face (mm)	אן		5 U 5	Distance from exposed	35 40 45 50 face (mm)

Fig. 1 Some of the EPMA mapping images and concentration distribution (M base material: Samples with the deepest carbonation and element mapping images with significant fluctuations are selected)

AAM(M) based solidified substances: There was no change in strength and mineral facies phase in the case of base material and IS mixture. Strength increased and changes in the mineral facies phase were observed only in the case of CS mixture.



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

- Accelerated carbonation test 5. Test results (AAM (MB40) based solidified substances) -

Results of the accelerated carbonation test

Table 1 Results of accelerated carbonation test of AAM(MB40) based solidified substances (up to 8 weeks of the acceleration period)

				Measur	rement items			Evaluation items	Period anticipated
Samplo namo	External	observation		Changes and chan	ge in properties from the	initial sample (Accelerat	ed carbonation period: 0 weeks)		environment
Sample hame	Precipitate	Cracks	Carbonation Depth (8 weeks)	Strength	XRD	MIP	ЕРМА	Carbonation resistance	weeks of the acceleration period
AAM (MB40) base material	-	+	35.6mm	- (41%)	Disappearance of C-S- H Formation of Aragonaite	No major change	Ca: Decrease in concentration in the carbonated part Na: Increase in surface concentration	+	
AAM (MB40+CS)	-	-	6.9mm	No change (107%)	Formation of Faujasite	Increase in number of pores of size 10 ⁻² to 10 ⁻¹	Ca, SI, AI, Mg: Decrease in concentration up to 7mm from the surface Na, CI: Increase in surface concentration, decrease in concentration up to 1 to 5mm from the surface	++	Outdoors: 26 years (Accelerated 167 times) Indoors: 8 years
AAM (MB40+IS)	+	-	5.3mm	+ (123%)	Formation of Faujasite Disappearance of Halite	Increase in number of pores of size 10 ⁻² to 10 ⁻¹	Ca: Increase in concentration up to 3 to 5 mm Si, Al: Increase in concentration up to 7mm from the surface Na, Cl: Increase in surface concentration, decrease in concentration up to 1 to 5mm from the surface	++	(Accelerated 50 times)
Description of the signs and symbols	+: Formed -: Not formed	+: Formed -: Not formed	-	+: Increase ∆: Decrease of less than 50% -: Decrease of 50% or more Rate of change from the initial sample is indicated by the numerical value in ()	-	-	-	Number of +: Extent of carbonation resistance	-
	8:00 2:00 4:00 4:00 1:00 2:00 2:00 2:00 5:00 5:00	60.45 60.55 60.55 60.65 60.65 60.65 60.65 60.65 60.65 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55 60.55	¹⁵ MB40 ¹⁰ Decrease in Ca (Carbonated pa	Carbonation 80 Carbonation dept 35.6mm concentration rt)	th	24.00 21.00 19.00 17.00 2.00 2.00 2.00 2.00 3.00 8.00 8.00	MB40 13.000 13.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.0000 15.00000 15.00000 15.0000 15.0000 15.000000000 15.000000000000 15.00000000000000000000	in Na oncentration	arbonation depth
[Ca	C.	OW 5ee	Distance fro	om exposed face (mm)	30	[Na]	Distance	from exposed face (m	m)

Fig. 1 Some of the EPMA mapping images and concentration distribution (MB40 base material: Samples with the deepest carbonation and element mapping images with significant fluctuations are selected)

AAM(MB40) solidified substance
 Accelerated carbonation test

: Cracks were formed on the base material. Zeolite mineral (Faujasite) was formed in the CS mixture and IS mixture.

: This implies the possibility of being able to evaluate the strength of solidified substances and changes in the mineral facies by quantitatively accelerating change in properties (carbonation due to carbon dioxide gas). (It is necessary to check whether it is the same as the status of change in properties due to carbonation in actual environment)



(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties - Heat curing test 1. Overview and test conditions -



solidified substances and in the pore structure.

Heat curing test

Intensity (a.u.)

⇒ The results of the heat curing test were consolidated by solidified substance.

AAM(M+CS) 80°C sealing 4weeks

needle crystals will be added in

accordance wit

the products.

AAM(M+CS) 20°C

sealing_4weeks

iv. Investigation and evaluation of factors affecting long-term change in properties

- Heat curing test 2. Test results (Cement based solidified substances) -

> Results of the heat curing test (Cement based solidified substances)

Table 1 Poculte of boat curing test of coment bac	ad calidifiad cubetancae (XRD	MID Curing pariod of 8 wooks)
Table T Results of fleat curring test of certient bas	כם שטווטוווכט שטשנמוונכש (אועט	

Sample name	Sealed			Humid				
	20 [°C]	50 [°C]	80 [°C]	20 [°C]	50 [°C]	80 [°C]		
Cement base material	_	_	Monosulfate that is the AFm phase disappeared Thermodyanmically stable Katoite was formed. Size of the pores increased due to drying, and number of pores that were of the size 10 ⁰ [µm] or less increased	•Number of pores of the size 10 ⁻¹ to 10 ⁰ [µm] decreased due to hydration	•Number of pores of the size 10 ⁻¹ to 10 ⁰ [µm] decreased due to hydration	<ul> <li>Monosulfate that is the AFm phase disappeared</li> <li>Thermodyanmically stable Katoite was formed.</li> <li>Belite of clinker mineral disappeared.</li> <li>Number of pores of the size 10⁻¹ to 10⁰ [µm] decreased due to hydration</li> </ul>		
Cement + CS	-	Belite of clinker mineral disappeared	<ul> <li>Belite of clinker mineral disappeared.</li> <li>Friedel's salt that is the AFm phase disappeared.</li> <li>Thermodyanmically stable Katoite was formed.</li> </ul>	•Number of pores of the size 10 ⁻¹ to 10 ⁰ [µm] decreased due to hydration •Number of pores of the size 10 ⁻² [µm] or lower increased due to structural change in C-S-H.	<ul> <li>Belite of clinker mineral disappeared.</li> <li>Number of pores of the size 10⁻¹ to 10⁰ [µm] decreased due to hydration</li> <li>Number of pores of the size 10⁻² [µm] or lower increased due to structural change in C- S-H.</li> </ul>	<ul> <li>Belite of clinker mineral disappeared.</li> <li>Friedel's salt that is the AFm phase disappeared.</li> <li>Thermodyanmically stable Katoite was formed.</li> <li>Number of pores of the size 10⁻¹ to 10⁰ [µm] decreased due to hydration</li> <li>Number of pores of the size 10⁻² [µm] or lower increased due to structural change in C-S-H.</li> </ul>		
Cement + IS	_	Belite of clinker mineral disappeared.     Thermodyanmically stable Katoite was formed.	<ul> <li>Belite of clinker mineral disappeared.</li> <li>Friedel's salt that is the AFm phase decreased.</li> <li>Thermodyanmically stable Katoite was formed.</li> <li>Size of the pores increased due to drying, and number of pores that were of the size 10^o [µm] or less increased</li> </ul>	_	<ul> <li>Belite of clinker mineral disappeared.</li> <li>Thermodyanmically stable Katoite was formed.</li> </ul>	<ul> <li>Belite of clinker mineral disappeared.</li> <li>Friedel's salt that is the AFm phase disappeared.</li> <li>Thermodyanmically stable Katoite was formed.</li> </ul>		

✓ The AFm phase (Monosulfate, Friedel's salt) disappeared at 80°C in all samples, and instead the thermodyanmically stable Katoite was formed.

- ✓ Katoite which was not formed in cement base material and cement + CS solidified substances even at 50°C was formed in the case of cement + IS solidified substances.
  ⇒ Due to the increase in Fe content associated with mixing of waste, Fe solid solution Katoite that can form even at room temperature is likely to be formed.
- In the case of humid curing samples, since hydration of the samples progressed further, it was confirmed that the peak of Belite disappeared and the pore structure changed.

(In the case of sealed curing samples as well, change in the peak of Belite and change in the pore structure were verified.)


(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

- Heat curing test 3. Test results (AAM (M) based solidified substances) -

> Results of the heat curing test (AAM(M) based solidified substances)

			5		,	- /	
Sample name		Sealed		Humid			
	20 [°C]	50 [°C]	80 [°C]	20 [°C]	50 [°C]	80 [°C]	
AAM(M) base material	_	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ to 10⁰μm increased.</li> </ul>	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ to 10⁰</li> <li>[µm] increased.</li> </ul>	_	• Faujasite was formed • Due to formation of Faujasite (crystallization), pores of size 10 ⁻¹ to 10 ⁰ [µm] increased.	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ to 10⁰</li> <li>[μm] increased.</li> </ul>	
AAM (M+CS)	<ul> <li>Faujasite was formed</li> <li>Pores of size about 10⁰ [µm] formed due to CS mixing decreased (cause unknown)</li> </ul>	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ [µm] increased.</li> <li>Pores of size about 10⁰ [µm] formed due to CS mixing decreased (cause unknown)</li> </ul>	<ul> <li>Faujasite was formed</li> <li>Sodalite was formed</li> <li>Due to formation of</li> <li>Sodalite (crystallization),</li> <li>pores of size 10⁻¹ [µm] or</li> <li>less decreased.</li> <li>Pores of size about 10⁰</li> <li>[µm] formed due to CS</li> <li>mixing decreased (cause unknown)</li> </ul>	• Faujasite was formed • Pores of size about 10 ⁰ [µm] formed due to CS mixing decreased (cause unknown)	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ [µm] increased.</li> <li>Pores of size about 10⁰ [µm] formed due to CS mixing decreased (cause unknown)</li> </ul>	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ [µm] increased.</li> <li>Pores of size about 10⁰ [µm] formed due to CS mixing decreased (cause unknown)</li> </ul>	
AAM (M+IS)	-	• Faujasite was formed • Due to formation of Faujasite (crystallization), pores of size 10 ⁻¹ to 10 ⁰ [µm] increased.	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ to 10⁰</li> <li>[µm] increased.</li> <li>Pores of size about 10⁻¹</li> <li>[µm] decreased (cause unknown)</li> </ul>	-	• Faujasite was formed • Due to formation of Faujasite (crystallization), pores of size 10 ⁻¹ to 10 ⁰ [µm] increased.	<ul> <li>Faujasite was formed</li> <li>Due to formation of</li> <li>Faujasite (crystallization), pores of size 10⁻¹ to 10⁰</li> <li>[µm] increased.</li> <li>Pores of size about 10⁻¹</li> <li>[µm] decreased (cause unknown)</li> </ul>	

Table 1 Results of heat curing test of AAM(M) based solidified substances (XRD, MIP Curing period of 8 weeks)

✓ Due to heat curing, N-A-S-H in the matrix portion of the solidified substance crystallized, and zeolite minerals (Faujsite, Sodalite) were formed.
⇒ In some of the samples, the same zeolite minerals were found even at 20°C, due to which it is believed that the reaction got accelerated due to heat curing.

The formed zeolite mineral differed depending on the compounding or curing conditions, and the change in pore structure also differed depending on the formed zeolite mineral.

⇒ It implies that the formed zeolite mineral changes depending on the composition of the solidified substance, and the difference in its formation process leads to change in pore structure.



iv. Investigation and evaluation of factors affecting long-term change in properties

- Heat curing test 4. Test results (AAM (MB40) based solidified substances) -

> Results of the heat curing test (AAM(MB40) based solidified substances)

Table 1 Results of heat curing test of AAM(MB40) based solidified substances (XRD, MIPCuring period of 8 weeks)

Sample name	Sealed			Humid			
	20 [°C]	50 [°C]	80 [°C]	20 [°C]	50 [°C]	80 [°C]	
AAM (MB40) Base material	•C-S-H was formed	·C-S-H was formed	•C-S-H was formed •Size of the pores increased due to drying, and number of pores that are of the size 10- 1 [µm] or less increased	•C-S-H was formed	•C-S-H was formed	•C-S-H was formed •Faujasite was formed	
MB40 +CS	• Faujasite was formed • Due to formation of Faujasite (crystallization), pores of size 10 ⁻¹ to 10 ⁰ [µm] increased.	• Faujasite was formed • Due to formation of Faujasite (crystallization), pores of size 10 ⁻¹ to 10 ⁰ [µm] increased.	<ul> <li>Faujasite was formed</li> <li>Sodalite was formed</li> <li>Due to formation of Sodalite (crystallization), pores of size 10⁻¹ [µm] or less decreased.</li> </ul>	• Faujasite was formed	<ul> <li>Faujasite was formed</li> <li>Due to formation of Faujasite (crystallization), pores of size 10⁻¹ to 10⁰ [µm] increased.</li> </ul>	<ul> <li>Faujasite was formed</li> <li>Due to formation of Faujasite (crystallization), pores of size 10⁻¹ to 10⁰ [µm] increased.</li> <li>Zeolite Na-P was formed</li> </ul>	
MB40 +IS	<ul> <li>C-S-H was formed</li> <li>Faujasite was formed</li> <li>Pores of size about 10^o [µm] or more increased (cause unknown)</li> </ul>	<ul> <li>C-S-H was formed</li> <li>Faujasite was formed</li> <li>Pores of size about 10^o [µm] or more increased (cause unknown)</li> </ul>	<ul> <li>C-S-H was formed</li> <li>Sodalite was formed</li> <li>Due to formation of Sodalite, pores of size 10⁻¹ [µm] or less decreased</li> <li>Pores of size about 10⁰ [µm] or more increased (cause unknown)</li> <li>Size of the pores increased due to drying, and number of pores of size about 10⁻¹ [µm] increased.</li> </ul>	• C-S-H was formed • Faujasite was formed • Pores of size about 10 ^o [µm] or more increased (cause unknown)	• C-S-H was formed • Faujasite was formed • Pores of size about 10 ^o [µm] or more increased (cause unknown)	• C-S-H was formed • Faujasite was formed • Sodalite was formed • Zeolite Na-P was formed • Pores of size about 10 ^o [µm] or more increases (cause unknown)	

✓ Due to heat curing, N-A-S-H in the matrix portion of the solidified substance crystallized, and zeolite minerals (Faujsite, Sodalite, Zeolite Na-P) were formed.
⇒ In some of the samples, the same zeolite minerals were found even at 20°C, due to which it is believed that the reaction got accelerated due to heat curing.

The formed zeolite mineral differed depending on the compounding or curing conditions, and the change in pore structure also differed depending on the formed zeolite mineral.

⇒ It implies that the formed zeolite mineral changes depending on the composition of the solidified substance, and the difference in its formation process leads to change in pore structure.

#### Results of the heat curing test

 It was found in the heat curing test that the formation of mineral facies can get accelerated due to factors leading to change in properties (temperature and moisture content), compared to the basic conditions (factors leading to change in properties are not manipulated) with sealed curing at 20°C.



 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties
 Efflorescence test 1. Overview and test conditions -



#### Efflorescence test

Table 1 Overview of the efflorescence test

IRID

iv. Investigation and evaluation of factors affecting long-term change in properties

- Efflorescence test 2. Test results (Cement based solidified substances) -

 Results of the efflorescence test (Cement based solidified substances)

Table 1 Results of efflorescence test of cement based solidified

	Test name*1				Measurement it	ems		Evaluation items
Sample		External observation		Rate of	Chan	ges and property changes f	irom the initial sample	Mass transfer due
name		Precipitate	Cracks	precipitate formation [%]	XRD (1, 4, 8 weeks)*2	MIP (1, 4, 8 weeks) ^{*2}	EPMA (Only 8 weeks)	to water absorption
Cement base	Efflorescence	-	-	-	No major change	No major change	Ca: Decrease in concentration of top surface 2 mm Na: Increase in concentration of the top surface 2 mm	+
material	Heat curing 20°C sealed	-	-	Not evaluated	No major change	No major change	Not evaluated	Not evaluated
Cement +	Efflorescence	+	-	4.2	No major change	No major change	Na, Cl: Decrease in concentration (overall) (Due to formation of precipitate)	+++
CS	Heat curing 20°C sealed	-	-	Not evaluated	No major change	No major change	Not evaluated	Not evaluated
Cement + IS	Efflorescence	-	-	-	No major change	No major change	Na: Decrease in concentration in the section that is 2 mm and deeper from the top surface CI: Decrease in concentration at the top surface and bottom surface	+
	Heat curing 20°C sealed	-	-	Not evaluated	No major change	No major change	Not evaluated	Not evaluated
Description	n of the signs and symbols	+: Formed -: Not formed	+: Formed -: Not formed	-: Not formed	-	-	Top surface: dry surface Bottom surface: immersed surface	Number of +: Extent of transferability

*1 The results of sealed curing at 20°C during the heat curing test are provided as reference for comparison with efflorescence test. *2 XRD and MIP measurement is carried out only at 4 and 8 weeks in the case of sealed curing at 20°C during the heat curing test.

- ✓ In cement based solidified substances, precipitate (Thermonatrite, Halite, Gaylussite) formed only in the case of Cement + CS.
- ✓ The XRD and MIP results after the 8 week test period were not much different than the initial sample.
- According to the EPMA results, the concentration of Na and Cl changed (mass transfer) even in the case of base material and Cement + IS in which precipitate was not formed.
- Compared to sealed curing at 20°C in the heat curing test in an environment without contact with water, since precipitate formed in the case of Cement + CS, it is believed that the efflorescence test accelerates change in properties of cement based solidified substances.



iv. Investigation and evaluation of factors affecting long-term change in properties

- Efflorescence test 3. Test results (AAM (M) based solidified substances) -

> Results of efflorescence test (AAM(M) based solidified substances)

Table 1 Results of efflorescence test of AAM(M) based solidified substances (Test period 8 weeks)

					Measurement items			Evaluation items
Sample name	Test name*1	External ob	External observation		Changes and	I property changes from the in	itial sample	Mass transfer due to
	Test hanc	Precipitate	Cracks	precipitate formation [%]	XRD (1, 4, 8 weeks) ^{*2}	MIP (1, 4, 8 weeks) ^{*2}	EPMA (Only 8 weeks)	water absorption
AAM(M) base material	Efflorescence	+	+	8.0	Formation and disappearance of Thermonatrite (Na dissolution in the base material)	on and disappearance of Thermonatrite Decrease in bulk density dissolution in the base Increase in porosity (overall) material)		+++
	Heat curing 20°C sealed	-	-	Not evaluated	No major change	No major change	Not evaluated	Not evaluated
	Efflorescence	*3	+	Not measured*3	Disappearance of Thermonatrite (Na dissolution in the base material and in CS)	Decrease in bulk density Increase in porosity	Na, CI: Decrease in concentration (overall) (Due to formation of precipitate)	+++
(M+CS)	Heat curing 20°C sealed	-	-	Not evaluated	Faujasite was formed	Decrease in pores of size about 10 ⁰ [µm] formed due to mixing of CS (cause unknown)	Not evaluated	Not evaluated
AAM (M+IS)	AAM (M+IS)		-	8.8	No major change	Decrease in bulk density Increase in porosity	Si, Al: Decrease in concentration (overall) Na, Cl: Decrease in concentration (overall) (Due to formation of precipitate)	+++
	Heat curing 20°C sealed	-	-	Not evaluated	No major change	No major change	Not evaluated	Not evaluated
Description of th	ne signs and symbols	+: Formed -: Not formed	+: Formed -: Not formed	-: Not formed	-	-	Top surface: dry surface Bottom surface: immersed surface	Number of +: Extent of transferability

*1 The results of sealed curing at 20°C during the heat curing test are provided as reference for comparison with efflorescence test.

*2 XRD and MIP measurement is carried out only at 4 and 8 weeks in the case of sealed curing at 20 °C during the heat curing test.

*3 Since there was water up to the top surface of the solidified substance, precipitate was not found, but when the solidified substance is dried, a large quantity of precipitate is formed.

 Precipitate was formed in all AAM(M) based samples. However, in the case of AAM(M+CS) precipitate was formed during the drying process after the test was concluded.

(Thermonatrite precipitated in all AAM(M) based samples, further Gaylussite precipitated in AAM(M+CS) based samples and Halite precipitated in AAM(M+IS) based samples.)

✓ As compared to sealed curing at 20°C in the heat curing test in an environment without contact with water, since it was found that precipitate was formed in all samples, that Thermonatrite formed and disappeared, that porosity increased and bulk density decreased, it is believed that the efflorescence test accelerates change in properties of AAM(M) based solidified substances.



iv. Investigation and evaluation of factors affecting long-term change in properties

- Efflorescence test 4. Test results (AAM (MB40) based solidified substances) -

> Results of efflorescence test (AAM(MB40) based solidified substances)

Table 1 Results of efflorescence test of AAM(MB40) based solidified substances (Test period 8 weeks)

		Measurement items								
Sample	Test name*1	External o	bservation	Rate of	Changes and property	y changes from the initial sam	ple	Mass transfer		
name		Precipitate	tate Cracks formation [%]		XRD (1, 4, 8 weeks) ^{*2}	MIP (1, 4, 8 weeks)* ²	EPMA (Only 8 weeks)	due to water absorption		
AAM (MB40)	AAM Efflorescence + +		+	0.9	Formation of Aragonaite and Calcite (Formed due to Ca dissolution in the base material and carbonation)	No major change	Ca, Si, Al, Na: Decrease in concentration	++		
material	Heat curing 20°C sealed	-	-	Not evaluated	C-S-H	C-S-H No major change		Not evaluated		
AAM	Efflorescence	+	+	0.5	No major change	Increase in bulk density Decrease in porosity	CI: Decrease in concentration (Due to formation of precipitate)	++		
(MB40 +CS)	Heat curing 20°C sealed	-	-	Not evaluated	Faujasite was formed	Due to formation (crystallization) of Faujasite, number of pores of size 10 ⁻¹ to 10 ⁰ [µm] increased	Not evaluated	Not evaluated		
AAM (MB40 +IS)	Efflorescence	Efflorescence + - 2.1 C-S-H (Formed due to the hydration reaction of slag) Calcite formation (Formed due to Ca dissolution in the base material and carbonation) Disappearance of Halite (Almost all the Cl in the solidified substance got		Decrease in bulk density Increase in porosity Decrease in the number of pores of size 10 ⁻² to 10 ¹	Na, Cl: Decrease in concentration (Due to formation of precipitate)	+++				
	Heat curing 20°C sealed	-	-	Not evaluated	C-S-H was formed Faujasite was formed	Pores of size 10 ⁰ [µm] or more increased (cause unknown)	Not evaluated	Not evaluated		
Descriptior s	n of the signs and symbols	+: Formed -: Not formed	+: Formed -: Not formed	-: Not formed	-	-	Top surface: dry surface Bottom surface: immersed surface	Number of +: Extent of transferability		

*1 The results of sealed curing at 20°C during the heat curing test are provided as reference for comparison with efflorescence test.

*F2 XRD and MIP measurement is carried out only at 4 and 8 weeks in the case of sealed curing at 20°C during the heat curing test.

✓ It was found that precipitate formed in all samples as compared to sealed curing at 20°C in the heat curing test in an environment without contact with water.

(Thermonatrite precipitated in all AAM(MB40) based samples, further Halite and Gaylussite precipitated in AAM(MB40+CS) based samples and Halite precipitated in AAM(MB40+IS) based samples.)

Even though the generation phase changed and changes took place in the porosity and bulk density under sealed curing at 20°C during the heat curing test, formation of precipitate was not being confirmed. Hence, it is believed that the efflorescence test accelerated changes in properties of AAM(MB40) based solidified substances.



#### (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

- Efflorescence test 5. Test results (EPMA mapping images) -

#### Results of the efflorescence test (Example of EPMA mapping image)

1 sample each with large precipitate formation is selected from cement based, AAM(M) based and AAM(MB40) based solidified substances, and these are shown as some of the EPMA analysis result. (Table 1) The initial samples were analyzed at a depth of 30 mm*, and the samples after an 8-week acceleration period were analyzed at a depth of 20 mm*². The results are listed focusing on the Na and CI contained in the precipitates (Thermonatrite, Halite) found.

Table 1 Some of the EPMA mapping images and concentration distribution





 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

 Summary of accelerated testing 

Accelerated			Applicability to evaluation of long-		
testing type	Purpose of the tests	Cement	AAM (M)	AAM(MB40)	term property change behavior
Freezing-thawing test	To observe changes in properties due to changes in the form of moisture in the solidified substances because of the effect of freezing and thawing, by a s u m ing storage environment of the solidified substances to be inland, in a cold region, in doors where air conditioning does not work, and outdoors.	<ul> <li>There was no major change in properties due to freezing and thawing in any of the cement based solidified substances. Hence it is believed that the substances are frost damage resistant.</li> </ul>	<ul> <li>Cracks formed on the AAM(M) base material in a short period. Hence its frost resistance is low.</li> <li>AAM(M) + CS showed a clear difference in behavior than the other solidified substances, but the cause was not clear.</li> <li>It was presumed that AAM(M)+IS was unlikely to be affected by freezing and thaving as compared to AAM(M) base material.</li> </ul>	<ul> <li>AAM(MB40) base material was largely affected by freezing and thawing because of its reduced compressive strength and the behavior of the mutual elastodynamic coefficient.</li> <li>Although the impact on AAM(MB40)+CS and AAM((MB40)+IS was small as compared to AAM((MB40), it is believed that they were affected by freezing and thawing.</li> </ul>	<ul> <li>Changes in properties can be accelerated, and the cracking of the solidified substance or decrease in strength can be evaluated.</li> <li>However, as water which is a medium for diffusion gets solidified, it is believed that changes in the mineral facies get suppressed.</li> </ul>
Accelerated carbonation test	To obtain information on the solidified substance property change behavior in response to the impact of carbon dioxide gas in atmosphere, assuming that the integrity of the container in which solidified substances are stored is lost.	<ul> <li>The cement based solidified substances were resistant to carbonation even if those solidified substances were mixed with simulated carbonate slurry or simulated iron co-precipitation slurry.</li> </ul>	<ul> <li>The carbonation depth of AAM(M) based solidified substances was small, and as major changes were not seen in compressive strength, it may be said that these solidified substances are carbonation resistant.</li> </ul>	<ul> <li>Since carbonation due to cracks and decrease in compressive strength were conspicuous in AAM(MB40) base material, it is believed that its carbonation resistance is low.</li> <li>The carbonation depth of AAM(MB40)+CS and AAM((MB40)+IS was 5mm or lower, and there was no decrease in the compressive strength, it is believed that these solidified substances are carbonation resistant.</li> </ul>	<ul> <li>This implies the possibility of being able to evaluate the strength of solidified substances and changes in the mineral facies by quantitatively accelerating change in properties (carbonation due to carbon dioxide gas). (It is necessary to check whether it is the same as the status of change in properties due to carbonation in actual environment)</li> </ul>
Heat curing test	To accelerate long term change in properties of AAM material (zeolite mineral formation, etc.) through heating on a trial basis, and observe the change in properties of solidified substances.	<ul> <li>Cement hydrates containing Al underwent phase change at 80°C curing temperature.</li> <li>This implies that there is interaction between the simulated iron co- precipitation slurry and cement hydrates in the solidified substances.</li> <li>The change in mineral facies had hardly any impact on the change in pore structure.</li> </ul>	<ul> <li>N-A-S-H crystallized due to heat curing, ar zeolite minerals formed even at 20°C).</li> <li>The formed zeolite minerals changed depe solidified substance, and the change in por mineral.</li> <li>Multiple factors such as difference in comp temperature, etc. compounded and gave right</li> </ul>	In a complex change in the port of the samples and zeolite minerals were formed (in some of the samples anding on the compounding or curing condition of the re structure also differed depending on the formed zeolite osition of the matrix part, impact of waste, impact of se to a complex change in the pore structure.	<ul> <li>It was verified by comparing with the reference (20°C sealed curing) that change in properties (change in mineral facies) was accelerated due to heat curing.</li> <li>It was found that 2 factors, namely temperature and moisture (vapor), contributed to the change in properties,</li> </ul>
Efflorescence test	To observe the impact on the solidified substances when nuclides get transferred to the top surface of the solidified substances, assuming that the container in which the solidified substances are stored has lost its integrity due to which the moisture in the solidified substances gradually evaporates.	<ul> <li>As the mineral composition or pore structure of all of the cement based solidified substances remained almost unchanged from the initial samples, it is believed that there was little mass transfer inside the solidified substances as compared to AAM(M) solidified substances and AAM(MB40) solidified substance.</li> </ul>	<ul> <li>In the case of AAM(M) base material and AAM(M)+IS, it was found that bulk density decreased and porosity increased due to the formation of precipitate.</li> <li>In the case of AAM(M)+CS, precipitate did not form during the test, but it was verified that precipitate formed when the surface was dried.</li> </ul>	<ul> <li>It was found the precipitate was formed in all AAM(MB40) solidified substances.</li> <li>The pore structure remained almost unchanged in the AAM(MB40) base material.</li> <li>Bulk density increased and porosity decreased in the case of AAM (MB40)+IS bulk density decreased rapidly and porosity exponentially increased.</li> </ul>	<ul> <li>It was verified by comparing with the reference (20°C sealed curing) that change in properties (change in mineral facies) was accelerated when efflorescence test was conducted.</li> <li>It was found that moisture (pore water), contributed to the change in properties.</li> </ul>

As it was confirmed during the accelerated carbonation test, heat curing test and efflorescence test that the mineral facies changed, there is a possibility of being able to accelerate long term change in mineral facies of the solidified substance and evaluate it.

 It was found that change in mineral facies differed depending on the difference in factors (water, temperature) leading to change in properties or the way in which those factors influence the change in properties.



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 (a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technique iv. Investigation and evaluation of factors affecting long-term change in properties

 Summary 

#### **Results so far**

- Accelerated testing that is expected to be applicable to the evaluation of long-term property change behavior was investigated.
- ✓ With regards to the freezing-thawing test, changes in properties (freezing and thawing) can be accelerated to bring about conditions equivalent to 48 years (288 times) in the Fukushima region, and the cracking of the solidified substance or decrease in strength can be evaluated. However, as water which is a medium for diffusion gets solidified, it is believed that changes in the mineral facies get suppressed.
- ✓ With regards to the accelerated carbonation test, it was implied that there is a possibility of being able to evaluate the strength of solidified substances and changes in the mineral facies by accelerating changes in properties (carbonation due to carbon dioxide gas) 167 times (outdoors) and 50 times (indoors). (It is necessary to check whether the status of change in properties in the case of AAM is the same as that caused by carbonation in actual environment)
- With regards to heat curing test, it was verified that it was possible to accelerate changes in properties (change in mineral facies) by means of the factors leading to change in properties (temperature and vapour).
- ✓ With regards to efflorescence test, it was verified that it was possible to accelerate changes in properties (change in mineral facies) by means of the factors leading to change in properties (pore water).

#### Issues, etc.

- Methods for measuring the amount of moisture passing into the solidified substances need to be studied.
- ✓ With regards to the efflorescence test and heat curing test, quantification of the extent of acceleration needs to be studied by comparing the presence of acceleration and the extent of development of property changes (change in mineral facies) over a long period of time.



i. Organization of evaluation axis and acquired data for comparing technologies

- Achievements up to FY2020
- The concept of solidification treatment equipment was studied, data pertaining to economic efficiency such as facility configuration, treatment efficiency, maintenance details, types of consumables and replacement frequency, types and quantity of secondary waste generated from contaminated water treatment, etc. was acquired, and it was organized along with the data acquired on a trial basis, for evaluating waste body specifications of various treatment technologies.
- In doing so, a series of approaches were developed for ① Selecting the applicable solidification treatment based on the characteristics of waste, ② Selecting a solidification treatment technology that is more suitable to the requirements even if it is for the same solidification treatment, ③ investigating the pro and cons of introducing the selected technology based on the technical achievements of the selected solidification treatment technology, process information, and economic efficiency data.
- The following issue was identified in detailed development of the selection method.
  - Expansion of data on an ongoing basis for evaluating the range of solid waste to which the treatment technologies are applicable

#### Goal

To present methods for evaluating the treatment technologies that are likely to be applicable to various solid waste, and their scope of application, by organizing the evaluation axis of the treatment technologies, their process data, etc. based on the variety of acquired data, in order to evaluate the range of solid waste to which the treatment technologies are applicable.

#### **Details of implementation**

Constant updating of the evaluation methods (approaches)

Ongoing investigation of multi-faceted evaluation methods (approaches) for selecting the solidification technology for secondary waste generated from contaminated water treatment

#### Reflection of data acquired this year

Updation by reflecting the findings from change in properties of solidified substances, long-term property change behavior, etc. due to heating, etc. acquired this year.



#### Indicators for the achievements of goal

Techniques (approach methods) for evaluating solidification technologies that are likely to be applicable to various solid waste are presented.





(a) Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology i. Organization of evaluation axis and acquired data for comparing technologies

#### Updating the FY2021 approach



- Investigation and evaluation of factors affecting long-term change in properties
- Investigation related to volume of Cs volatilization during high temperature treatment and its control
- Data acquired on solidified substances containing chemical admixtures was add section concerning "Characteristics of solidified substances" from among the 5 iter

Evaluation axis	Evaluation sub-items	ICV Joule heating (GeoMelt)	Plasma heating	Induction heating (Super-high frequency)	Cement in-drum	Cement out-drum	AAM in-drum
	Homogeneity	There was no problem as viscosity at the treatment temperature was controlled by the composition (Except when evaluating TiO ₂ )	Melting solidified LLW was found to be homogeneous.	There was stirring due to heat convection and electromagnetic force created by electromagnetic induction. After melting the entire quantity, by securing about 1 hour or more of retention time, the solidified substance became homogeneous.	Has not been evaluated, but is presumed to be high	Has not been evaluated, but is presumed to be high	Has not been evaluated, but is presumed to be high
	Approx. 300Mpa (When it was ensure that the solidification material is borosilicate glass)		Same as slag with CaO, MgO, SiO ₂ as the main ingredients (about 20MPa or more)	Almost the same as the quality of slag with CaO-MgO-SiO ₂ based components (SiO ₂ about 40wt%) or vitreous material	• 7MPa (28 days, carbonate slurry filling rate 30%) • 32MPa (26 days, iron co- precipitation slurry filling rate 20%)	• 7MPa (28 days, carbonate slurry filling rate 30%) • 32MPa (28 days, iron co- precipitation slurry filling rate 20%)	<ul> <li>SMPa or more (28 days, carbonate slurry filling rate 30%), (M, MB20, MB40)</li> <li>SMPa or more (28 days, iron co-precipitation slurry filling rate 20%) (M, MB20, MB40)</li> </ul>
fied substances	Leaching resistance	NMLu⊶c0.2 g/m²/d 10−4 kg/m2/d order (In the case of borosilicate glass)	Same as slag with CaO, MgO, SiO ₂ as the main ingredients (Estimated as 1E-7kg/m2/d (28d) or less (Cs, Sr))	Actual measurements are not available. Estimated to be almost the same as the quality of slag with CaO-MgO-SiO ₂ based components (SiO ₂ about 40wt%) or vitreous material.	Cs:95%, Sr:5%, Sr:≦0.1%, Ce:≦0.1% (Leaching rate evaluated assuming carbonate slurry filling rate 30%, ANS/ANSI-16.1)	Cs:95%, Sr:5%, Sr:≦0.1%, Ce:≦0.1% (Leaching rate evaluated assuming carbonate slurry filling rate 30%, ANS/ANSI-16.1)	Cs-23%, Sr.0.5%, Sn.5%, Ce:0.6% (Leaching rate evaluated assuming carbonate slurny filling rate 30% M solidified substance, ANS/ANSI-16.1)
naracteristics of solidi	Radiation resistance (Hydrogen G value and property change of solidified substance organization)	Negligible	Estimated to be a level at which there is no particular problem since it is a melting solidification substance.	Estimated to be a level at which there is no particular problem since it is a melting solidification substance.	• 0.16-0.17 (Carbonate slurry simulated powder filling rate 30%)	• 0.16-0.17 (Carbonate slurry simulated powder filling rate 30%)	0.13-0.28 (Carbonate slurry simulated powder filling rate 30% M solidified substance) 0.05-0.15 (Carbonate slurry simulated powder filling rate 30% MB solidified substance)
C	Heat resistance (Impact of thermal load on performance)	Negligible	Estimated to be a level at which there is no particular problem since it is a melting solidification substance.	Actual measurements are not available. Heat resistance is estimated to be high since it has the same characteristics as slag and glass.	Strength residual rate 90% or more when dried for 28 days at 80°C	Strength residual rate 90% or more when dried for 28 days at 80°C	Strength residual rate 20% or less when dried for 28 days at 80°C
	Long-term stability	Stable Estimated to be a level at which there is no particular problem since it is a melting solidification substance.		Almost the same as slag or glass with that composition			
				of a clidified ou	botopoo		
		Data on cr		solidilled su	bstances		
		obtained fro	om this year's	implementatio	on items is		
	and a second sec	reflecte	d as evaluatio	nd the	1		

approaches are upgraded and updated.

aluation sub-items of the on axis for FY2021.

Evaluation items	Evaluation sub-items
Technical achievements	Development stage/trends; proven record of application to general waste, radioactive waste, and secondary waste generated from water treatment; and main trouble case examples
Process performance	Number of processes, treatment temperature, treatment speed, maximum treatment unit, Cs volatilization rate, volatilization control measure, and required specifications for solidification containers
Operability and safety	Process risk, maintenance details and frequency, remote operability, and ease of solidification process control
Economic efficiency	Main facility configuration, power consumption, consumables, facility lifetime, work area, volume reduction rate, secondary waste/amount, and the possibility of treatment that mixes a different type of secondary waste generated from water treatment
Characteristics of solidified substances	Homogeneity, strength, leaching resistance, radiation resistance (hydrogen G value and property change of solidified substance organization), thermal resistance (the effects of a thermal load on the performance), long-term stability, effect of adding chemical admixtures



(a) Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology i. Organization of evaluation axis and acquired data for comparing technologies

The evaluation method (approach) was improved by adding data acquired on solidified substances containing chemical admixtures (Table 1).

Table 1 Details of the effect of adding chemical admixtures

Evaluation	Evaluation sub-	Cement in-drum	AAM in-drum
items	items	/ Cement out-drum	
Characteristics of solidified substances	[Addition] Effect of adding chemical admixtures	<ul> <li>Verified chemical admixtures</li> <li>&gt; Lignin sulfonic acid based</li> <li>&gt; Melamine based</li> <li>&gt; Naphthalene sulfonic acid based</li> <li>&gt; Polycarboxylic acid based</li> <li>&gt; Polycarboxylic acid based admixture was selected for carbonate slurry and polycarboxylic acid based admixture was selected for iron co-precipitation slurry.</li> <li>The amount of carbonate slurry filled can be increased by adding chemical admixtures.</li> <li>&gt; Scope of application:40%→45%</li> <li>In the case of iron co-precipitation slurry, since condensation gets delayed when filling rate increases, the scope of application cannot be expanded.</li> <li>Irradiation characteristics of base material containing chemical admixtures</li> <li>&gt; G value: 0.1 or less (irradiation of 3kGy Small cylinder of φ10x20mm)</li> </ul>	<ul> <li>Verified chemical admixtures</li> <li>Lignin sulfonic acid based</li> <li>Melamine based</li> <li>Naphthalene sulfonic acid based</li> <li>Polycarboxylic acid based</li> <li>Polycarboxylic acid based</li> <li>Solidified substances mixed with carbonate slurry showed a fluidity improvement effect.</li> <li>Fluidity of base material and all mixed systems reduced significantly in the case of polycarboxylic acid based admixture.</li> <li>The scope of application of carbonate slurry can be expanded to some extent.</li> <li>M: By adjusting the Na concentration, Si concentration, the filling rate of carbonate slurry can be improved from 30% up to 40%.</li> <li>MB40: There are concerns about expanding the scope of application as there is a significant drop in strength.</li> <li>Iron co-precipitation slurry mixture system does not show fluidity improvement effect.</li> </ul>

- ✓ The effect of addition of chemical admixtures (4 types) to carbonate slurry and iron coprecipitation slurry from among the secondary waste generated from contaminated water treatment, was added. The composition range that can contain secondary waste generated from contaminated water treatment was extended for cement + carbonate slurry by addition of chemical admixtures. (Fig. 1, Table 2)
- The acquired data was organized as the evaluation axis and was reflected in the investigation results for each solidification material.

Table 2 Scope of feasibility of solidification treatment when chemical admixtures are added (Cement + CS system)

	Without chemical admixtures	With chemical admixtures
Carbonate slurry	15 to 40wt%	15 to <mark>45</mark> wt%
Cement	30 to 55wt%	20 to 55wt%
Water	30 to 45wt%	30 to 45wt%



Fig. 1 Scope of application to cement + carbonate slurry



i. Organization of evaluation axis and acquired data for comparing technologies

#### - Summary -

#### **Results so far**

- ✓ Based on The investigation carried out in "①-i", the evaluation sub-item "Effect of adding chemical admixtures" was added to the section concerning "Characteristics of solidified substances" in the proposed evaluation axis.
- The results of studies on secondary waste generated from contaminated water treatment (simulated carbonate slurry, simulated iron co-precipitation slurry) in the solidified substances containing chemical admixtures were reflected, and the scope of application of the improved low temperature solidification treatment technology items was presented.

#### Issues, etc.

 Establishing methods for acquiring data that is common for waste bodies with different characteristics.

ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control

- Details of implementation -

- Achievement of the previous year
- In order to more appropriately select the high temperature treatment technology in which Cs volatilization becomes an issue, Cs volatilization and control mechanism was investigated, the Cs volatilization characteristics were consolidated based on the treatment method and operating conditions, and in addition, data related to the amount of volatilization was acquired and evaluated on a trial basis.
- Based on the above-mentioned investigation, the following issues were identified in developing a detailed selection method.
  - Expansion of data on an ongoing basis for evaluating the range of solid waste to which the treatment technologies are applicable
  - ✓ Further acquisition of data under conditions considering actual operation
- > Goal
- Cs volatilization is an issue in high temperature treatment technologies and has a strong influence on the type or facility configuration, etc. Hence, tests related to the Cs volatilization control mechanism will be conducted at the laboratory level using cold cap, top off frit, etc. regardless of the unique technology, to investigate the effect of Cs volatilization control.



Fig. 1 Overview of the test apparatus

#### Details of implementation

Acquisition of data related to Cs volatilization control based on tests Implementation of tests related to the Cs volatilization control mechanism at the laboratory level using cold cap, top off frit, etc. regardless of the unique technology, to investigate the effect of Cs volatilization control.



#### Indicators for the achievements of goal

Results of investigation of the volatilization control effect of various events are presented.





ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control

- Test conditions -

#### Test Conditions (Table 1)

• There are 2 loading formats as mentioned below.

Mixed sample: Loading format "Cs adsorbed zeolite + glass forming agent" for the sample being primarily assumed (Table 2).

<u>Cullet:</u> Mixed samples are melted to make glass cullets in order to reduce the volume of mixed samples due to testing related constraints (Next page Fig. 1)

• During test (5) (Top Off Frit (TOF)), 2 types of low melting point glass are selected and studied.

Table 1 Overview of the implemented test

The maximum glass temperature is set at 1200°C which is the maximum temperature of an electric furnace.

Table 2 Mixing ratio of zeolite and components of glass forming agent

No.	Test	Loading format ^{*1}	Quantity of sample loaded	Heating temperature	Surface temperature	Bubbling flow rate [mL / min]	Component	Reagent added	Weight added (%)
1	Air supply flow rate	Cullet	100	1200	1150		Zeolite	Zeolite	53.88
	dependency test			About 2	About 2		component	(Cs)	(3.81)
2	Supply mode dependency verification	Mixed sample	30	conditions between 800 to	conditions between 800 to		Simulated carbonate slurry	CaCO ₃	10.05
				1100 About 2 to 3	1100 About 2 to 3		component	MgO	6.95
3	Temperature dependency verification	Cullet	100	conditions between 800 to 1200	conditions between 800 to 1150		Simulated iron co-precipitation	Eo O	4 90
_		Cullet (Lower	80	About 2 conditions			slurry component	Fe ₂ O ₃	4.00
(4)	Cold cap	Mixed sample	20	between 1000 to 1200	800-900			SiO ₂	4.21
		(Upper layer)					Glass forming	B ₂ O ₃	4.71
5	Top off frit (2 types of glass with a	Cullet (Lower layer)	40 40	1200	1150		agent component	Na ₂ CO ₃	2.23
	low mening point)	TOF (Opper layer)				About 2 to 3		LiOH•H ₂ O	8.75
6	Bubbling	Cullet	100	1200	1150	conditions		ZrO ₂	4.42
		Cullet (Lower				About 2 to 2		Total	100.00
Ī	Cold cap + Bubbling	layer) Mixed sample (Upper layer)	80 20	1200	800	conditions between 10 to 200			

*1: When the loading method involves lower layer + upper layer, if loading cannot be performed at 2 layers, the method of loading the samples need to be re-examined.



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(a Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control - Test conditions -

Although the loading format for the samples being primarily assumed was supposed to be mixed samples, as the volume of the sample had to be reduced due to circumstances pertaining to this test, the samples were supplied in the form of glass cullets.

#### $\geq$ Preparation of sample to be loaded

Mixed sample made by adding glass forming agent to zeolite was heated at 1100°C and glass cullets were prepared (Fig. 1). The condition at the time of heating is shown (Fig. 2).







powder

Fig. 1 Preparation of the sample to be loaded



After heating













Along with increase in temperature, the precipitation phase* reduces



the cullets, but it was found that these dissolve when heated at 1200°C.

The surface

becomes flat, and the sample gets liquified

fuse

Fig. 2 Melting behavior of the prepared glass cullets *Pollucite (nepheline) which does not easily dissolve at low temperatures and ZrO₂ were found in

Preparation of the top off frit material  $\geq$ 

The top off frit materials (2 types) were heated under the following conditions respectively, to prepare top off frit. The condition at the time of heating is shown (Fig. 3).

Top off frit 1 (hereinafter, TOF-1)

Top off frit 2 (hereinafter, TOF-2)



The mixed reagent is heated at 1200°C and melted to test manufacture the top off frit material

Melting point 950 to 1000°C



Particle fusion



Disintegration of the shape of particles



Shape of the particles is lost, they get fused, and the surface becomes uneven



Particle fusion



950°C



Uneven surface

1000°C

The surface

becomes flat,

and the sample

aets liquified

of the shape of particles Fig. 3 Melting behavior of the prepared TOF









•



Melting point 1050 to 1100°C





Particle separation





(a) Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control

- Test results 1. (1) Air supply flow rate dependency, (2) Temperature dependency, (3) Supply mode dependency -

Test ① Air supply flow rate dependency test

• The air supply flow rate was changed and the volume of Cs volatilization was measured* (Fig. 1). Also, Cs was collected by means of a 6 stage offgas trap (Fig. 2). *Cs volatilization volume: Amount of volatilized Cs collected from each site in the test system





- volatilized Cs
- Fig. 2 Amount of Cs collected from each site

- ✓ With the increase in air supply flow rate, the amount of volatilized CS increased and the quantity that migrates to the subsequent stages of the off-gas trap increased.
- ✓ When the air was supplied at 100 [mL/min], and 500 [mL/min], the entire amount of volatilized Cs was collected in the 6 stage off-gas trap.

✓ It was decided to reflect the conditions when air was supplied at 500 [mL/min] in the test from then on.

- Test (2) + (3) Temperature dependency and supply mode dependency verification test
  - · The difference between the amount of volatilized Cs from the mixed sample obtained by adding glass forming agent to zeolite and the cullets was acquired by changing the temperature (Table 1, Fig. 3).

temperature and surface temperature							
Heating	Surface temperature [°C]						
temperature [°C]	Cullet	Mixed sample					
1100	1053	1075					
1000	955	977					
850	819	811					

Table 1 Relation between heating



Fig. 3 Comparison of the volume of Cs volatilization in case of the cullets and the mixed sample

- ✓ The amount of volatilized Cs was less in the case of mixed sample as compared at cullets at heating temperature 850°C and 1000°C.
- ✓ At 1100°C when both types of samples melted, the amount of volatilization increased in the case of the mixed sample.
  - ✓ Melting of samples can be considered to be an impact of the difference in surface temperature (Table 1).



 $\checkmark$  In the cold cap test (Test (4)) in which the surface temperature is low, it was decided that cullets would be used in the lower layer, the mixed sample would be placed in the upper layer part where the temperature will be 1000°C or lower, and the test would be conducted.



(a) Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control
 Test results_2.④ Cold cap ⑤ Top off Frit -

#### Test ④ Volatilization control effect of cold cap

The surface temperature of molten material was controlled to form a cold cap as illustrated in Fig. 1, and the volume of Cs volatilization under test conditions (Table 1) was measured (Fig. 2).





the volume of Cs volatilization

and the volume of Cs volatilization

- By letting a cold cap form on the surface of the sample, the volume of Cs volatilization was reduced to 1/50 or lower.
- ✓ It was confirmed that the volume of Cs volatilization depends on the temperature of the cold cap (sample surface temperature) and not on the temperature of the sample inside.

Test (5) Investigation of the volatilization behavior of Cs when Top off frit is used

• Top off frit material was loaded as shown in Fig. 3, and the volume of Cs volatilization respectively when 2 types of Top off frit material were used under temperature conditions mentioned in Table 2 was measured (Fig. 4).



- ✓ When a top off frit with a low melting point is used, the volume of Cs volatilization is likely to actually increase. Hence it is important to use frits that have a melting point closer to the heating temperature.
- ✓ By making sure that the surface temperature is lower than the melting point of the frits, high volatilization control effect was seen, but the amount of frits loaded increased (increase in volume of waste). Hence a balance between the amount loaded and the volume of volatilization is important.

#### IRID

(a) Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control

- Test results_3.6 Bubbling, 7 Cold cap + bubbling -

0

Test (6) Investigation of the Cs volatilization behavior as a result of bubbling

• Bubbling was performed and the volume of Cs volatilization was measured (Fig. 1, 2)



Fig. 1 Overview diagram of the test (Same for test 6 and 7)



Fig. 2 Dependency of the volume of Cs volatilization on the bubbling flow rate

- The moisture content from the air in the laboratory was removed and the resulting gas was used as bubbling gas.
- ✓ The volume of volatilization increased approximately linearly with the increase in bubbling flow rate in Region A where individually independent air bubbles were formed, but in Region B where the air bubbles were continuous, the volume of Cs volatilization showed a tendency to remain constant regardless of the flow rate.
- ✓ The volume of Cs volatilization when bubbling was performed at 10 [mL/min] (this is increased 4.9 times for in-furnace temperature) for melted samples with a volume of approx. 32 [mL] was 2.1 times, and it was 4.0 times when the flow rate was 50 [mL/min].

Test ⑦ Cold cap + bubbling

• Volume of Cs volatilization when bubbling is performed upon controlling volatilization by means of co ld cap, was measured (Table 1, Fig. 1, 3).





Fig. 3 Results of measuring the volume of Cs volatilization

- ✓ By letting a cold cap form, the volume of volatilization at the time of bubbling was reduced to 1/10 to 1/30.
- ✓ The volume of Cs volatilization tended to increase along with an increase in bubbling flow rate, but it was not significant.
- ✓ During Test 1 and Test 3, bubbles did not form at the surface (formation of hot spot) other than several occasions when they were observed in the initial stages.

Further, during the test white plume came up from the surface. It is believed that the bubbling gas escaped from between the sample particles of the cold cap.



(a) Establishment of selecting advance treatment methods [2] investigate of approach to assessing applicability for treatment technology ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control

- Volume of Cs volatilization for each volatilization control effect and its evaluation -

- > Changes in the volume of Cs volatilization for each volatilization control effect
  - The volume of Cs volatilization was organized by measurement site in the test equipment (Table 1).

 Table 1
 Volume of Cs volatilization by measurement site

Location		Reference	Test ④ Cold cap	Test ⑤ Top off frit	Test ⑥ Bubbling	Test ⑦ Cold cap + bubbling
Glass	temperature [°C]	1205	1185	1166	1198	1139
Surfac	e temperature [°C]	1147	803	1035	1184	919
Gla	ass sample [%]	99.092	99.983	99.956	96.353	99.798
	Tammann pipe	0.338	0.006	0.015	1.167	0.029
	T pipe	0.414	0.006	0.022	1.395	0.076
Volotilizod	Off-gas trap 1	0.069	0.003	0.002	0.989	0.054
substance	Off-gas trap 2	0.045	0.001	0.002	0.073	0.022
collection	Off-gas trap 3	0.021	0.000	0.001	0.016	0.012
site [%]	Off-gas trap 4	0.012	0.000	0.001	0.003	0.006
	Off-gas trap 5	0.009	0.000	0.000	0.002	0.002
	Off-gas trap 6	0.002	0.000	0.000	0.001	0.001
Total volatilization [%]		0.908	0.017	0.044	3.647	0.202
	Total [%]	100.000	100.000	100.000	100.000	100.000

- It was possible to reduce the volume of Cs volatilization to 1/50 or lower by letting a cold cap form with surface temperature approx. 800°C. It was indicated that formation of a cold cap was effective in controlling volatilization.
- ✓ The volume of Cs volatilization increased to maximum 5 times as a result of bubbling, but by letting a cold cap form, the volume of volatilization at the time of bubbling reduced to 1/10 1/30.
- It was indicated that it was possible to reduce the volume of Cs volatilization to 1/20 or lower by appropriately selecting and using the top off frit.
- ✓ Cs was collected the most at the T pipe site. This suggested that Cs is likely to accumulate at low temperature sites inside the equipment.

ii. Investigation related to volume of Cs volatilization during high temperature treatment and its control - Summary -

#### **Results so far**

- Data related to the volume of volatilization for each volatilization control factor was acquired, the volatilization control effect was compared, and results were presented.
- ✓ It was possible to reduce the volume of Cs volatilization to 1/50 or lower by letting a cold cap form with surface temperature approx. 800°C. It was indicated that formation of a cold cap was effective in controlling volatilization.
- ✓ It was indicated that it was possible to reduce the volume of Cs volatilization to 1/20 or lower by appropriately selecting and using the top off frit.
- ✓ The volume of Cs volatilization increased to maximum 5 times as a result of bubbling, but by letting a cold cap form, the volume of volatilization at the time of bubbling reduced to 1/10 1/30.

#### Issues, etc.

- The surface coverage of the cold cap is important for controlling the volume of Cs volatilization. Hence the method of capturing visible images needs to be studied.
- The impact of bubbling is expected to change depending on the test system. Hence further studies need to be carried out considering the difference with the actual equipment.
- The investigation on migration behavior of elements besides Cs at the time of melting needs to be studied as well.



- Achievements up to FY2020
- 1F waste was classified into 27 types, and waste information was organized by type.
- Based on investigation of cases from Japan and overseas, multiple dose evaluation conditions (scenario / model / parameters) were tentatively established while taking uncertainties into consideration, and preliminary dose evaluation was conducted for 27 types of 1F waste.
- The method for deriving the disposal classification of typical 1F waste depending on the treatment options was pointed out based on the results of the preliminary dose evaluation.

#### Goal

- To point out multiple disposal concepts for typical waste streams and items required for their safety evaluation and study, based on the results of the latest characterization and treatment studies.
- Details of implementation
- Study of multiple disposal concepts for typical waste streams based on the content of investigations related to disposal concepts and safety evaluation methods in Japan and overseas, as also information related to the properties of solid waste. And, identification of items that need to be studied for establishing the disposal concepts and safety evaluation methods for each disposal concept based on treatment technologies applicable to solid waste, and collection and organization of the required information.
- Indicators for determining goal achievement
- Presentation of typical waste streams along with the reason for their selection.
- Presentation of multiple disposal concepts for typical waste streams
- Presentation of items that need to be studied for establishing the disposal concepts and safety evaluation methods based on the treatment technologies for typical waste streams
- Collection and organization of information on the items that need to be studied as mentioned above.



Fig. 1 Flow of study of the disposal concepts and safety evaluation methods implemented in FY2019-2020



Fig. 2 Results of the FY2020 dose evaluation (Trench, groundwater)





FY2021 Study Plan Overview





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# (b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods FY2021 Study Plan Implementation items

#### Consolidation of input information

- Information on 1F waste studied until FY2020 (characterization, treatment method, materials with impact on disposal, etc.) will be collected and consolidated, and parameters such as waste amount, inventory, waste body properties, etc., used for evaluation will be established.
- Dose evaluation parameters such as the distribution coefficient, etc. for each nuclide that might impact nuclide migration will be established based on the latest domestic trend of the NUMO Comprehensive Technical Report, etc.

#### Implementation of sensitivity evaluation

- Models of multiple disposal concepts will be created with depths corresponding to trench disposal, pit disposal, and medium depth disposal based on the results of studies on disposal concepts conducted up to FY2020.
- Based on the results of studies on consolidation of input information, parameters for the sensitivity evaluation base case and the parameters for the sensitivity evaluation will be established.
- The models of multiple disposal concepts will be analyzed and evaluated by changing the parameters (barrier material, barrier thickness, distribution coefficient, etc.) of the facilities, and the sensitivity of parameters will be evaluated.

#### Study of preliminary proposals for the disposal concepts

• Based on the results of sensitivity evaluation and the results of consolidating input information, preliminary proposals for the disposal concepts suitable for the properties of 1F waste will be presented.

Dose evaluation

• Dose evaluation will be carried out with respect to the preliminary proposal of the disposal concepts, and revisions will be made based on the results.



# (b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Consolidation of input information (1/10)

- The amount and inventory used for investigating disposal concepts was updated based on the results of the analytical inventory studied in FY2020 as part of "Characterization".
- Treatment methods were established for each waste based on the results of studies on treatments conducted until FY2020 and the weight and amount when waste conditioning is carried out was calculated.
- The diffusion coefficient inside the solidified substance and the leaching rate of the solidified substance were calculated for the leaching rate from glass, melt, cement, AAM solidified substances, based on the outcome of studies on advance treatments up to FY2020.
- Some of the set values of distribution coefficients of FY2020 based on the application for changing the JNFL Low-level radioactive waste burial center, were reviewed while referring to the latest domestic disposal trend (NUMO Comprehensive Technical Report, etc.).



(b) Proposal of disposal methods and development of safety evaluation methods

① Consolidation of information to investigate the disposal concepts and to establish safety assessment methods

Consolidation of input information (2/10)

<Amount of waste (weight and volume)>

The amount of waste in which the outcome of characterization carried out in FY2020 was reflected, was used in the investigate of sensitivity analysis and disposal concept proposals.

•The sum total of the net volume of metal and concrete respectively in the rubbles in FY2021 are listed.

 $\Rightarrow$  The amount that takes waste conditioning into consideration is assumed to have an impact on the exposure dose mainly in the nearing scenario.

FY2021/2020 Comparison of amount of waste	ļ A	Amount of waste in FY202	20	Amount of waste in FY2021			
	m3	ton	ton/ m3	m3	ton	ton/ m3	
KURION	2847	11565	4.06	2877	11685	4.06	
AREVA	597	597	1.00	597	597	1.00	
SARRY	1196	5184	4.33	1274	5520	4.33	
Slurry 1_iron co-precipitation	1833	1833	1.00	2001	2001	1.00	
Slurry 2_Carbonate precipitation	7887	7887	1.00	8610	8610	1.00	
ALPS-4_Ag impregnated activated carbon	222	184	0.83	181	181	1.00	
ALPS-2_titanate	332	276	0.83	272	272	1.00	
ALPS-3_ferrocyanide	222	184	0.83	181	181	1.00	
ALPS-6_chelating resin	443	368	0.83	363	363	1.00	
ALPS-5_titanium oxide	222	184	0.83	181	181	1.00	
ALPS-7.1	46	57	1.25	68	68	1.00	
Rubble 1	190000	380000	2.00	34656	186873	5.39	
Rubble 2	48000	96000	2.00	30140	177951	5.90	
Rubble 3	26700	53400	2.00	6079	33705	5.54	
Rubble 4	52000	104000	2.00	20729	103528	4.99	
Rubble 5	36000	72000	2.00	19617	106797	5.44	
Felled trees	18000	14400	0.80	17264	13811	0.80	
Pressure vessel waste (equivalent to L1 before the accident)	230	216	0.94	231	222	0.96	
Pressure vessel waste (equivalent to L2 before the accident)	185	1466	7.91	185	1467	7.91	
Waste in the containment vessel (metal) (equivalent to L2 before the accident)	1164	873	0.75	1164	877	0.75	
Waste in the containment vessel (metal) (equivalent to L3 before the accident)	1211	1831	1.51	1211	1829	1.51	
Waste in the containment vessel (concrete) (equivalent to L2 before the accident)	431	328	0.76	431	329	0.76	
Waste in the containment vessel (concrete) (equivalent to L3 before the accident)	449	682	1.52	449	679	1.51	
Waste inside the building (metal) (equivalent to L3 before the accident)	23677	35677	1.51	23677	35677	1.51	
Waste inside the building (metal) (equivalent to nonradioactive waste before the accident)	6454	9725	1.51	6454	9725	1.51	
Waste inside the building (concrete) (equivalent to L3 before the accident)	8766	13208	1.51	8766	13208	1.51	
Waste inside the building (concrete) (equivalent to nonradioactive waste before the accident)	370586	558403	1.51	370586	558403	1.51	



Consolidation of input information (3/10)

#### <Radioactivity inventory>

The inventory that serves as input for the investigate of sensitivity analysis and disposal concept proposals was established, based on the outcome of characterization carried out in FY2020

- The radioactivity of 68 nuclides that have a major impact on disposal were established for 27 types of wastes (5 representative nuclides are listed below). Considering the uncertainty of the inventory, evaluation was carried out in two ways, one using the single most frequent value (mode) (using some of the average values) and another using the 95% itle.
- The amount of radioctivity from C-14 and I-129 from some of the secondary waste generated from contaminated water treatment (SARRY, KURION, AREVA, etc.) was changed substantially ⇒ It is assumed to mainly have an impact on exposure dose in the groundwater scenario.

			C	-14			Sr-	90			I-1	29			Cs	Cs-137 Pu-239					
	Amount of radioactivity	G	eomean_Mode		95%ile	Ge	omean_Mode		95%ile	G	eomean_Mode		95%ile	(	Geomean_Mode		95%ile	G	eomean_Mode		95%ile
	Comparison table of EY2020 and EY2021	2021 Year	Comparison with the	2021 Year	Comparison with the	2021 Year	Comparison with the	2021 Year	Comparison with the												
		(Bq)	previous year	(Bq)	previous year	(Bq) p	previous year	(Bq)	previous year	(Bq)	previous year	(Bq)	previous year								
			(FY2021 / FY2020)		(FY2021 / FY2020)		(FY2021 / FY2020)		(FY2021 / FY2020)												
1	KURION	9.6E+11	164.833	1.4E+12	90.202	9.1E+14	0.056	2.0E+17	0.687	5.8E+10	162.530	1.0E+11	106.176	+17.7E+1	1.001	+17.7E+1	1.000	+10.9E+1	117.334	8.0E+11	245.389
2	AREVA	6.0E+08	16.929	+09.9E+2	30.096	1.5E+16	51.440	3.8E+16	21.187	3.7E+07	16.696	+08.7E+1	30.162	1.0E+15	1.001	1.0E+15	1.000	5.7E+08	6.167	2.0E+11	20.349
3	SARRY	7.5E+11	180.367	1.1E+12	95.300	6.8E+16	5.170	1.6E+17	0.711	+10.6E+4	178.208	7.7E+10	115.615	1.3E+17	1.053	1.3E+17	1.053	1.4E+10	122.963	4.9E+11	247.027
4	Slurry 1_Iron co-precipitation	2.7E+11	32.652	2.3E+12	0.976	3.9E+16	33.374	2.0E+17	0.999	4.4E+08	0.850	1.3E+09	0.788	2.0E+12	0.162	+13.7E+1	0.001	1.1E+09	0.495	6.0E+11	0.586
5	Slurry 2_Carbonate precipitation	1.1E+11	1.685	1.4E+12	0.540	8.9E+15	1.881	1.1E+17	0.512	4.3E+08	0.751	1.3E+09	0.767	+11.9E+1	0.017	4.7E+12	0.000	5.4E+09	0.653	7.9E+11	0.610
6	ALPS-4_Ag impregnated activated carbon	1.1E+06	0.370	9.3E+06	0.411	8.2E+10	2.021	5.6E+11	0.605	4.2E+10	2.505	1.5E+11	0.847	7.8E+08	0.005	7.3E+09	0.001	4.0E+04	0.476	2.3E+06	0.553
-	11 DO A //					0.05 10	1.080			115.00		1.05.04		0.15.04		0.05.00		105.01		0.15.01	
7	ALPS-2_titanate	1.1E+06	0.388	8.8E+06	0.403	8.8E+12	1.876	1.1E+14	0.512	4.1E+05	0.762	1.3E+06	0.777	8.4E+08	0.006	8.2E+09	0.001	4.0E+04	0.512	2.4E+08	0.574
8	ALPS-3_terrocyanide	1.1E+06	0.381	9.0E+06	0.390	8.5E+07	520.448	6.2E+08	0.722	4.1E+05	0.761	1.2E+06	0.738	2.2E+10	0.002	+12.7E+1	0.000	40.E+04	335.516	2.4E+08	0.611
9	ALPS-6_chelating resin	1.0E+06	0.392	7.0E+06	0.313	7.8E+07	1.993	4.8E+08	0.629	4.0E+05	0.784	1.2E+06	0.724	7.8E+05	0.006	7.5E+06	0.001	2.2E+05	0.295	2.3E+08	0.568
10	ALPS-5_titanium oxide	1.1E+06	0.408	8.7E+06	0.386	7.8E+09	84993.994	5.2E+10	57.300	4.2E+05	0.816	1.3E+06	0.774	+07.7E+1	124.363	6.2E+08	0.049	1.2E+04	1185.287	8.1E+05	0.491
11	ALPS-7, 1	1.1E+06	0.391	8.7E+06	0.383	2.4E+07	1424.382	2.2E+08	0.240	4.3E+05	0.807	1.3E+06	0.789	2.3E+05	1.//2	3.6E+06	0.000	1.2E+04	9815.208	9.2E+05	0.583
12	Rubble 1	3.6E+08	0.007	+09.9E+2	0.012	1.4E+10	0.015	1.3E+11	0.011	6.0E+06	0.002	+08.7E+1	0.011	1.1E+13	0.017	1.1E+13	0.017	1.1E+06	0.008	+07.9E+1	0.004
13	Rubble 2	1.1E+11	0.697	9.1E+11	1.200	+12.5E+4	1.436	4.1E+13	1.042	+09.9E+1	0.210	5.4E+10	1.045	3.4E+15	1.678	3.5E+15	1.680	3.4E+08	0.817	6.0E+09	0.350
14	Rubble 3	1.2E+11	0.188	9.8E+11	0.334	4.9E+12	0.387	4.4E+13	0.281	2.1E+09	0.057	5.8E+10	0.330	3./E+15	0.452	3./E+15	0.453	3./E+08	0.220	6.5E+05	0.094
15	Rubble 4	1.8E+12	0.610	1.4E+13	4.756	7.1E+13	0.405	6.5E+14	0.294	3.0E+10	0.190	8.5E+11	4.096	5.4E+16	0.474	5.4E+16	0.474	5.4E+09	0.231	9.4E+10	0.099
16	Rubble 5	3.6E+12	1.242	+13.9E+2	9.680	1.4E+14	0.466	1.3E+15	0.338	6.1E+10	0.387	+12./E+1	8.336	1.1E+1/	0.544	1.1E+1/	0.545	1.1E+10	0.265	+11.9E+1	0.114
17	Felled trees	2.1E+11	0.753	9.9E+11	0./14	5.8E+12	1.102	4./E+13	0.621	1.2E+10	0./14	6.2E+10	0.656	3.9E+15	1.041	3.9E+15	1.042	3.3E+08	0.439	6.3E+05	0.219
18	Pressure vessel waste (equivalent to L1 before the accident)	+13.6E+4	1.000	+13.6E+4	1.004	4.4E+16	1.790	4.8E+16	1.013	3.1E+07	0.258	3.2E+10	1.413	8.2E+16	1.246	8.7E+16	0.984	2.2E+14	1.502	2.3E+14	0.960
19	Pressure vessel waste (equivalent to L2 before the accident)	5.4E+10	0.980	4.7E+11	1.386	4.0E+16	1.665	4.8E+16	1.024	1.4E+07	0.161	2.6E+10	1.417	6.6E+16	1.207	7.0E+16	0.984	2.1E+14	1.484	2.3E+14	0.965
20	Waste in the containment vessel (metal) (equivalent to L2 before the accident)	3.3E+10	0.824	4.9E+11	1.404	9.7E+15	1.441	1.2E+16	1.004	8.6E+07	0.170	+10.9E+2	1.419	7.3E+16	1.351	7.8E+16	0.976	5.4E+13	1.455	5.8E+13	3 0.953
21	Waste in the containment vessel (metal) (equivalent to L3 before the accident)	6.9E+10	0.905	5.5E+11	1.377	1.1E+16	1.647	1.3E+16	1.015	8.8E+07	0.168	3.0E+10	1.419	7.7E+16	i 1.353	8.1E+16	0.976	5.5E+13	1.479	5.8E+13	0.968
22	Waste in the containment vessel (concrete) (equivalent to L2 before the accident)	9.6E+08	0.275	+11.7E+1	1.451	9.0E+15	1.424	1.2E+16	1.008	3.4E+07	0.177	1.1E+10	1.412	2.7E+16	1.278	+16.9E+2	0.977	5.1E+13	1.378	5.8E+13	0.968
23	Waste in the containment vessel (concrete) (equivalent to L3 before the accident)	1.4E+09	0.339	1.8E+11	1.449	1.0E+16	1.630	1.2E+16	1.003	3.5E+07	0.176	1.1E+10	1.415	+16.9E+2	1.285	3.0E+16	0.976	4.9E+13	1.367	5.7E+13	3 0.971
24	Waste inside the building (metal) (equivalent to L3 before the accident)	1.3E+12	0.991	1.3E+12	0.976	8.5E+12	0.040	1.1E+15	0.182	3.0E+08	0.417	5.0E+08	0.199	9.3E+14	0.299	1.4E+15	0.257	2.8E+10	0.862	5.0E+11	0.344
25	Waste inside the building (metal) (equivalent to non-radioactive waste before the accident)	1.2E+09	0.355	2.0E+09	0.182	8.1E+12	0.142	2.6E+14	0.168	6.3E+07	0.316	1.2E+08	0.184	2.4E+14	0.281	3.5E+14	0.240	5.4E+08	0.431	1.3E+11	0.300
26	Waste inside the building (concrete) (equivalent to L3 before the accident)	1.6E+10	0.846	+10.7E+1	0.577	2.4E+14	3.194	3.1E+14	0.143	8.6E+07	0.314	1.4E+08	0.156	3.3E+14	0.282	4.3E+14	0.214	6.5E+08	0.380	1.6E+11	0.295
27	Waste inside the building (concrete) (equivalent to non-radioactive waste before the accident)	6.7E+10	0.361	1.1E+11	0.174	6.6E+14	0.211	1.6E+16	0.172	4.7E+09	0.407	6.8E+09	0.175	1.3E+16	0.280	+16.9E+1	0.229	2.7E+10	0.378	7.2E+12	2 0.346



Radioactivity in 2021 and 2020 is less than 0.5 Radioactivity in 2021 and 2020 is more than 2





Consolidation of input information (4/10)

#### <Set-up for the treatment>

During the analysis and evaluation carried out in FY2021, treatment methods were established for each waste based on the results of studies on treatments conducted until FY2020 and the weight and amount when waste conditioning is carried out was calculated.

#### Up to FY2020: Waste was evaluated*

*: Treatment methods were established and evaluated in some of the studies on typical waste.

FY2021: Weight and amount were set up considering treatment methods for each waste based on the results of studies on treatments conducted until FY2020



Raw waste

Weight and volume is the same as raw waste



Dehydration, etc.

Solidification, etc.

Set by multiplying an appropriate coefficient to the weight and volume of raw waste





#### Consolidation of input information (5/10)

#### <Values established for the volume and weight of waste>

Type of waste	Waste conditioning volume ratio	Waste conditioning weight ratio	Original volume (m ³ )	Original weight (t)	Waste body volume (m ³ )	Waste body weight (t)
KURION	1	2	2,877	11,685	2,877	5,753
AREVA	0.03	0.04	597	597	18	24
SARRY	1	2	1,274	5,520	1,274	2,548
Slurry 1_Iron co-precipitation	0.3	0.5	2,001	2,001	600	1,000
Slurry 2_Carbonate precipitation	0.3	0.4	8,610	8,610	2,583	3,444
ALPS-4_Ag impregnated activated carbon	0.5	0.5	181	181	91	91
ALPS-2_titanate	0.5	0.5	272	272	136	136
ALPS-3_ferrocyanide	0.5	0.5	181	181	91	91
ALPS-6_chelating resin	0.5	0.5	363	363	181	181
ALPS-5_titanium oxide	0.5	0.5	181	181	91	91
ALPS-7, 1	0.5	0.5	68	68	34	34
Rubble 1	3.7	2	34,656	186,873	129,089	373,747
Rubble 2	4.0	2	30,140	177,951	120,113	355,902
Rubble 3	3.8	2	6,079	33,705	23,114	67,410
Rubble 4	3.5	2	20,729	103,528	73,019	207,055
Rubble 5	3.8	2	19,617	106,797	73,588	213,594
Felled trees	1.3	3	17,264	13,811	22,443	41,434
RPV waste (L1 before the accident)	1	2	231	222	231	443
RPV waste (L2 before the accident)	5	2	185	1,467	927	2,934
PCV waste (metal) (L2 before the accident)	1	2	1,164	877	1,164	1,753
PCV waste (metal) (L3 before the accident)	1	2	1,211	1,829	1,211	3,658
PCV waste (concrete) (L2 before the accident)	1	2	431	329	431	658
PCV waste (concrete) (L3 before the accident)	1	1	449	679	449	679
Waste inside the building (metal) (L3 before the accident)	1	2	23,677	35,677	23,677	71,353
Waste inside the building (metal) (non-radioactive waste before the accident)	1	2	6,454	9,725	6,454	19,449
Waste inside the building (concrete) (L3 before the accident)	1	1	8,766	13,208	8,766	13,208
Waste inside the building (concrete) (non-radioactive waste before the accident)	1	1	370,586	558,403	370,586	558,403
Total			558,245	1,274,740	863,237	1,945,073





#### References

Consolidation of input information (6/10)

#### <The approach while establishing the values for the volume and weight of waste>

#### **①KURION / SARRY**

 $\Rightarrow$  The current weight has been calculated "including the radiation shielding container".

(KURION: 2877m³, 11685t, SARRY: 1274m³, 5520t)

 $\Rightarrow$  It is assumed that volume does not change and the density becomes 2t/m³, as an assumption for solidified substances.

2 AREVA / Iron co-precipitation slurry / Carbonate slurry

- $\checkmark$  Assumed that Dehydration, etc. treatment  $\rightarrow$  solidification would be conducted
- ✓ Value obtained by calculating the volume ratio and weight ratio assuming that the filling rate for solidified substances is 30% for waste (solid) (20% for iron co-precipitation slurry) and 70% for filler material (80% for iron co-precipitation slurry), based on the weight ratio and volume ratio of solid/liquid raw waste, and rounding it up.

Waste conditioning coefficient	AREVA	Iron co- precipitation slurry	Carbonate slurry		
Volume ratio of raw waste	0.03	0.3	0.3		
Weight ratio of raw waste	0.04	0.5	0.4		



⇒ As analytical data on raw waste is not available, based on results of iron co-precipitation slurry and carbonate slurry, the weight ratio and volume ratio of raw waste during waste conditioning was assumed to be 0.5.

#### (4) Rubble 1 to 5

⇒ The filling rate of metal rubble was assumed to be 20% (that is, the waste conditioning volume ratio: 5) and the filling rate of concrete rubble was assumed to be 50% (that is, the waste conditioning volume ratio: 2). Also, the waste conditioning weight ratio was considered to be 2 by conservatively (on the lower side) rounding up the weight ratio that includes the container and filler material. The waste conditioning volume ratio of Rubble 1 to Rubble 5 in which concrete and metal are combined, changed in the range of 3.5 to 4 based on an overall average, and changed in the range of 2 to 5 depending on the proportion of concrete and metal in individual waste bodies.

Evaluation Committee meeting

#### 5 Felled trees

⇒ ⇒ The amount of raw waste is the amount after incineration. Hence the filling rate of incineration ash was considered to be 30wt% and the density of solidified substance was considered to be 2t/m3, the waste conditioning weight ratio was set at 3 and the waste conditioning volume ratio was set at 1.3.



Secretariat meeting

Secretariat meeting



### **References** Consolidation of input information (7/10)

<The approach while establishing the values for the volume and weight of waste>

6 RPV / PCV waste and waste inside the building

- ⇒ Density was calculated based on the volume and weight of raw waste and was organized from the following perspectives.
- (i) Metal waste
- ✓ Density about 7.9t/m³ (Only RPV waste (L2 before the accident))
  - ⇒ Assuming a filling rate of 0.2 similar to that of metal rubble, the waste conditioning volume ratio was set at 5, and the waste conditioning weight ratio was set at 2.
- ✓ Density about 0.7 to 1.5t/m³ (Other than RPV waste (L2 before the accident))
  - ⇒ Assumed that it is stored in containers. The waste conditioning volume ratio was assumed to be 1, and the waste conditioning weight ratio was assumed to be 2.

(ii) Concrete waste

The waste conditioning volume ratio was assumed to be 1, and when the density was 1 or less, the waste conditioning weight ratio was considered to be 2, whereas when the density was 1 or more, the waste conditioning weight ratio was considered to be 1.



# (b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Consolidation of input information (8/10)

<Leaching rate and diffusion coefficient inside the waste body>

The diffusion coefficient inside the solidified substance and the leaching rate of the solidified substance were calculated for the leaching rate from glass, melt, cement, and AAM solidified substances, based on the outcome of studies on advance treatments up to FY2020.

	Diffusion dominated (When	it is assumed that leachin	Dissolution dominated**			
Nuclides*	Diffusion coefficient D (cm ² /s)	Leaching period (Years)	Leaching rate (1/y)	Leaching rate (1/y)	Leaching period for the entire amount (y)	
		1	1.6E-01		3.2E+00	
Carbonate slurry Cs	6.3E-08	100	Entire amount gets leached out in 41 years	3.1E-01		
Carbonata alurry Sr	10.75.1	1	8.2E-03	1 65 02	6.1E+01	
Carbonate sturry Sr	-10.72+1	100	8.2E-04	1.02-02		
Carbonata alumu En. Ca	6 0E 11	1	1.6E-04	2.25.04	3.0E+03	
Carbonate siurry Sn, Ce	0.9E-14	100	1.6E-05	3.3E-04		
Iron co-precipitation		1	1.3E-01		3.8E+00	
slurry Cs	4.4E-08	100	Entire amount gets leached out in 58 years	2.6E-01		
Iron co-precipitation	1 65 00	1	2.5E-02	4 0E 02	2.05.01	
slurry Sr	1.02-09	100	2.5E-03	4.9E-02	2.0E+01	
Iron co-precipitation	6 0E 11	1	1.6E-04	2.25.04	3.0E+03	
slurry Sn, Ce	0.9⊏-14	100	1.6E-05	3.3⊏-04		

Table 1 (Example) Leaching rate of each nuclide in cement (OPC)

*Corresponds to the solidification mentioned in ② on p281

** Since the results of a short (91 days) immersion test are extrapolated linearly, it is dissolution dominated. The leaching rate (when it is assumed that leaching is mainly due to diffusion) becomes conservative.

#### Table 2 (Example) Leaching rate of glass solidified substance

Evaluation temperature (°C)	Leaching speed (kg/m ² d)	Leaching rate (1/y)	Leaching period for the entire amount (y)
10°C	1.0E-08	1.6E-08	6.4E+07
25°C	5.0E-08	7.8E-08	1.3E+07
60°C	1.0E-06	1.6E-06	6.4E+05
90°C	1.0E-04	1.6E-04	6.4E+03





Consolidation of input information (9/10)

The set values of some of the distribution coefficients of FY2020 based on the application for changing the JNFL Lowlevel radioactive waste burial center, were reviewed while referring to the latest domestic disposal trend (NUMO Comprehensive Technical Report, etc.).

<Set values of distribution coefficient (Kd)>

Kd	Cement	Bentonite	Rock/Soil	Farm soil
Н	0	0	0	0
Be	0.1	0.004	0.01	3
С	0.004	0	0.0001	0.002
CI	5E-04	0	0	0.00025
Ca	0.009	0.01	0.01	0.11
Со	0.1	0.004	0.01	0.99
Ni	0.009	0.004	0.1	1.1
Se	0.01	0.01	0.001	1.8
Rb	0.01	0.1	0.1	0.67
Sr	0.009	0.01	0.01	0.15
Zr	0.1	0.1	0.02	7.3
Nb	0.1	0.1	0.02	2
Мо	0.01	0.0003	0.0001	0.027
Tc	0.0002	0	0.0001	0.0015
Pd	0.009	0.004	0.1	0.67
Ag	0.01	0.1	0.1	15
Cd	0.009	0.004	0.1	0.81
Sn	0.25	0.67	0.13	1.6
	0.0001	0	0	0.027
Cs	0.01	0.1	0.1	0.27

# The values in red indicate the values that were revised from last year

Kd	Cement	Bentonite	Rock/Soil	Farm soil
Ba	0.009	0.01	0.01	0.06
La	0.009	0.004	0.1	0.65
Pm	0.009	0.004	0.1	1
Sm	0.009	0.004	0.1	3
Eu	0.009	0.004	0.1	3
Tb	0.009	0.004	0.1	0.65
Ho	0.009	0.004	0.1	3
Pt	0.009	0.004	0.1	0.09
Pb	0.009	0.004	0.1	22
Po	0.009	0.004	0.1	6.6
Ra	0.009	0.01	0.01	2.4
Ac	0.1	1	0.1	5.4
Th	0.4	0.03	0.02	89
Ра	0.4	0.03	0.02	6.6
U	0.02	0.009	0.001	0.4
Np	0.009	0.004	0.0009	1.2
Pu	0.4	0.03	0.02	1.8
Am	0.1	1	0.1	110
Cm	0.1	1	0.1	12



#### References

Consolidation of input information (10/10)

The non-conservative set value was revised based on the NUMO Comprehensive Technical Report and JAEA database.

Status of setting the distribution coefficient Kd The values in red indicate the values that were revised from last year											
	FY2020 set value	NUMO Comprehensive Technical Report JAEA Database									
		Neogene period sedimentary rock	Neogene period sedimentary rock	e periodNeogene period entary sedimentary sedimentary Sandstone Sandstone ck rock rock				Rocks, soil			
Element	Rocks, soil (Based on the JNFL application for change)	Base case HWL, Gr.1, 2, 4 Low Cl  Concentration	Base case HWL, Gr.1, 2, 4 High Cl  Concentration	Base case Gr.3 (Up to 1000 years after closing of disposal site) Low Cl ⁻ Concentration	Base case Gr.3 (Up to 1000 years after closing of disposal site) High Cl ⁻ Concentration	Logarithmic mean	Lower limit value of the range with the largest quantity of data	revised based on the FY2020 JNFL application for change)			
Со	0.1	3	0.5	3E-4	5E-5	0.08357	0.01	0.01			
Ni	0.1	3	0.5	3E-4	5E-5	0.5422	0.1	0.1			
Se	0.15	0.04	0.05	0.04	0.05	0.02818	0.01	0.001 *			
Sr	0.1	0.2	0.07	2E-4	7E-5	0.01361	0.01	0.01			
Nb	0.02	6	6	6	6	1.774	0.1	0.02			
Sn	0.13	100	100	100	100	2.433	0.1	0.13			
Cs	0.9	1	0.1	0.001	1E-4	0.1916	0.1	0.1			
Th	0.02	30	5	30	5	0.3566	0.1	0.02			
U	0.001	6	2E-6	6	2E-6	0.2294	0.01	0.001			
Np	0.0009	30	5	30	5	0.02128	0.001	0.0009			
Pu	0.02	30	5	30	5	1.019	0.1	0.02			
Am	0.1	200	20	200	20	1.124	1	0.1			

*: The distribution coefficient for Se in rocks and soil is set conservatively at 0.001m³/kg considering that the numerical values under oxidizing conditions for sandstone are distributed in the range of 0.001 to 0.01m³/kg according to the JAEA database.





(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of sensitivity evaluation (1/15)

- Multiple disposal concepts with depths corresponding to trench disposal, pit disposal, and medium depth disposal were planned and models were created for sensitivity analysis. (Page 287 to 290)
- The parameters for sensitivity evaluation were set for each disposal concept. (Page 287 to 290)
- For the sensitivity evaluation, it was assumed that 1GBq nuclides are distributed evenly in the waste filler material, the outlet nuclide migration rate at each barrier (filler material, low diffusion layer, low permeability layer, natural barrier) was compared, and the attenuation in the nuclide migration rate once the nuclides have passed each barrier was evaluated. (Page 291 to 295)
- The sensitivity of the barrier composition and the sensitivity of the barrier parameters were organized. (Page 296 to 300)





(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of sensitivity evaluation (2/15)

#### <Overview of sensitivity evaluation>

The outlet nuclide migration rate at each barrier (filler material, low diffusion layer, low permeability layer, natural barrier) considered in groundwater migration in the case of trench disposal, pit disposal and medium depth disposal in the groundwater scenario, was compared, and the attenuation in nuclide migration rate once the nuclides have passed each barrier was understood. (Refer to Fig. 2)



#### Fig. 1 Image of evaluation model

(Note) The barrier composition illustrated in this diagram is an example. Models will be created according to the disposal concept (Refer to the following page onwards)

An indicator for discussing the necessity of the abilities required of barriers while evaluating each waste.



Fig. 2 Image of nuclide migration rate at each evaluation point

The peak nuclide migration rate after the nuclides pass through each barrier was evaluated.  $\rightarrow$  As there is delay during migration, the peak nuclide migration rate decreases due to broadening and attenuation



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(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of sensitivity evaluation (3/15)

<Study of sensitivity analysis cases (Disposal concept equivalent to trench disposal)> Trench disposal evaluation model (Fig. 1), disposal concept (Fig. 2), and sensitivity analysis parameters (Table 1) were established.



#### IRID

Set value

100

Set value

x 100

30

x 100

x 10

300

x 10

x 0.1

x 0.1

x 0.01

x 0.01

 $\mathbf{288}$ 

m³/kg

m

m³/ka

Distribution coefficient Kd

Migration distance

Distribution coefficient Kd

Natural barrier

(b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of sensitivity evaluation (4/15)

<Study of sensitivity analysis cases (Disposal concept equivalent to pit disposal)> Pit disposal evaluation model (Fig. 1), disposal concept (Fig. 2), and sensitivity analysis parameters (Table 1) was established.



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(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of sensitivity evaluation (5/15)

<Study of sensitivity analysis cases (Disposal concept equivalent to medium depth disposal)> Medium depth disposal evaluation model (Fig. 1), disposal concept (Fig. 2), and sensitivity analysis parameters (Table 1) were established.



Natural barrier

Migration distance

Materia

Distribution coefficient Kd

#### IRID

300

rock

Set value

100

x 100

900

x 10

x 0.1

m

m³/kg



x 0.01

(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of sensitivity evaluation (6/15)

<Sensitivity evaluation of barrier structure>

Sensitivity of each nuclide with respect to <u>impact of barrier structure</u> was evaluated for typical nuclides (result of studies from last year) that contribute to the dose from the groundwater evaluation picked out in Table 1. The nuclides were evaluated by grouping them as shown below.

- C-14, Tc-99, I-129: Long half-life (up to 5000y), delayed attenuation due to sorption is not expected as much
- Cs-135, Pu-239: Long half-life (up to 5000y), delayed attenuation due to sorption is expected to some extent
- Sr-90, Cs-137: Short half-life, delayed attenuation due to sorption is expected to some extent

		Distribution coefficient Kd (m ³ /kg)						
Nuclides	Half-life (y)	Cement	Bentonite	Rocks, soil, sand				
C-14	5700	0.004	0	0.0001				
Sr-90	29	0.009	0.01	0.01				
Tc-99	211100	0.0002	0	0.0001				
I-129	15700000	0.0001	0	0				
Cs-135	2300000	0.01	0.1	0.1				
Cs-137	30	0.01	0.1	0.1				
Pu-239	24110	0.4	0.03	0.02				

Table 1 Distribution coefficient of each nuclide for each barrier material Kd

Implementation of sensitivity evaluation (7/15)

<Evaluation of the impact of barrier structure in trench disposal*> The sensitivity to leaching rate, and controlling the amount of water penetration was evaluated, and it was verified that C-14, Tc-99, I-129 reduce due to leaching rate and because of controlling the amount of water penetration, but that there is almost no delay during migration (Fig. 2)



#### IRID

Trench disposal

I ow permeation of water

1 m lets

Earth's surfac

Evaluation point ①

Evaluation point (2)

C-14 Sr-90

Tc-99

I-129

Cs-135

Cs-137

Drainage lay

Implementation of sensitivity evaluation (8/15)

<Evaluation of the impact of barrier structure in trench disposal*>

The sensitivity to leaching rate, and controlling the amount of water penetration was evaluated, and it was verified that C-135, Pu-239 reduce due to leaching rate and because of controlling the amount of water penetration, and in addition, that there is attenuation due to sorption (In particular, the waste filled layer has a major impact) (Fig. 2).

*: In this document only trench disposal is introduced as a typical disposal method.





#### IRID

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Implementation of sensitivity evaluation (9/15)

<Evaluation of the impact of barrier structure in trench disposal*>

The sensitivity to leaching rate, and controlling the amount of water penetration was evaluated, and it was verified that Sr-90, Cs -137 reduce due to leaching rate and controlling the amount of water penetration, and that the peak largely reduces at each barrier due to delayed attenuation. (Fig. 2)

*: In this document only trench disposal is introduced as a typical disposal method. Amount of water penetration







Fig. 2 Results of evaluation of sensitivity to amount of water penetration and leaching rate

Fig. 1 Trench disposal evaluation model

Red: Parameters that were caused to fluctuate



(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of sensitivity evaluation (10/15)

Summary of evaluating sensitivity to barrier structure

O Nuclides with low sorption property and long half-life (C-14, Tc-99, I-129)

The engineered barrier is effective in controlling leaching and controlling the amount of water penetration.

O Nuclides with sorption property and long half-life (Cs-135, Pu-239)

- The engineered barrier is effective in controlling leaching and controlling the amount of water seepage.
- The engineered barrier and natural barrier are expected to lead to delayed attenuation to a certain extent.

O Nuclides with sorption property and short half-life (Sr-90, Cs-137)

Leaching and amount of water penetration from waste is expected to be controlled, but the effect of delayed attenuation due to the engineered barrier and natural barrier is dominant.



 (b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of sensitivity evaluation (11/15)

<Evaluation of sensitivity to the parameters of the barrier>

Sensitivity of each nuclide with respect to <u>the parameters of the barrier (amount of water penetration, leaching rate, confinement</u> <u>period, Kd of each barrier, migration distance)</u> was evaluated for typical nuclides (result of studies from last year) that contribute to the dose from the groundwater evaluation picked out in Table 1.

The nuclides typical to each group were evaluated as shown below.

- C-14 : Low sorption property, long half-life nuclide. Half life is long (up to 5000y), not much delayed attenuation is expected due to sorption.
- Cs-135: Sorption property, long half-life nuclide. Half life is long (up to 5000y), some delayed attenuation is expected due to sorption.
- Sr-90: Sorption, short half-life nuclide. Half-life is short, some delayed attenuation is expected due to sorption.

Nuolidoo	Half life (v)	Distribution coefficient Kd (m ³ /kg)					
nuclides	Hall-life (y)	Cement	Bentonite	Rocks, soil			
C-14	5700	0.004	0	0.0001			
Sr-90	29	0.009	0.01	0.01			
Tc-99	211100	0.0002	0	0.0001			
I-129	15700000	0.0001	0	0			
Cs-135	2300000	0.01	0.1	0.1			
Cs-137	30	0.01	0.1	0.1			
Pu-239	24110	0.4	0.03	0.02			



Implementation of sensitivity evaluation (12/15)

<Evaluation of the impact of barrier parameters in trench disposal*> The sensitivity to barrier parameters was evaluated, and it was found that amount of water penetration, filler material Kd, leaching rate, damage period have a major impact on C-14. (Fig. 2)

*: In this document typical examples of trench disposal and medium depth disposal are introduced.



Fig. 2 Results of evaluating the sensitivity to each parameter



Drainage layer

g (Low permeation of water as compa surrounding areas

Waste Jav

Flow of groundwat

hielding structure as shown in the above figure

trench disposal

Evaluation point (3) After migratine from the natural barrier

Fig. 1 Conceptual diagram of

1 m³/y/m Earth's surface

Evaluation point (1

Evaluation point (2)

C-14

Sr-90

Tc-99 I-129

Cs-135

Cs-137

Pu-239

x 0.01

0.0001

10000

10000

x 0.1 x 0.01

2

x 0.1

x 0.1

x 0.01

x 0.01

Lon Vivino Earth's surface

Evaluation point (1)

Evaluation point (2)

Flow of groundwater

(Low permeation of water as compared to

Evaluation point (3) After migrating from the natural barrier

Waste lav

#### Implementation of sensitivity evaluation (13/15)

<Evaluation of the impact of barrier parameters in trench disposal*> The sensitivity to barrier parameters was evaluated, and it was found that amount of water penetration, leaching rate, leaching rate, confinement period damage period have a major impact on Sr-90. (Fig. 2)

*: In this document typical examples of trench disposal and medium depth disposal are introduced.



Fig. 2 Results of evaluating the sensitivity to each parameter

C-14

Sr-90

Tc-99

I-129

Cs-135

Cs-137

Pu-239

Implementation of sensitivity evaluation (14/15)

<Evaluation of the impact of barrier parameters in medium depth disposal*>

The sensitivity to barrier parameters was evaluated, and it was found that amount of water penetration and filler material Kd, have a major impact on C-14. (Fig. 2)

*: In this document typical examples of trench disposal and medium depth disposal are introduced.



Fig. 2 Results of evaluating the sensitivity to each parameter



Evaluation of the natural barrier outlet (Evaluation point ④) (Refer to Fig. 1)

is the reference case

 ${\ensuremath{ \bullet} }$  : Cases that do not consider controlling water penetration

(The permeability coefficient (1E-7) for the area around the facility is considered to be the same as that for rocks.)

#### Table 1 List of sensitivity analysis parameters (Repeated)

			Initial value	Sensit	ivity param	eter/magnif	ication
	Facility configuration	L(m)	100				
		W(m)	100				
		H(m)	5				
	Amount of water penetration	m ³ /y/m ²	0.16, 3.2E-03	x 10	x 0.1	x 0.01	
Waste	Nuclide leaching rate	1/y	1	1.E-02	1.E-03	1.E-04	
	Confinement period	у	0	100	1000	10000	
	Damage period	у	0	100	1000	10000	
Filler material	Filler material ingredients		Cement type				
	Distribution coefficient Kd	m ³ /kg	Set value	x 100	x 10	x 0.1	x 0.01
Low diffusion layer	Material		Cement type				
	Thickness	m	0.5	0			
	Diffusion coefficient	m²/y	1E-11				
Bottom barrier layer	Material		bentonite				
	Thickness	m	1	0	0.5	2	
	Distribution coefficient Kd	m ³ /kg	Set value	x 100	x 10	x 0.1	x 0.01
Natural barrier	Migration distance	m	300	100	900		
	Material		rock				
	Distribution coefficient Kd	m ³ /kg	Set value	x 100	x 10	x 0.1	x 0.01

····Parameters to which the nuclides are highly sensitive



# Implementation of sensitivity evaluation (15/15) Summary of evaluating sensitivity to barrier parameters

The sensitivity of each nuclide to the barrier parameters was evaluated, and the parameters to which each nuclide is highly sensitive were identified. (Table 1)

	Trench	Pit	Medium depth
Nuclides with low sorption property and long half-life (C-14 was evaluated as a typical nuclide)	Amount of water penetration Filler material Kd Leaching rate Damage period	Amount of water penetration Filler material Kd Leaching rate Damage period	Amount of water penetration Filler material Kd
Nuclides with sorption property and long half-life (Cs-135 was evaluated as a typical nuclide)	Amount of water penetration Filler material Kd Leaching rate	Amount of water penetration	— (Does not become the controlling nucide even if there is no engineered barrier)
Nuclides with sorption property and short half-life (Sr-90 was evaluated as a typical nuclide)	Amount of water penetration Leaching rate Confinement period Damage timing	— (Does not become the controlling nucide even if there is no engineered barrier)	— (Does not become the controlling nucide even if there is no engineered barrier)

## Table 1 Parameters to which the nuclides are highly sensitive

## IRID

Preliminary proposals of disposal concepts (1/8)

- The method for deriving the disposal category for each waste based on the dose obtained through sensitivity evaluation was studied. (Page 302 to 303)
- Preliminary proposals of disposal concepts for each waste were consolidated on receiving the results of sensitivity analysis. (Page 304 to 308)

(b) Provision of disposal methods and development of safety assessment methods

[1] Information organization to investigate the disposal concepts and to establish safety assessment methods

Preliminary proposals of disposal concepts (2/8)

The disposal category for each waste was studied using the dose evaluation methods studied until last year (In the table below, there is no engineered barrier and the inventory is the MODE value). The 12 wastes selected until last year were studied.

In the case of waste for which the disposal category is determined based on the nearing scenario, even if an engineered barrier is installed, it is difficult to revise the disposal category. Hence, wastes for which disposal category is determined based on the groundwater scenario when there is no engineered barrier, were identified. (Refer to Table 1) Table 1 Results of investigating disposal categories

					_		•		
		Nearing	scenario		Groundwater scenario (without barrier)				Waste for which the disposal
			Control period						category is determined by groundwater evaluation
Waste name		400y	400y	100,000y					
	Disposal category	L3	L2	L1	Disposal category	L3	L2	L1	
KURION	L1	2.1E+05	8.8E+04	1.5E+03	L1-strata	2.3E+03	6.9E+01	9.3E+00	<ul> <li>Image: A set of the set of the</li></ul>
AREVA	L1	6.9E+06	2.6E+06	4.3E+03	L2	7.5E+03	4.3E-02	5.8E-03	
SARRY	L1	7.2E+05	3.0E+05	2.7E+03	L1-strata	3.3E+04	5.4E+01	7.3E+00	~
Slurry 1_iron co-precipitation	L1	4.7E+05	+05.9E+1	6.9E+02	L1-strata	+04.9E+1	+01.9E+1	2.6E+00	1
Slurry 2_Carbonate precipitation	L1	2.7E+04	1.0E+04	4.8E+01	L2-strata	4.3E+03	7.9E+00	1.1E+00	1
Rubble 1	L3	3.0E-01	1.4E-01	8.4E-03	L3	8.7E-01	2.6E-02	3.5E-03	
Rubble 2	L3	1.0E+02	4.7E+01	+00.9E+2	L2 - strata	2.8E+02	8.2E+00	1.1E+00	1
Rubble 5	L1	5.3E+03	2.5E+03	1.5E+02	Strata	8.8E+03	2.6E+02	3.5E+01	1
RPV waste (L1 before the accident)	Strata	9.1E+07	3.8E+07	4.0E+06	Strata	1.1E+05	3.3E+03	4.4E+02	
PCV waste (metal) (L2 before the accident)	Strata	+06.6E+4	1.8E+06	2.0E+05	L1	4.7E+03	3.1E+01	6.5E-01	
Waste inside the building (metal) (L3 before the accident)	L2	7.0E+02	3.3E+02	3.8E+00	Strata	3.2E+03	9.4E+01	1.3E+01	1
Waste inside the building (Concrete) (Non-radioactive waste before the accident)	L3	+02.9E+1	7.2E+01	8.0E+00	L2 - L1	3.2E+02	4.8E+00	6.5E-01	<i>✓</i>
L3         L2           300         1000         20	ring scenario	Dose criteria in the gr 10	undwater scenario µSv/y	For waste the scenario, converse of the scenario, converse the scenario, converse that e	hat does not onsidering th exceeds the	exceed the weight of approximat	dose criter other nuclic e dose of 1	ia in the gro les and was JSv/h are co	undwater te, the blor coded

red.

Exceeds the dose criteria

*: Disposal category with the least depth wherein dose criteria is satisfied in the nearing scenario and approximate dose is satisfied in the groundwater scenario.



(Unit: uSv/v)

### Preliminary proposals of disposal concepts (3/8)

If there is no engineered barrier, in the case of waste for which the disposal category is determined based on groundwater scenario, barriers or barrier performance that can control the dose up to approximate dose: 1µSv/y should be selected. The method for selecting barriers or barrier performance for Rubble 5 as an example of relatively low dose rubble, and for KURION as an example of high dose waste generated from contaminated water treatment is provided from next page onwards.

Table 1	Dose
---------	------

#### and main nuclides in each disposal concept

(Unit:  $\mu Sv/y$ )

	Nearing		Groundwater scenario Person using rivers Without barrier							
	scenario	L3				L2		L1		
Waste name	Disposal category	Nuclides	1/10 <	Dose	Nuclides	1/10 <	Dose	Nuclides	1/10 <	Dose
KURION	L1	C14	Sr90	2.3E+03	C14	1129	6.9E+01	C14		9.3E+00
AREVA	L1	Sr90		7.5E+03	C14	Se79, I129	4.3E-02	C14	Se79	5.8E-03
SARRY	L1	Sr90		3.3E+04	C14	1129	5.4E+01	C14		7.3E+00
Slurry 1_Iron co-precipitation	L1	Sr90		+04.9E+1	C14		+01.9E+1	C14		2.6E+00
Slurry 2_Carbonate precipitation	L1	Sr90		4.3E+03	C14		7.9E+00	C14		1.1E+00
Rubble 1	L3	C14		8.7E-01	C14		2.6E-02	C14		3.5E-03
Rubble 2	L3	C14		2.8E+02	C14		8.2E+00	C14		1.1E+00
Rubble 5	L1	C14		8.8E+03	C14		2.6E+02	C14		3.5E+01
RPV waste (L1 before the accident)	Strata	C14	Sr90	1.1E+03	C14		3.3E+03	C14		4.4E+02
PCV waste (metal) (L2 before the accident)	Strata	Sr90	Pu240, Pu239 Pu238, Am241	4.7E+03	Pu239	Pu240	3.1E+01	Pu239	Np237, C14 U234, Po210	6.5E-01
Waste inside the building (metal) (L3 before the accident)	L2	C14		3.2E+03	C14		9.4E+01	C14		1.3E+01
Waste inside the building (concrete) (non-radioactive waste before the accident)	L3	Sr90	C14	3.2E+02	C14	1129	4.8E+00	C14		6.5E-01

Disposal concept in the groundwater scenario that exceeds approximate dose: 1µSv/y from among the disposal concepts in which disposal is carried out at a depth that is shallower than the nearing scenario



Preliminary proposals of disposal concepts (4/8)

<Application of sensitivity evaluation to KURION>

In the nearing scenario the disposal category of KURION is medium depth disposal, but if disposal category is medium depth disposal in the groundwater scenario, the approximate dose (1µSv/y or lower) is exceeded. (Refer to Table1)

The dominant nuclide in the groundwater scenario is C-14. The barrier or barrier performance for reducing the dose of  $9.3\mu$ Sv/y to the approximate dose or lower, should be studied based on the sensitivity evaluation results. (Refer to Fig. 1)

	I able I	Dose t	or each dis	sposal con	cept in the	groundwat	er scenario	)		
	Nearing		Groundw	ater scenaric	Person us	ing rivers (V	Vithout barrie	r) Target dos	e 1µsV/y	
	Scendilo	L3 L2 L1								
Waste name	Disposal category	Nuclides	1/10 <	Dose	Nuclides	1/10 <	Dose	Nuclides	1/10 <	Dose
KURION	L1	C14 Sr90 2.30E+03 C14 I129 6.90E+01 C14						9.30E+00		

Kurion

"Dominant nuclides ingroundwater scenario: C-14" "Dose in groundwater scenario: 9.3µSv/y (C-14)"



#### Preliminary proposals of disposal concepts (5/8)

#### <Application of sensitivity evaluation to KURION>

In the case of KURION, in order to reduce the dose of  $9.3\mu$ Sv/y to the approximate dose:  $1\mu$ Sv/y or lower, based on the results of sensitivity evaluation, the amount of water penetration at the bentonite layer needs to be kept at about  $3.2E-3(m^2/ym^2)$  or lower. (Fig. 1) (Water penetration control barrier is essential.) Since even the sensitivity of concentration reduction at the filler material due to sorption is comparatively high, it is determined that it is important to take actions that would not have an adverse effect on filler Kd, such as limiting the materials with impact on disposal, etc. (Fig. 2)

Table 1





Fig. 2 Dependence on waste and filler material Kd

Change in dose due to change in the amount of water penetration and nuclide migration rate

	No barrier	Referen ce	Target
Amount of water penetration (m ³ /y/m ² )	0.16	0.0032	0.0032
Nuclide migration rate (Bq/y)	6.5E+5	7.0E+4	7.0E+4
Dose (µSv/year)	9.3	1.0	1.0

In order to keep the dose equivalent to  $1\mu$ Sv/year or lower (approx. 1/10 or lower), the amount of water penetration at the bentonite layer needs to be kept at about 3.2E-3 (m³/y/m²) (permeability coefficient 1E-10m/s) or lower.



(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Preliminary proposals of disposal concepts (6/8)

<Application of sensitivity evaluation to Rubble 5>

In the nearing scenario the disposal category of Rubble 5 is medium depth disposal, but if disposal category is medium depth disposal in the groundwater scenario, the approximate dose (1µSv/y or lower) is exceeded. (Refer to Table 1)

The dominant nuclide in the groundwater scenario is C-14. The barrier or barrier performance for reducing the dose of  $35\mu$ Sv/y to the approximate dose: 1  $\mu$ Sv/y or lower, should be studied based on the sensitivity evaluation results. (Refer to Fig. 1)

	Nearing	(	Groundwater	scenario	Person u	Person using rivers (Without barrier) Target dose 1µSv/y				
	scenario		L3		L2			L1		
Waste name	Disposal category	Nuclides	1/10 <	Dose	Nuclides	1/10 <	Dose	Nuclides	1/10 <	Dose
Rubble 5	L1	C14		8.80E+03	C14		2.60E+02	C14		3.50E+01

# Table1

Dose for each disposal concept in the groundwater scenario

#### Rubble 5

Dominant nuclides in groundwater scenario: C-14 Dose in groundwater scenario: 35µSv/y (L1)



Fig. 1 Time dependence of dose when there is no L1 barrier

(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Preliminary proposals of disposal concepts (7/8)

<Application of sensitivity evaluation to Rubble 5>

In the case of Rubble 5, in order to reduce the dose of  $35\mu$ Sv/y to the approximate dose:  $1\mu$ Sv/y or lower, based on the results of sensitivity evaluation, the amount of water penetration at the bentonite layer needs to be kept at about 1E-3(m²/y/m²) or lower (Fig. 1) (Water penetration control barrier is essential). Since even the sensitivity of concentration reduction at the filler material due to sorption is comparatively high, it is determined that it is important to take actions that would not have an adverse effect on filler Kd, such as limiting the materials with impact on disposal, etc. (Fig. 2)



 Table 1 Change in dose due to change in the amount of water penetration

 and nuclide migration rate

		No barrier	Reference	Target
	Amount of water penetration (m ³ /y/m ² )	0.16	0.0032	0.001 (Low diffusion barrier present)
	Nuclide migration rate (Bq/y)	6.5E+5	7.0E+4	2E+4
~ *	Dose (µSv/year)	35	3.8	1

Controlling the amount of water penetration has a major dose reduction effect.



Preliminary proposals of disposal concepts (Summary) (8/8)

Preliminary proposals of disposal concepts for each waste on receiving the results of sensitivity analysis are indicated in Table 1.

Waste name	Disposal category preliminary proposal	Scenario for determining the disposal category	Dominant nuclides in nearing scenario	Dominant nuclides in groundwater scenario	Water penetration control barrier	Amount of water penetration (m³/y/m²)	Filler material Kd (*2) (m³/kg)
KURION	L1	Ground water	I-129, Cs-135 (>10%)	C-14, I-129 (>10%)	Required	3E-3 or less	0.004 or more
AREVA	L1	Nearing	Se-79, Tc-99 (>10%)	C-14, Se-79 (>10%)	Not required	-	0.004 or more
SARRY	L1	Ground water	I-129, Cs-135 (>10%)	C-14, I-129 (>10%)	Required	3E-3 or less	0.004 or more
Slurry 1_iron co-precipitation	L1	Ground water	Se-79, Tc-99 (>10%)	C-14, I-129 (>1%)	Required	3E-3 or less	0.004 or more
Slurry 2_Carbonate precipitation	L1	Ground water	Se-79, Tc-99 (>10%)	C-14, I-129 (>1%)	Required	3E-3 or less	0.004 or more
Rubble 1	L3	Nearing / Groundwater	Cs-137, Sr-90 (<1%)	C-14, I-129 (>1%)	Not required	-	0.004 or more
Rubble 2	L2	Groundwater	Cs-137, C-14 (>1%)	C-14, I-129 (>1%)	Required	1E-2 or less	0.004 or more
Rubble 5	L1	Groundwater	Tc-99, Cl-36 (>10%)	C-14, I-129 (>1%)	Required	1E-3 or less	0.004 or more
RPV waste (L1 before the accident)	Strata (*1)	Nearing	Tc-99, Ra-226 (>10%)	C-14, TRU (up to 10%)	Required	1E-4 or less	0.004 or more
PCV waste (Metal) (L2 before the accident)	Strata (*1)	Nearing	Tc-99, Ra-226 (>10%)	TRU	Not required	-	-
Waste inside the building (Metal) (L3 before the accident)	L1	Ground water	C-14, Cs-137 (>10%)	C-14, I-129 (<1%)	Required	3E-3 or less	0.004 or more
Naste inside the building (Concrete) Non-radioactive waste before the accident)	L2	Nearing / Groundwater	Cs-137, Tc-99 (>10%)	C-14, I-129 (>10%)	Required	1E-2 or less	0.004 or more

## Table 1 Preliminary proposals of disposal concepts for each waste

*1: In this evaluation, waste that cannot be disposed at L1 is tentatively assumed to be disposed geologically. L1 values are provided for dominant nuclides in the groundwater scenario.

*2: Filler material Kd for dominant nuclides in groundwater scenario





## Implementation of dose evaluation (1/14)

- Dose evaluation was carried out for the disposal concept proposals obtained as a result of sensitivity evaluation. (Only groundwater scenario) (Page 310 to 318)
- Dose evaluation was conducted using 2 models, namely, "Conservative conceptual model (Analysis model similar to the models that have a domestic track record)" and "Cautiously realistic conceptual model" using the same parameters as the sensitivity evaluation, and the results were compared. (Page 310 to 318)
- Based on the results of dose evaluation using the conservative model (Analysis model similar to models that have a domestic track record), it was projected that the disposal concept that was determined through sensitivity evaluation was generally satisfactory when its dose was evaluated. (Page 319 to 321)
- According to the results of dose evaluation using the cautiously realistic conceptual model, the maximum dose showed a decline. It was verified that the components of the disposal concepts have a major impact on dose evaluation, in particular when diffusion of waste / backfill and ageing degradation are considered. (Page 317 to 318)



 (b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of dose evaluation (2/14)

- Dose evaluation was carried out for the disposal concept proposals obtained as a result of sensitivity evaluation. (Only groundwater scenario)
- Dose evaluation was conducted using 2 models, namely, **"Conservative conceptual model (Analysis model similar to the models that have a domestic track record)**" and **"Cautiously realistic conceptual model"** using the same parameters as the sensitivity evaluation, and the results were compared. (Refer to Fig. 1)



Fig. 1 Comparison of the broad outline of the dose evaluation models



 (b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of dose evaluation (3/14)

•Based on the results of investigation of overseas disposal cases carried out from FY2017 - FY2019, it was decided to use the cautiously realistic conceptual model for dose evaluation on a trial basis.

•The cautiously realistic conceptual model adopts an approach that is based on safety cases from the UK. It consists of "Observation", "Process model", "Numerical model", and "Data elicitation" and is updated in accordance with updates in safety cases based on latest findings. (Refer to Fig. 1)

## Observation

## Numerical model



IRID

Implementation of dose evaluation (4/14)

When dose evaluation is conducted, while a conservative evaluation that includes uncertainties can be conducted if a conservative set of parameters and a conservative model are used, the resulting dose is likely to be excessive. (Likely to have an impact on the required disposal depth and concept) (Refer to Fig. 1)

•Hence, the evaluation was carried out using both, the conservative model and the cautiously realistic model, and the impact due to the differences in the models was compared.



Fig. 1 Impact of the difference in evaluation models on the results of dose evaluation (Image)



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(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of dose evaluation (5/14)

•Conservative model and cautiously realistic model were arranged respectively for each component (low permeability layer, waste bodies, external wall of the facility, etc.) of the disposal facilities conducting dose evaluation.

• Dose evaluation was carried out for multiple cases in accordance with the combination of models for each component (barrier).

Dose evaluation cases (Example of L2 disposal facility) (Refer to Fig. 1)

·Setting all components in the conservative model

•Setting the low permeability layer in the cautiously realistic model and all the other components in the conservative model

•Setting the waste layer in the cautiously realistic model and all the other components in the conservative model

·Setting all components in the cautiously realistic conceptual model





Etc.

## Implementation of dose evaluation (6/14)

• The difference in the conservative model and cautiously realistic model in the case of the low permeability layer is shown in Fig. 1. • The changes in infiltration ratio due to ageing degradation of the low permeability layer are taken into consideration in the cautiously

realistic conceptual model.





(b) Provision of disposal methods and development of safety assessment methods
 [1] Information organization to investigate the disposal concepts and to establish safety assessment methods
 Implementation of dose evaluation (7/14)

•The difference in the conservative model and cautiously realistic model in the case of waste bodies is shown below.

•Ageing degradation of the container and delay in diffusion of each nuclide in the waste body are incorporated in the cautiously realistic conceptual model.



Fig. 1 Comparison of the condition of waste bodies in the conservative model and cautiously realistic conceptual model

Instant degradation of container

- Instant mixing of water and waste
- •Rapid emission of radioactive nuclides
- Continuous and constant speed process

Ageing degradation of container

- Consideration of cracks in cement
- ·Sequential emission of radioactive nuclides
- Time dependent process

## Fig. 2 Characteristics of each model



# (b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of dose evaluation (8/14)

• The impact of differences in the models (conservative model / cautiously realistic conceptual model) on the dose evaluation results is indicated below.

• Decline in dose was seen at Slurry-1 at L2 when time dependency of the low permeability layer was considered. (Table 1)

Table 1Results of dose evaluation with L2 as the disposal concept proposal and Slurry-1 as the target waste (iron co-precipitation)

Waste name:	Slurry_1						[					
Disposal concep	1 L2b											
Calculation				Well		River		Sea				
Realization	case	Scenario		Peak dose (uSv/v/h	Time (vr)	Key RN	Peak dose ((Sv/vr)	Time (vr)	Key RN	Peak (uSv/yr)	Time (yr)	Key RN
1	Pb_Ref	Reference case (conversative)	Conservati	ve model	333	Sr-90	1.27E+01	118	C-14	6.70E-01	238	C-14
2	Pb_2	Waste and backfill controlled variant (con	servative Kd)	9.63E-02	70000	Se-79	9.21E-03	6220	C-14	7.10E-04	5790	C-14
3	Pb_3	Waste and backfill controlled variant (cautiously realistic Kd)		8 27E-03	8530	1-129	9.15E-03	6720	C-14	5.99E-04	6850	C-14
4	Pb 4	Container Considering the	time depende	ency of the	e low r	bermea	bility lave	er ¹⁸¹	C-14	4.96E-01	762	C-14
5	Pb_5	Buffer controlico variant		8.1/6100	0010	00-18	1.044-100	850	C-14	1.19E-01	1980	C-14
6	Pb_6	Engineered vault controlled variant		8.83E+00	2920	Se-79	1.81E+00	1820	C-14	1.26E-01	2000	C-14
7	Pb_7	Integrated (conservative Kd)		9.60E-02	74000	Se-79	8.37E-03	7030	C-14	6.57E-04	5170	C-14
8	Pb_8	Integrated (cautiously realistic Kd)		8.08E-03	9250	I-129	8.32E-03	7540	C-14	5.46E-04	7670	C-14



Fig. 1 Temporal changes in the infiltration ratio of the low permeability layer Impact of using the cautiously realistic conceptual model for the low permeability layer

(In the case of the scenario in which river is used)

•Decline in maximum dose is seen

•The time period when the dose is maximum is delayed about 1700 years



Physical delay has a major impact

(The period of 5000 years by when degradation of the low permeability layer is assumed to happen, is equivalent to the half-life of the main nuclide C-14 (Fig. 1)





Implementation of dose evaluation (9/14)

•As a result of dose evaluation, the highest dose reduction effect was seen when the cautiously realistic conceptual model was used for the waste bodies and backfill.

 $\rightarrow$  The evaluation of waste body performance with respect to Slurry-1 (iron co-precipitation) is likely to have a major impact on the disposal safety evaluation.

(Further, in the case in Fig. 1, the analysis was conducted using the same value as Kd (conservative value, refer to P.284))



IRID

# (b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of dose evaluation (10/14)

Confinement of C-14 and Pu-240 into the waste body is seen when diffusion inside the waste body is considered.
Confinement of I-129 into the backfill is seen due to the effect of delayed diffusion.

(Further, in the case in Fig. 1, the analysis was conducted using the same value as Kd (conservative value, refer to P.284))





Implementation of dose evaluation (11/14)

• It was projected that the disposal concept proposal L3 would be suitable for "Rubble 1" even when the evaluation was carried out using the conservative model. (Refer to Table 1)

•In the case of "Waste inside the building (Concrete) (Non-radioactive waste before the accident)", the maximum dose was high when the conservative model was used. Hence it is believed that dose evaluation needs to be carried out using a model in which ageing degradation of the cap, etc. is appropriately evaluated. (Refer to Fig. 1)

•Results can be said to be the same as The investigation of preliminary proposals for the disposal concepts



#### IRID

It was projected that the disposal concept proposal L2 would be suitable for "AREVA" and "Rubble 2" even when the evaluation was carried out using the conservative model. (Refer to Table 1)

•As "Rubble 2" is close to the dose criteria in the Ref case, it is believed that the amount of water penetration and the leaching at the waste body layer need to be controlled.

• The maximum dose is high when the conservative model is used for the "Waste inside the building (Metal) (L3 before the accident)" and "Slurry 1 iron co-precipitation". L2 is difficult to accomplish even in the preliminary proposal evaluation. Hence an L1 concept that includes barriers is necessary.

In order to dispose these at L2, it is believed that dose evaluation needs to be carried out using a model that appropriately evaluates ageing degradation and diffusion, etc. of the waste bodies and backfill.

Table 1

		Case		Scenario in which river is used			
0.05 m ^{3/} y/m ² = 500 m ³ /y/10000 m ²	Waste		Model conditions	Maximum dose (µSv/y)	Time period when the dose is the maximum (y)	Main nuclides	
Soil covering T Ground water level	AREVA*	Pa_Ref	Conservative model	3.00E-02	108	C-14	
Filler mortar Low permeability layer (Bentonite)		Pa_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	2.48E-04	97000	Cs-135	
Waste body	Waste inside the building (Metal) (L3 before the accident)	Pa_Ref	Conservative model	6.20E+01	119	C-14	
concrete) Bedrock (Internal partition) Concrete pit		Pa_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	4.38E-02	6630	C-14	
	Slurry-1 Iron co-precipitation*	Pa_Ref	Conservative model	1.27E+01	118	C-14	
Evaluation point ③ After migrating from the natural barrier		Pa_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	9.21E-03	6220	C-14	
Fig. 1 Disposal concept proposal L2	Rubble 2	Pa_Ref	Conservative model	5.42E+00	116	C-14	
		Pa_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	6.62E-03	2350	I-129	

Summary of dose evaluation with respect to the disposal concept proposal L2

*: Waste for which disposal concepts for disposal deeper than L1 are used in the nearing scenario



(Porous concre

Fig.

# (b) Provision of disposal methods and development of safety assessment methods [1] Information organization to investigate the disposal concepts and to establish safety assessment methods Implementation of dose evaluation (13/14)

 It was projected that the disposal concept proposal L1 which includes engineered barriers can be used for "SARRY", "Slurry-2" and "PCV waste (metal)(L2 before the accident)" even when the evaluation was carried out using the conservative model. (Refer to Table 1)

• The maximum dose in "KURION", "Rubble-5" and "RPV waste (L1 before the accident)" is high when penetration is not controlled. Hence it is believed that dose evaluation needs to be carried out using a model that appropriately evaluates ageing degradation and diffusion, etc. of the low permeability layer, waste bodies and backfill.



Fig. 1 Disposal concept proposal L1

			Scenario in which river is used					
Waste	Case	Model conditions	Maximum dose (µSv/y)	Time period when the dose is the maximum (y)	en Main nuclides e			
KURION	I_Ref**	Conservative model	1.15E+01	1380	C-14			
	I_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	1.42E-01	2740	I-129			
SARRY	I_Ref**	Conservative model	9.06E+00	1380	C-14			
	I_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	1.11E-01	2750	I-129			
Slurry-2	I_Ref**	Conservative model	1.30E+00	1390	C-14			
	I_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	3.31E-03	6700	C-14			
Rubble 5	I_Ref**	Conservative model	4.31E+01	1390	C-14			
	I_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	1.88E-01	3250	I-129			
PCV waste (Metal) (L2 before the accident)*	I_Ref**	Conservative model	1.73E+00	5760	Np-237			
	I_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	8.70E-02	180000	Ra-226***			
RPV waste (L1 before the accident)*	I_Ref**	Conservative model	5.40E+02	1390	C-14			
	I_2	Cautiously realistic conceptual model (Waste bodies / Backfill)	1.32E+00	8030	C-14			

Table 1 Summary of dose evaluation with respect to the disposal concept proposal L1

*: Waste for which disposal concepts with disposal at L1 are difficult in the nearing scenario as well

**: Controlling the amount of water penetration through the low permeability layer is not considered in I_Ref

***: Ra-226 has been formed through migration of U. Hence the contribution of Rn-222 is not considered.



> Implementation of dose evaluation (14/14) Summary of dose evaluation

- Based on the results of dose evaluation of the disposal concepts that were determined through sensitivity evaluation, it was projected that the disposal concepts were generally satisfactory.
   Further, the barrier compositions and parameters for some of the waste in which the dose criteria was exceeded, need to be carefully decided if overlap with other waste is considered.
- Dose evaluation was carried out using both, the conservative model and the cautiously realistic conceptual model, considering a number of barrier compositions.
   As a result, it became clear that by taking into consideration the decline in amount of water penetration due to the low permeability layer, diffusion control, and ageing degradation of barrier material, the maximum dose can be reduced. In the evaluation using parameters that are based on the cautiously realistic model that takes diffusion in the waste layer into consideration and the safety cases from UK, a comparatively large decline in dose similar to the decline in leaching rate was seen.
- Since it became clear when the cautiously realistic conceptual model was used that creating models for the waste body layer and considering changes due to ageing can be expected to contribute to dose reduction, developing parameters and incorporating them in the future is presumable.
- In the waste stream that was studied, even though determining the disposal category differed depending on the waste, C-14 was dominant most of the times in the groundwater scenario and I-129 followed.



# Summary

#### **Results so far**

- Parameters concerning dose evaluation were developed based on the outcomes of the studied conducted by IRID until FY2020 (characterization, treatment, etc.) and the latest domestic disposal trends.
- Sensitivity evaluation of the barrier composition and barrier parameters was conducted for the disposal depths of each disposal method, namely, trench disposal, pit disposal, and medium depth disposal, and the parameters that were sensitive were identified for each nuclide.
- The method for deriving the disposal category for each waste based on the dose obtained through sensitivity evaluation was studied. Methods for deriving the barrier composition and required performance of the barrier using the results of sensitivity analysis were proposed, and preliminary proposals of disposal concepts for each waste were consolidated.
- Based on the results of dose evaluation of the preliminary proposals of disposal concepts, it was projected that the disposal concepts determined through sensitivity evaluation would generally meet the dose criteria.

#### Issues, etc.

- Since it became clear when the cautiously realistic conceptual model was used that creating models for the waste body layer and considering changes due to ageing can be expected to contribute to dose reduction, developing parameters and incorporating them in the future is presumable.
- In the waste stream that was studied, even though determining the disposal category differed depending on the waste, C-14 was dominant most of the times in the groundwater scenario and I-129 followed.


(b) Provision of disposal methods and development of safety assessment methods[2] Development of techniques for assessing impact of affecting substances, etc. on disposal

- Achievements until the previous year
- Nuclide migration parameters, barrier material, and affecting substances (6 substances) that have a high priority in impact assessment were selected based on the investigation of affecting substances likely to be contained in waste or the assumed disposal concepts.
- The idea behind the method of evaluating the impact on nuclide sorption parameters depending on the affecting processes of the affecting substances, the level of understanding the processes and the amount of data on the processes, was developed. (Fig. 1)
- Trial runs and derivations were carried out concerning the sorption reduction effect due to direct impact of the 6 substances, and sorption reduction effect including their indirect impact under some of the conditions that have a comparatively large amount of information contributing to the investigation. (Table 1)
- O Issues
- Lack of findings on change in solid phase of borate, ferrocyanide, etc. in particular
- Need for a selective response while evaluating the extent of contribution to exposure dose in the case of the disposal concepts being investigated

#### Goal

- To organize the set of parameters to be applied to dose evaluation considering direct as well as indirect impact and the idea behind them, based on the findings so far and data collected on cases pertaining to which sufficient findings are not available.
- Implementation items
- a. Expansion of findings and data that are insufficient for evaluating direct and indirect impact of nuclide migration, and quantitative evaluation of the impact.
- b. investigation of the impact of the sorption effect of the affecting substances on dose evaluation.
- Indicators for the achievements of goal
- Expansion of data on the combination of impact affecting substances and nuclides, which is required for impact assessment, and presentation of impact assessment techniques
- b. Results of analyzing the impact of various types of parameters on dose evaluation including the impact of decline in the nuclide sorption parameters, and identification of important parameters
- ⇒ b. Will be implemented as part of "[1] Information organization to investigation the disposal concepts and to establish safety assessment methods".



- Fig. 1 Overview of methods for assessing the impacts of affecting substances on nuclide sorption
  - Table 1Organization of sorption reduction factorsfor barrier materials against affecting substances

	Poprocontot	Concentration for the onset of impact (threshold value) and the sorption reduction factors (SRF)									
Element group	ive element	Organic mat	ter (ISA)	Sea-water	Boric a	cid	Ferrocyanide				
		Threshold value	SRF	SRF*3	Threshold value	SRF	Threshold value	SRF			
Alkali metal	Cs	- 1		2	-	1	-	1			
Alkali earth metal	Sr	1×10 ⁻² M ^{*1}	10	8	-	1	-	1			
Divalent transition metal	Ni	-	1	1	—	1	-	1			
Tetravalent transition metal	Sn	1×10 ⁻⁴ M	100	10	—	1	Issues exist	Issues exist			
Pentavalent transition metal	Nb	1×10 ⁻⁴ M	100	10	Issues exist	Issues exist	Issues exist	Issues exist			
Trivalent actinide	Am	1×10 ⁻⁴ M	10	10	-	1	—	1			
Tetravalent actinide	Th	1×10 ⁻⁴ M	100	10	Issues exist	Issues exist	Issues exist	Issues exist			
Pentavalent actinide	Np	1×10 ⁻⁴ M	10	10	-	1	1×10 ⁻³ M	3			
Hexavalent actinide	U	5×10 ⁻⁴ M	10	10	1×10 ⁻² M	500	1×10 ⁻³ M	3			
Halogen	I	- ^{*2}	1	-*2	1×10 ⁻⁴ M	1.3	—	1			
Anionic species	Se	-*2	1	-*2	Issues exist	Issues exist	Issues exist	Issues exist			

(Sorption reduction factor to cement caused by affecting substances (direct impact))

pH Affecting	State I	State II	State Illa	State IIIb
Absent	0.1	0.03	0.1	0.1
ISA - 0.021M	0.01	1.6×10-3	0.01	0.01
Boric acid - 0.4M	0.025	7.5×10-3	0.025	0.025
Boric acid - 0.04M	0.1	0.03	0.1	0.1
Sea-water - 0.6M	0.017	5×10-2	0.017	0.017
Sea-water - 6M	1.7×10-3	5×10-4	1.7×10-3	1.7×10-3

(Sorption distribution factor to cement caused by the affecting substance Sr (including indirect impact))





(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 Flow of investigation (In the case of direct impact assessment) -







Fig. 2 Overview of methods for assessing the impacts of affecting substances on nuclide sorption



(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 Direct impact assessment: Setting of data acquisition targets and conditions for setting sorption reduction factors -

- The relationship between test conditions from last year and the acquired data was analyzed.
- Elements pertaining to which the acquired data was not sufficiently accurate (Table 1 Red frame) and elements pertaining to which some of the data was insufficient (Table 1 Green frame), were identified.
- The consistency, etc. of the factors due to which distribution coefficient that serves as the basic data for calculating the sorption reduction factor that was unable to be acquired, and the assumed geological environmental conditions was verified against the elements that were identified.
- The conditions such as the nuclides used, liquid to solid ratio, etc. were changed, and test condition was set wherein the concentration of affecting substances and the pH conditions were considered to be parameters. (Table 2, Accuracy improvement: Red, Sufficiency of data: Green)

	Boric acid	Ferrocyanide			
Bentonite	Cement	Bentonite	Cement		
NaCl	NaCl	NaCl	NaCl		
7, 9.3	Cement equilibrium w ater (Approx. pH12.5)	4-12	Cement equilibrium w ater (Approx. pH12.5)		
Borio	c acid concentration	Ferrocyanide concentration			
Sn ^{*1} , Se ^{*2}	Sn ^{*1} , Se ^{*2} Sn ^{*1} , Nb ^{*3} , Se ^{*3}		Ni ^{*3} , Sn ^{*1} , Nb ^{*3} , Am(Eu) ^{*4} , Se ^{*3}		

Table 2 List of test conditions for the elements to be retested this year

#### Table 1 Elements to be re-tested this year

		Poprocontot	Concentration for the onset of impact (threshold value) and the sorption reduction factors (SRF)								SRF)
	Element group	ivo element	0	ganic mat	ter (ISA)	Sea-water		Boric a	cid	Ferrocya	anide
σ		ive element	Thresh	old value	SRF	SRF*3	Thr	eshold value	SRF	Threshold value	SRF
<b>U</b>	Alkali metal	Cs		-	1	2		-	1	-	1
ပ္ဆ	Alkali earth metal	Sr	1x1	0 ⁻² M ^{*1}	10	8		-	1	-	1
	Divalent transition metal	Ni		-	1	1		-	1	—	1
	Tetravalent transition metal	Sn	1x1	0 ⁻⁴ M	100	10		-	1	Issues exist	Issues exist
Ē	Pentavalent transition metal	Nb	1x1	0 ⁻⁴ M	100	10	ls	sues exist	Issues exist	Issues exist	Issues exist
Ð	Trivalent actinide	Am	1x1	0 ⁻⁴ M	10	10		-	1	-	1
1	Tetravalent actinide	Th	1x1	0 ⁻⁴ M	100	10	k	sues exist	Issues exist	Issues exist	Issues exist
Ð	Pentavalent actinide	Np	1x1	0 ⁻⁴ M	10	10		-	1	1×10 ⁻³ M	3
$\circ$	Hexavalent actinide	U	5×1	0 ⁻⁴ M	10	10		1×10 ⁻² M	500	1×10 ⁻³ M	3
	Halogen	1	-	*2	1	- ^{*2}		1×10 ⁻⁴ M	1.3	-	1
	Anionic species	Se	-	-*2		-* ²	ls	sues exist	Issues exist	Issues exist	Issues exist
		Depres	ntoti vo	Concent	ration for th	e onset of imp	act (th	nreshold value	e) and the sor	ption reduction fa	ctors (SRF)
$\overline{\mathbf{D}}$	Element group	Represe	ent Thresh		Boric acid				Ferrocyanide		
Ð		CICITI			old value	SRF		Thresh	old value	SRF	:
S.	Alkali metal	Cs	\$		-	1			-	1	
ğ	Alkali earth metal	S	r		-	1			-	1	
	Divalent transition metal	N			-	1		Issue	s exist	Issues e	exist
Ð	Tetravalent transition metal	Sr	۱	1×1	10 ⁻² M	100		1×1	10 ⁻³ M	2.7	
÷	Pentavalent transition meta	I Nt	)	1×1	10 ⁻⁴ M	2.6			-	1	
	Trivalent actinide	An	n	1×1	10 ⁻⁴ M	1.3			-	1	
Ĕ	Tetravalent actinide	ſT	1	lssue	is exist	Issues exi	st	Issue	s exist	Issues e	exist
	Pentavalent actinide	Np	)		-	1		1×1	10 ⁻³ M	4	
ഷ്	Hexavalent actinide	U			-	1		1×1	10 ⁻³ M	2	
	Halogen	1		lssue	s exist	Issues exi	st		-	1	
	Anionic species	Se	e	lssue	s exist	Issues exi	st	1×1	10 ⁻⁴ M	2.0	

The behavior of each target nuclide such as container sorption, sedimentation, etc. was verified and evaluated through blank tests, etc. that do not include a solid phase.

*1: The data collected so far was collected under conditions wherein the initial concentration was high, and the disposal environment was not sufficiently simulated. Hence data will be collected using radionuclides under conditions wherein the initial concentration is low.

*2: Since the difference between the initial concentration and the concentration after sorption equilibrium was small, distribution coefficient was unable to be calculated. Hence data will be acquired once again by changing the liquid to solid ratio.

*3: Since there was large amount of sorption and the concentration after sorption equilibrium was lower than the lower limit of quantification of the equipment, the distribution coefficient was unable to be calculated. Hence, data will be acquired once again by changing the liquid to solid ratio.

*4: Since data under conditions wherein ferrocyanide that is to be compared did not coexist, was not available, the dependency of distribution coefficient on the ferrocyanide concentration was unable to be sufficiently evaluated. Hence data will be acquired once again.

*5: Since data on the circum-neutral range, which is required for ascertaining the disposal environment is not available, it will be acquired once again.



(b) Provision of disposal methods and development of safety assessment methods

- [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
  - Direct impact assessment: Assessment and investigation of boric acid impact Evaluating the Sorption
- Sn The distribution coefficient to cement (OPC) declined under conditions wherein the Na concentration was 0.05 mol/l and the boric acid concentration was high (0.1 mol/l).
  - The distribution coefficient to bentonite declined under conditions wherein the boric acid concentration was high and the pH was either 7 or 9.3 (Fig. 1).
- Nb As there was large amount of sorption to cement (OPC), the concentration after the test was below the lower limit of quantification. Hence the distribution coefficient was unable to be evaluated.
- Se The distribution coefficient to cement (OPC) declined under conditions wherein the boric acid concentration was high (Fig. 2).
  - The distribution coefficient to bentonite declined under conditions wherein boric acid coexisted and the pH was either 7 or 9.3 When the pH was 9.3, if boric acid coexisted, the volume of sorption reduced. Hence the distribution coefficient was unable to be calculated.
- The sorption reduction factors (SRFs) were derived for the combination of barrier material and nuclides pertaining to which the dependency of the distribution coefficient on boric acid concentration was verified, SRFs were newly evaluated or were updated to highly reliable values.

**Newly evaluating the SRFs** (mentioned hereinafter in the format of "Element - target solid phase: SRF value")

Se - OPC: 32, Se - Bentonite: 1.5

Updating the SRFs with highly reliable values

Sn - OPC: 1  $\rightarrow$  2, Sn - Bentonite: 100  $\rightarrow$  2.0



Fig. 2 Results of the sorption test of Se to cement (OPC) in the presence of boric acid



(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal

- Direct impact assessment: Assessment and investigation of ferrocyanide impacts -





(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 Direct impact assessment: Summary of the results of the trials on the threshold value for onset of impact and the sorption reduction factors -

		Representa	Concentration for the onset of impact (threshold value) and the sorption reduction factors (SRF)									
	Element group	tive	Organic matte	er (ISA)	Sea-water	Boric aci	d	Ferrocyanide				
		element	Threshold value	SRF	SRF*3	Threshold value	SRF	Threshold value	SRF			
D D	Alkali metal	Cs	-	1	2	—	1	—	1			
S	Alkali earth metal	Sr	1×10-2 M*1	10	8	—	1	—	1			
a a	Divalent transition metal	Ni		1	1	—	1	—	1			
	Tetravalent transition metal	Sn	1 × 10 ⁻⁴ M	100	10	1 × 10 ⁻² M	2	1 × 10 ⁻² M	1.8			
С О	Pentavalent transition metal	Nb	1 × 10 ⁻⁴ M	100	10	Issues exist	Issues exist	(During analysis)	(During analysis)			
Ē	Trivalent actinides	Am/Eu	1 × 10 ⁻⁴ M	10	10	—	1	-	1			
Ð	Tetravalent actinides	Th	1 × 10 ⁻⁴ M	100	10	Issues exist	Issues exist	Issues exist	Issues exist			
$\bigcirc$	Pentavalent actinides	Np	1 × 10 ⁻⁴ M	10	10	—	1	1 × 10 ⁻³ M	3			
	Hexavalent actinides	U	5×10⁻⁴ M	10	10	1 × 10 ⁻² M	500	1 × 10 ⁻³ M	3			
	Halogen	I	-*2	1	-*2	1 × 10 ⁻⁴ M	1.3	—	1			
	Anionic species	Se	-*2	1	-*2	1 × 10 ⁻² M	32	—	1			

		Representa	Concentration for the onset of impact (threshold value) and the sorption reduction factors (SRF)								
	Element group	tive	Boric aci	d	Ferrocyanide						
		element	Threshold value	SRF	Threshold value	SRF					
	Alkali metal	Cs	—	1	—	1					
	Alkali earth metal	Sr	—	1	1 × 10 ⁻² M	2.9					
	Divalent transition metal	Ni	—	1	(During analysis)	(During analysis)					
	Tetravalent transition metal	Sn	1 × 10 ⁻² M	2.2	1 × 10 ⁻² M	100					
	Pentavalent transition metal	Nb	1 × 10 ⁻⁴ M	2.6	-	1					
	Trivalent actinides	Am/Eu	1 × 10 ⁻⁴ M	1.3	—	1					
	Tetravalent actinides	Th	Issues exist	Issues exist	Issues exist	Issues exist					
	Pentavalent actinides	Np	—	1	1 × 10 ⁻³ M	4					
	Hexavalent actinides	U	_	1	1 × 10 ⁻⁴ M	2.1					
	Halogen	I	Issues exist	Issues exist	_	1					
_	Anionic species	Se	1 × 10 ⁻³ M	1.5	1 × 10 ⁻⁴ M	2.0					

- *1 M:mol/L
- *2  $K_d = 0 (m^3 kg^{-1})$
- *3 SRF between precipitation conditions and seawater conditions
- Set based on past information
- Set based on data obtained through sorption tests
- Red: Data that was updated based on results of tests conducted this year
- * Findings from complex formation in the past suggested that neither sulfate nor carbonate had a significant impact and hence these have not been mentioned.

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Bentonite based

(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 Indirect impact assessment: Methods of assessing indirect impact based on past disposal research

- If there is a change in the pore water pH or solid phase properties of cement or bentonite due to coexisting substances, their indirect impact needs to be considered.
- For assessment of indirect impact, the methods for setting sorption distribution coefficients can be utilized which were established through the past investigations of geological disposal and are adaptable to various environmental conditions and their changes.
- The setting methods include the following: a thermodynamic sorption model that predicts changes in conditions based on a theoretical model; a condition conversion method that provides corrections based on the relationships between changes in environmental conditions and sorption distribution coefficients; and experts' judgments based on limited actual measurement data or knowledge of sorption mechanisms. The applicability of these methods depends on the processes in which affecting substances exert an impact on the properties of barrier materials or depends on the level of understanding of the actual measurement data or the mechanisms.

Concept of evaluation methods for sorption distribution coefficients under various environmental conditions for performance evaluation of geological disposal (NEA, 2005; Tachi et al., 2017)



 Indirect impact assessment for bentonite: The condition conversion method that has been investigated during the geological disposal evaluations conducted so far, for setting the sorption parameters considering environmental transition impact on bentonite, is applicable.

Indirect impact assessment for cement: It is difficult to assess indirect impact of cement based materials using the method used for bentonite, because of the properties of its solid phase, the transition process, and the complexity of their relationship with the radionuclide sorption and retention mechanisms. Hence an estimation and assessment method based on the relationship between the solid phase transition process and its impact on nuclide sorption is required.



# (b) Provision of disposal methods and development of safety assessment methods [2] Development of techniques for assessing impact of affecting substances, etc. on disposal Indirect impact assessment: Approach towards concretization of the assessment of indirect impact on cement -

### Approach towards investigating the assessment of indirect impact on cement

- In the past, the method that considers the gradual transition process of the pore water and mineral facies of cement, and the associated changes in the properties of nuclide sorption has been considered for establishing the parameters for nuclide sorption to cement. While following the methods for setting sorption that were used so far, a method of assessing the indirect impact was presented wherein the changes in mineral facies of cement caused by affecting substances in the accident wastes, and their impact on nuclide sorption, were additionally considered.
- In that assessment method (Fig. 1), the transitions in constituent minerals of cement are consolidated (Step ①) based on the past evaluations of sorption in cement during radioactive waste disposal, and thereafter the constituent minerals of cement that dominantly contribute to the sorption of nuclides being evaluated are specified (②). Then, the changes taking place in the mineral facies of cement caused by the affecting substances are identified (③), and their impact on nuclide sorption is qualitatively estimated based on the relationship between the properties of nuclide sorption and the cement minerals specified in steps ① and ②. Further, based on these estimation results, parameters can be conservatively set considering the indirect impacts, depending on the type and contained amount of the affecting substances.

Past evaluations of sorption of nuclides to cement during radioactive waste disposal	① Specification of the changes in the composition of minerals during the cement transition process Consolidation of the cement mineral composition considering cement hydration, the pore water chemistry, and their transition process	<b>→</b>	<ul> <li>2 Specification of the mineral facies that dominate sorption of radionuclides</li> <li>Consolidation of mineral facies that contribute to or dominate sorption to cement of radionuclides being evaluated</li> </ul>
Assessment of	<ul> <li>3 Analysis of impact of affecting</li></ul>		(4) Assessment of impact on nuclide
indirect impact of	substances on the mineral composition of		sorption associated with mineral changes
affecting	cement <li>Analysis and organization of the impact of</li>		caused by affecting substances
substances in the	affecting substances in the Fukushima		Assessment of impact of changes in
Fukushima	accident wastes on the constituent		cement mineral facies caused by affecting
accident waste on	minerals of cement and their transition		substances, on nuclide sorption, based on
nuclide sorption	states		information from (1 - (3).

Fig. 1 Approach towards assessment of indirect impact of affecting substances on nuclide sorption in cement (Steps ① - ④)



(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 - Indirect impact assessment: Assessment of indirect impact on cement (1) -

Specification of the changes in the composition of constituent minerals during the cement transition process (Step ①)

- Cement, which is represented by OPC, passes through several transition states due to interactions such as long-term hydration reaction with groundwater components, etc. (Fig. 1)
- The transition of the mineral facies of cement has been understood through experiments and models. The relative importance of the constituent mineral facies differs depending on the transition states respectively. The changes in the main mineral compositions can be organized depending on the category of the transition state of cement as shown in Table 1 (Berner, 1990; Taylor, 1997; Ochs et al., 2016).



porewater exchange cyles (log scale) ->

Fig. 1 Transition process (State I to IV) of the pore water of OPC (Ochs et al., 2016)

the composition of the	Transition state (State)	State I	State II	State IIIa	State IIIb	State IV
main constituent	Pore water pH	>13.1	12.5	11	11	<9
cement (OPC) transition process	Portlandite	Roughly 20 wt%				0.00
	CSH1.7	Roughly 45 wt%				
	CSH1.5-1.7	0 ,				
	CSH0.83					
	AFt phase / Ettringite	Roughly 10 wt%				
	AFm phase	Roughly 10 wt%				
	Hydrotalcite	Roughly 2 wt%				
	Calcite	Roughly 1 wt%				
	Non-hydrated clinker minerals	Roughly 10 wt%	Can exist in a	ll states		

(Gray: Main mineral composition during the transition process)



(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 - Indirect impact assessment: Assessment of indirect impact on cement (2) -

### Specification of the mineral facies that dominates sorption of radionuclides (Step 2)

- During the past safety evaluations of waste disposal, nuclide sorption evaluations that considered the transition in state of mineral facies and pore water of cement, were taken into consideration (Wieland, 2014; Ochs et al., 2016). Specifically, the sorption distribution coefficients were set on the basis of data obtained by actual measurement during the respective transitions, and by taking the relation between the cement mineral facies and nuclide sorption properties into consideration.
- In order to investigate the impact of affecting substances in the accident wastes on cement mineral facies and nuclide sorption, the relationship between the cement mineral facies and the sorption properties of a variety of nuclides were investigated based on cases that examined setting of sorption distribution coefficient to cement so far, and in addition, the relationships were consolidated as shown in Table 1.

Table 1 Relationship between the constituent minerals of cement and the sorption properties of radionuclides (extent of sorption: Sorption present +++ > ++, sorption absent -, unclear*)

Cement constituent minerals	Alkal alkali	Alkali and alkali earth		Transition metal			Actinides and lanthanides				Anion		
minerals	Cs(I)	Sr(II)	Ni(II)	Sn(IV)	Nb(V)	Am/Eu(III)	Th(IV)	Np(V)	U(VI)	Ι	Cl	Se(IV)	
Portlandite	_	_	+	—	_	—	—	—	—	_	_	_	
CSH1.7	+	+	+	_	+	+++	+~++	+~++	+	+++	++	++	
CSH1.5-1.7	++	++	+	—	+	+++	+~++	+~++	+~++	++	+	++	
CSH0.83	+++	+++	+	_	+	+++	+++	+++	+	+	#	++	
AFt phase / Ettringite	#	+	++	_	++	+	_	—	_	+++	+	++	
AFm phase	—	_	_	—	_	+	_	_	—	+++	++	+	
Hydrotalcite	_	_	+	+	—	—	—	—	—	++	#	+	
Calcite	_	+	+	+	#	+	+	+	+	_	_	+	



(b) Provision of disposal methods and development of safety assessment methods [2] Development of techniques for assessing impact of affecting substances, etc. on disposal

- Indirect impact assessment: Assessment of indirect impacts on cement (3) -

Assessment of impact of affecting substances on the mineral composition of cement (Step (3))

- By investigating and analyzing the past findings related to each of the 6 affecting substances, the impacts of seawater component were organized in the form of an impact flow as shown in the example (Fig. 1), and from among the indirect impact processes to be considered, the possibility of transition of mineral facies in particular was specified.
- For example, as an indirect impact of seawater components on cement, in addition to the changes in the main constituent minerals such as CHS, AFt phase (Ettringite), AFm phase, and hydrotalcite as shown in Fig. 1, there may be possibilities of solid phase transition such as formation of brucite due to Mg components, formation of ettringite and gypsum due to sulfuric acid components, formation of Friedel's salt due to CI, etc.





(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 - Indirect impact assessment: Assessment of indirect impacts on cement (4) -

Assessment of impact on nuclide sorption associated with mineral changes caused by affecting substances (Step (4))

- The impact of respective affecting substances on nuclide sorption through the intermediary of transition of the cement mineral facies was qualitatively evaluated based on the impact of affecting substances investigated in Step ③ above, on cement mineral facies (dissolution / sedimentation), and the relationship between cement mineral facies and properties of nuclide sorption investigated in Steps ① and ②.
- For example, as an impact of seawater component, indirect impact evaluation is possible as shown in Table 1. Hence the possibility of sorption reduction of a variety of nuclides due to CSH dissolution, increase in sorption of anions due to Aft phase (Ettringite) sedimentation, etc. needs to be considered.

 Table 1 Changes in cement constituent minerals caused by the impact of seawater component, and results of assessment of the impact on nuclide sorption associated with it

	Changes	s in cem	ent transi	tion state (	State)	Impact on nucli		
Cement constituent minerals	State I	Π	IIIa	IIIb	Alkali and alkali earth	Transition metal	Actinides and lanthanides	Anion
Portlandite	-	-						
CSH1.7	-				-	—	-	
CSH1.5-1.7		-			-	-	—	
CSH0.83			_	_	_	_		
AFt phase / Ettringite	+	+	+		±	+	+	+
AFm phase								
Hydrotalcite	+	+	+	+				Ŧ
Calcite	+	+	+		+	+	-	Ŧ

(Legend) Gray: Main mineral composition during the transition process, Red/-: Dissolution of minerals or decline in nuclide sorption, Blue/+: Sedimentation of minerals, increase in nuclide sorption, Green/±: Unclear impacts



(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 Current status of the sorption impact assessment (direct and indirect impact) considering the affecting substances and the issues thereof -

The current status of assessment of sorption impact of the 6 affecting substances namely, organic matter, seawater components, boric acid, ferrocyanide, sulfates and carbonates, including both direct and indirect impact, and the future issues have been summarized below based on the results of investigations and studies conducted so far.

Affecting substance	Current assessment of sorption impacts during disposal	Future issues
Organic matter	<ul> <li>Past findings on the impact of organic matter are comparatively substantial. It is realistic to use the SRF set values based on actual measured data on the sorption impact of isosaccharinic acid (ISA) which is believed to have the largest impact in particular.</li> <li>The method for estimating the SRFs for ISA and EDTA based on thermodynamic data was investigated. It was believed that it would be possible to evaluate the estimation of SRF if highly reliable thermodyamic data can be used. However, thermodynamic data on organic matter cannot be considered sufficient.</li> </ul>	<ul> <li>The direct impact of organic matter due to complex formation is highly likely to become evident compared to that of other affecting substances. Therefore it is necessary to investigate more realistic impact assessment by assuming the organic matter likely to be actually contained in accident wastes and its concentration.</li> <li>In particular, the investigation of applicability of impact assessment methods to a variety of organic matters, and the investigation of the systematicity evaluation methods, etc. between organic matters is important.</li> </ul>
Seawater component	<ul> <li>Regarding the impacts of seawater components on nuclide sorption, there is a relatively large amount of knowledge, so it is possible to some extent to quantitatively evaluate sorption reduction (direct impact) based on the understanding of sorption mechanisms such as sorption competition with seawater components.</li> <li>Regarding the assessment of indirect impact of high concentration seawater components, the sorption impact based on the transition of cement solid phase can be qualitatively assessed.</li> </ul>	<ul> <li>The impact of seawater components is highly likely to become evident. Hence the settings need to be revised further based on the latest findings on seawater component impact.</li> <li>Also, a more realistic indirect impact assessment based on the more realistic analysis and evaluations of long term transition of barrier material such as cement, etc. as a result of seawater components is required.</li> </ul>
Sulfates	<ul> <li>It was presumed based on the actual measured data obtained from this project and from the assessment based on thermodynamic data that the direct impact of the formation of sulfate complex on sorption is not conspicuous.</li> <li>Meanwhile, the indirect impact of transitioning of the cement mineral facies assumed to have occurred due to the impact caused by the presence of sulfates, can be assessed based on the qualitative and conservative methods proposed in this project.</li> </ul>	• Regarding the solid phase transition in the presence of sulfates, in addition to understanding that transition process, it is important to understand the impact on barrier performance other than nuclide migration. Hence further investigations including analytical assessment are required.
Carbonates	<ul> <li>The impacts of carbonates can be assessed using methods that have been investigated in the past, because carbonates are components originally contained in natural rock or groundwater environments.</li> <li>The indirect impact of transitioning of the cement mineral facies assumed to have occurred due to the impact caused by the presence of carbonates, can be assessed based on the qualitative and conservative methods proposed in this project.</li> </ul>	<ul> <li>As mentioned in the column on the left, although carbonates have been included in the disposal evaluation conducted so far as well, including their direct and indirect impacts, investigations based on the status of carbonates contained in accident wastes are necessary.</li> </ul>
Boric acid	<ul> <li>Regarding the impact of boric acid, data on sorption impact was obtained and based on those results, it can be assessed that boric acid has little impact on alkali metals and alkali earth metals, whereas in the case of actinides, if the presence of carbonic acid is considered where boric acid has an impact to some extent, that impact is not prominent.</li> <li>When the waste has high concentration of boric acid, the impact of solid phase transition needs to be considered as well, but there is lack of information on this point.</li> </ul>	<ul> <li>It is believed that the presence of high concentration boric acid will be concretized while investigating waste conditioning considering the waste conditioning process and the concentration limit as boron is a substance subject to environmental standards.</li> </ul>
Ferrocyanide	<ul> <li>Regarding ferrocyanide, data on sorption in the presence of ferrocyanide was obtained which suggested that even though it has a slight impact on some nuclides, that impact is not significant.</li> <li>When there is high concentration of ferrocyanide, the impact of solid phase transition is likely to occur, but there is lack of information on this point.</li> </ul>	<ul> <li>Along with the fact that total cyanide is a substance subject to environmental standards, the behavior of ferrocyanide inside the waste body or in the disposal environment needs to be evaluated, and investigations need to be conducted including measures to be taken at the time of the pretreatment</li> </ul>



(b) Provision of disposal methods and development of safety assessment methods
 [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
 Summary -

#### **Current results**

✓ The impact processes of the impacts of affecting substances (6 main substances) that are likely to be present in accident wastes, on nuclide sorption to the engineered barriers (cement, bentonite) which are used in common in a variety of disposal concepts were identified.

✓ The affecting substances were categorized into those that have a "direct impact" due to formation of complexes with the nuclides, and those that have an "indirect impact" by giving rise to changes in the solid phase or pore water of engineered barriers, and the methods for evaluating the respective impacts were proposed.

✓ For direct impact, a method of deriving sorption reduction factors (SRF) depending on the status of data was presented, and in addition, by investigating existing information and expanding data that was insufficient, the SRFs and concentration at which the impact appears (threshold) were organized respectively for the 6 main affecting substances - the engineered barriers (cement, bentonite) - the main elements.

✓ With regards to indirect impacts, the method of qualitatively assessing the impact of affecting substances on nuclide sorption as a result of changes in the cement mineral facies, was presented, whereas for bentonite, using the conventional method was proposed.

✓ The possibility that the impact of organic matter and seawater components would be relatively important in the case of direct impacts, whereas the impact of seawater components and sulfates would be relatively important in the case of indirect impacts, was presumed.

### Future issues

✓ The method for evaluating direct impact of important organic matters and other affecting substances included in accident wastes considering their diversity and concentration needs to be concretized.

✓ Assessment of direct and indirect impacts considering the concentration of affecting substances in actual disposal systems, their spread, etc. is required.

✓ It is important to investigate impact assessment and identification of issues in conjunction with the analysis of nuclide migration and the analysis of uncertainties.

✓ Investigations need to be carried out including processing measures considering perspectives such as impact at the time of waste conditioning, substances subject to environmental standards, etc.



## 2. Project details (No. 3)

## c. Characterization of waste

(a) Efficient characterization

- [1] Investigation on analytical inventory estimation methods
- [2] Proposal for mid-and-long-term analysis plan
- (b) Development of sampling technology



## c. Characterization of waste Overview of analysis and R&D with data from analysis





## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis

- Achievements up to FY2020
- Collection, management and analysis of analysis samples required for characterization of solid waste continued to be carried out.
- Rubbles, secondary waste generated from contaminated water treatment, contaminated water, plants, and soil were analyzed by assigning the task of analysis to 5 facilities (JAEA NSRI / NCL / Oarai, NDC and NFD) located in Ibaraki prefecture.
- The obtained analyzed data is being used for decommissioning (Fig. 1) and in addition, it has been included in the database so that it can be used by concerned parties (Fig.2).
- Goal
- To accumulate analyzed data on solid waste so as to contribute to the development of analytical inventory estimation methods.
- Details of implementation
- Obtaining the cooperation of TEPCO, collecting analysis samples and storing them inside the power station with the idea of continuing to analyze the solid waste in the mid-and-long-term.
- Transporting the analysis samples to analytical facilities such as JAEA, etc. located in Ibaraki prefecture, and conducting analysis
- Recording the obtained analyzed data into the database.
   "Fukushima Daiichi Radwaste Analytical Data Library (FRAnDLi)", and making it available for general use (Fig. 2) Also, investigating databases for managing analysis samples.
- Indicators for the achievements of goal
- O Continued disclosure of analytical data.





Fig. 2 Analysis database that continues to be used https://frandli-db.jaea.go.jp/FRAnDLi/

Fig. 1 Results of analyzing the water from Unit 2 Torus room (Referenced from TEPCO) *

## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis plan)

This year, the plan for conducting analysis focusing on rubble and secondary waste generated from contaminated water treatment is being drafted. (Table 1) In order to reduce the uncertainties in estimating inventories, contamination mechanism and waste that contributes to accumulating statistical data will be selected and  $\circ$ analyzed.

#### Table 1 Plan for this year's analysis and status of implementation

			Sample		Status	Status		
Classification		Focus points for inventory estimation	samples	Collection	Transport	Analysis method	Analysis	Analyzed by
Dismantling	Units 1, 2 exhaust stack core	Location (Off-gas system)	3	Completed	This year First time		Completed	NDC
Waste	Smear solution in Unit 1 SGTS piping	Same as above	1	Completed	Completed		Completed	NFD
Rubble	Unit 1 Reactor Building, 1-3 Radwaste Building	Location (inside the building), Material (rubbles)	8	Completed	This year First time		Completed	NDC
	Unit 1 Reactor Building stagnant water (containing slud	Location (Basement floor)	To be determined	Pending	Planned for FY2022			JAEA
	Unit 3 Reactor Building accumulated water (Unitaining Suage)	Same as above	2	Completed	Received on August 19		Completed	JAEA
	Water from contaminated water storage tank (containing sludge)	Location (residual water in the tank) Contamination mechanism	1	Completed	Received on August 19		Completed	JAEA
	Unit 3 Turbine Building sludge	Same as above	1	Completed	Completed		Completed	NFD
Secondary waste generated from contaminated water treatment	Iron co-precipitation slurry	Ort Actual waste data	4	Completed	Completed		Completed	JAEA
	Carbonate precipitation slurry	Same as above	6	Completed	Completed	Verified	Completed	JAEA
	Carbonate precipitation slurry	Same as above	4	Completed	This year First time		Completed	NFD
	Concentrated liquid waste slurry	Same as above	2	Completed	This year First time		Completed	NFD
	Cerium oxide This rep	Ort Same as above	5	Completed	Completed	Needs to be investigated	Analysis in process	JAEA
	Chelating resin 2	Same as above	1	Completed	Completed	Investigated	Completed	JAEA
	Chelating resin 1	Same as above	1	Completed	Completed	Investigated	Completed	JAEA
	Silver zeolite	Same as above	2	Completed	Completed	Needs to be investigated	Analysis in process	JAEA
	Titanium oxide This rep	Ort Same as above	2	Completed	Completed	Needs to be investigated	Completed	JAEA
	Ferrocyanide	Same as above	1	Completed	Completed	Investigated	Completed	JAEA
	Titanate	Same as above	2	Completed	Completed	Investigated	Analysis in process	JAEA
	Titanate 2	Same as above	3	Completed	This year Second time	Needs to be investigated		JAEA
	Silver zeolite	Same as above	1	Completed	This year Second time	Needs to be investigated		JAEA
	Cerium oxide	Same as above	2	Completed	This year Second time	Needs to be investigated		JAEA
	Activated carbon	Same as above	To be determined	Completed	To be determined			JAEA
	Sub-drain adsorbent	Same as above	4	Completed	This year First time		Completed	NDC
	Activated carbon sandba	Same as above	1	Completed	Completed		Completed	JAEA

# (a) Efficient characterization ① Investigation on analytical inventory estimation methods

### i. Implementation of analysis (Analysis plan)

- Technology is being developed for collecting adsorbent from the used cesium adsorption vessel ((b) Development of sampling technology).
- The dose rate of the adsorbent is extremely high and is expected to reach the Sv order every hour with about 10 cm³ of adsorbent. Hence, considering safety, specific procedures right up to analysis need to be investigated.
- Impacts to be considered at each stage of storage and transportation were identified and technical constraints were investigated (Table 1) Further, the handling method was investigated considering 2 facilities of the JAEA as examples of analysis facilities.
- Since the operation of chemical separation is carried out in the glove box or the hood where contamination is controlled, radioactive Cs needs to be removed or fractionated in advance in the cell. Investigations continue to be carried out.

Process	Mass*(g)	Dimensions (cm)	Radioactivity (Bq)	Dose rate (mSv/h)
Collection	<ul> <li>Measurement work</li> <li>Transportation of heavy weight objects</li> </ul>	<ul> <li>Dimensions of sampling head</li> </ul>		<ul> <li>Exposure during work such as sampling, etc.</li> </ul>
Storage	<ul> <li>Transportation of heavy weight objects</li> </ul>	<ul> <li>Dimensions of the canister</li> <li>Storage location</li> </ul>		<ul> <li>Conformance with the control value</li> <li>Exposure during handling work</li> </ul>
Transport	<ul> <li>Specifications of transport cask</li> <li>Transportation of heavy weight objects</li> </ul>	<ul> <li>Type of transport cask</li> </ul>	<ul> <li>A₂ value</li> <li>Formation of hydrogen</li> </ul>	<ul> <li>Conformance with the regulatory value</li> <li>Exposure during work such as measurement, packing, etc.</li> </ul>
Analysis	<ul> <li>Transportation of heavy weight objects</li> </ul>	Dimensions of the service entrance	<ul> <li>Quantity permitted</li> <li>Limit of fixed quantity</li> </ul>	<ul> <li>Conformance with the control value</li> <li>Exposure during work</li> </ul>

### Table 1 Impact of the properties of the sample on the handling method



Fig. 1 Flow leading to the analysis of the cesium adsorbent (being investigated)



## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Sampling)

- The analysis samples of solid waste were obtained in coordination with the construction work and the operation of the processing facility. A plan for collecting around 75 rubble samples from Units 1 to 4 and waste storage locations was created while referring to plans such as those concerning construction work, etc. 90 samples were collected from buildings in Units 1, 2 and 3, Unit 3 southern yard, common pool, temporary storage area C for rubble, and flange tank dismantling facility. (Table 1)
- Also, the process water was collected from the contaminated water treatment facility. A total of 18 samples were collected from between the adsorption vessels in Sarry II over the course of 3 sampling rounds (August 27, 2021, November 20, 2021 and February 22, 2022) with 6 samples collected during each round by means of the sampling line for operation management.
- O Sampling of the ALPS adsorbent had been planned. However, it was unable to be pursued as the equipment were in operation.

Table 1 Breakdown of the number of samples of rubble, etc. collected

	Sampling location	S	Numb	per of samples coll	lected
Unit, etc.	Building	Floor	Rubble and other debris	Smear	Total
Unit 1	R/B	First floor	-	3	3
		Third floor	2	1	3
	T/B	First floor	1	1	2
	Rw/B	First floor	1	1	2
Unit 2	R/B	First floor	-	2	2
	T/B	Second floor	-	2	2
	T/B	First floor	3	4	7
		Second floor	3	3	6
	Rw/B	Basement Level 1	1	1	2
Unit 3	R/B	First floor	5	11	16
		Second floor	-	7	7
Unit 3 southern ya	ard (Fig. 1)		6	-	6
Common pool			8	-	8
Temporary storag	e area C for rubble	(Fig. 2)	20	-	20
Unit 2 R/B First floor Second floor T/B First floor Second floor Second floor Second floor Rw/B Basement Level 1 Unit 3 R/B First floor Second floor Unit 3 southern yard (Fig. 1) Common pool Temporary storage area C for rubble (Fig. 2) Flange tank dismantling facility			4	-	4



Fig. 1 Unit 3 Southern yard (Numbers indicate the dose rate (mSv/h), Red dot indicates the sampling location)



Fig. 2 Crushed rubble at temporary storage area C

### RID

# (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Sampling)

- Incinerated ash generated during the operation of the miscellaneous solid waste incineration facility is filled in drums and stored in the temporary storage facility.
- O There are 2 incineration systems (System A and B), and both systems are identical.
- A total of 25 samples were collected form the incinerated ash generated by both the systems (Table 1). The samples were collected after the ash was filled in the drums before closing the lid. (Fig. 1)
- The dose rate of the samples showed a tendency to have a correlation with the surface dose rate of the drum. It does not form a straight line perhaps due to the background (Fig. 2).

Table 1 Breakdown of collection of incinerated ash samples

	System	Number of samples collected			System A
	А	13			System B
	В	12		0.01	
			entry of the second sec	0.001	
the second s	and the second second		- All	0.01	0.1

Sampling location (Temporary storage chamber) The incinerated ash drum and the collected samples

Fig. 1 Collection of incinerated ash samples

Fig. 2 Dose rate of the collected incinerated ash samples (mSv/h)



Drum

(a) Efficient characterization ① Investigation on analytical inventory estimation methods
 i. Implementation of analysis (Storage management and transportation of samples)

- The collected and obtained samples are stored in the solid waste storage facility (Fig. 1) and continue to be managed as before.
- The analysis samples were transported to the analysis facilities, namely JAEA (Nuclear Fuel Cycle Engineering Laboratories), NDC, and NFD.
- Samples were transported in 2 batches with the first batch sent on September 16. The second batch was sent on February 17. (Table 1)
- And, the Unit 3 torus room stagnant water reached the Nuclear Fuel Cycle Engineering Laboratories (NCL) in August.





Classification	Sample	Number of samples	Batch	Destination
emolition waste	Units 1, 2 exhaust stack core	3	First	NDC
Rubble	Unit 1 Reactor Building, Unit 2 Turbine Building, Units 1 - 3 Radwaste building	11	First	NDC
	Carbonate precipitation slurry	4	First	NFD
	Concentrated liquid waste slurry	2	First	NFD
econdary waste generated from	Sub-drain adsorbent	4	First	NDC
vater treatment	Titanate 2 (FST)	3	Second	JAEA, NCL
	Silver zeolite	1	Second	JAEA, NCL
	Cerium oxide	2	Second	JAEA, NCL

#### Table 1 Transported analyzed samples



## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis results Rubbles)

- With regards to the properties of the rubble and demolition waste, the contamination of the exhaust stacks in Units 1 and 2 was analyzed.
- Approximately 60 m of the top portion of the exhaust stacks in Units 1 & 2 were dismantled. Core samples from the exhaust stack were obtained, and the inner surface was analyzed (Fig. 1).
- ⁹⁰Sr, ¹²⁵Sb, ^{134, 137}Cs, ²³⁸U were detected. Actinide nuclides other than ³H, ¹⁴C, ⁶⁰Co, ⁶³Ni, ⁷⁹Se, ⁹⁹Tc, ¹⁰⁶Ru, ¹²⁹I, ^{152, 154}Eu and ²³⁸U were not detected (Fig. 2).
- When the transport ratio (Cs-137 as reference) of the detected nuclides was calculated along with the analytical value of the smear solution obtained by wiping off the inside of the Standby Gas Treatment System (SGTS) pipes in Units 1 and 2 (analyzed this year), and the contaminated water^{*2} from the exhaust stack drain sump pit in Units 1 and 2, regardless of the sampling location, the values for ⁹⁰Sr and ¹²⁵Sb were the same.
- O Meanwhile, the results from the inner surface of the exhaust stack and the inside of the SGTS pipes differed for actinide nuclides.



*1 TEPCO HD, "Fukushima Daiichi Nuclear Power Station Units 1 and 2 exhaust stack dismantling work progress status" February 27, 2020. *2 A. Shimada et al., Sci Rep 12, 2086 (2022). https://doi.org/10.1038/s41598-022-05924-2



## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis results Stagnant water sludge)

- The solid components (sludge) in the stagnant water in the building basement is important for ascertaining the status of contamination of demolition waste, and hence data on the sludge is being accumulated. The tank water collected while dismantling the tank in which stagnant water from the Unit 3 torus room and the main steam isolation valve (MSIV) room as well as the treated water were stored, was analyzed.
- All the water samples contained dark brown deposits (Fig. 1). When the samples were filtered sequentially using filters with hole size 10, 1, 0.1, and 0.02 µm, almost all the sludge was collected using the filter with hole size 10 µm in the case of all the samples (Fig. 2).
- 99.6% or more of the Pu, Am and Cm nuclides were detected from the residue of the filter with hole size 10 µm. Fe was the main element that the residue (sludge) of the filter with hole size 10 µm consisted in the case of all the samples.*
- When the nuclide composition of Pu, Am, and Cm was compared with that of fuel, it was similar in the case of stagnant water, and in the case of tank water the proportion of Cm was small (Fig. 3)
  - * ICP-AES analysis of the solution obtained by dissolving the residue in nitric acid, etc. was conducted. It was found that 100% weight proportion was the most common with respect to the detected elements.



Unit 3 Torus room water Unit 3 MSIV room water E-D1 tank water

Fig. 1 Appearance of stagnant water samples

Filter hole size	10 µm	1 µm	0.1 µm	0.02 µm
Mass (g)	0.036	ND	ND	ND
ZAppearance of the filtration residue				

Fig. 2 Filtration results of Unit 3 Torus room water

Fig. 3 Analysis results of Pu, Am and Cm from the stagnant water sludge (relative proportion)





## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis results Stagnant water)

- Ascertaining the concentration of radionuclides in stagnant water and treated water is helpful in estimating the status of contamination of structural material of buildings and secondary waste generated from contaminated water, and in investigating the treatment and disposal methods. Hence ⁷⁹Se was analyzed.
- If the usual method using hydrochloric acid is used in equipment made of stainless steel, the equipment gets corroded. Hence the method of conducting the separation operation in a nitric acid solution system was developed and applied (Fig. 1).
- ⁷⁹Se was not detected in any of the 17 samples of stagnant water and treated water that were submitted for analysis so far (Table 1). Further, the collection rate of Se carriers was good at 74 96%.



 Table 1
 Results of analyzing ⁷⁹Se present in stagnant water and treated water

ample number	Sampling date	Sampling locations	Concentration (Bq/cm ³ )
-2RB5-1	August 7, 2013	Unit 2 PCV	< 5 × 10- 2
-3RB5-1	October 22, 2015	Unit 3 PCV (near the water surface)	< 5 × 10- 2
-TS-02	February 13, 2020	Unit 2 Torus room (filtrate of 0.22µm filter)	< 5 × 10 ⁻²
-1RB-1	December 8, 2016	Unit 1 R/B basement (collected from the water sampling port)	< 5 × 10- 2
-1TB5-6	September 30, 2015	Unit 1 T/B (top of OP1900)	< 4 × 10- 2
-2TB7-1	September 25, 2015	Unit 2 T/B basement	< 4 × 10- 2
-3TB7-1	October 15, 2015	Unit 3 T/B basement	< 5 × 10 ⁻²
-RW2-2	March 11, 2014	Centralized RW basement (KURION inlet)	< 4 × 10 ⁻²
-RW4-2	March 9, 2015	Centralized RW basement	< 5 × 10- 2
-HTI2-2	February 11, 2014	HTI/B basement stagnant water (SARRY inlet)	< 5 × 10 ⁻²
-KU3-1	July 13, 2011	Cesium absorption apparatus outlet	< 7 × 10 ⁻²
-KU3-3	September 3, 2014	Cesium absorption apparatus outlet	< 5 × 10 ⁻²
-KU4-3	March 9, 2015	Cesium absorption apparatus outlet	< 5 × 10- 2
-SA2-2	August 13, 2013	Second cesium adsorption apparatus (SARRY) System-B outlet	< 5 × 10 ⁻²
-SA2-4	February 11, 2014	Second cesium adsorption apparatus (SARRY) System-B outlet	< 5 × 10 ⁻²
-SA3-2	August 5, 2014	Second cesium adsorption apparatus (SARRY) System-B outlet	< 4 × 10 ⁻²
-AR3-1	July 13, 2011	Decontamination system outlet	< 5 × 10 ⁻²

Fig. 1 Method of analyzing  $^{79}\mbox{Se}$  developed for the nitric acid solution system





(a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis results Adsorbent)

- During the early days of the accident, sandbags containing zeolite and activated carbon got introduced into the Process Building (PMB) and the High Temperature Incinerator Building (HTI)*. These sandbags were sequentially analyzed and data on activated carbon was obtained (Fig. 1).
- ⁶⁰Co, ⁹⁰Sr, ¹²⁵Sb, ¹³⁴Cs, ¹³⁷Cs, Pu, ²⁴¹Am and ²⁴⁴Cm were detected. (Table 1)
- As compared to zeolite, except ⁹⁰Sr, ¹³⁴Cs, ¹³⁷Cs, the concentration of activated carbon was high.



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Table 1 Results of analyzing the samples of activated carbon from the sandbags  $(Bq/g)^{*1}$ 

		⁵⁴ Mn	⁶⁰ Co	⁹⁴ Nb	¹³⁴ Cs	¹³⁷ Cs	¹⁵⁴ Eu	¹²⁵ Sb
No.	Sample number	(Approx. 312 days)	(Approx. 5.3 years)	(Approx. 2.0 × 10 ⁴ years)	(Approx. 2.1 years)	(Approx. 30 years)	(Approx. 8.6 years)	(Approx. 2.8 years)
1	PMB-AD-CH-1	< 5x10¹	$(1.0\pm0.1)$ x10 ³	< 2x10 ¹	(3.3±0.1)x10 ⁴	(5.4±0.1)x10⁵	< 3x101	(7.9±1.0)x10 ²
Refer ence	Sandbag zeolite ^{*2} PMB-Ze-01	ND	ND	ND	(8.5±0.1)x10 ⁶	(1.4±0.1)x10 ⁸	ND	ND

		⁹⁰ Sr	²³⁸ Pu	²³⁹ Pu+ ²⁴⁰ Pu	²⁴¹ Am	²⁴⁴ Cm
No.	Sample number	(Approx. 29 years)	(Approx. 88 years)	(Approx. 2.4 × 10 ⁴ year, Approx. 6.6 × 10 ³ year)	(Approx. 4.3 × 10 ² year)	(Approx. 18 years)
1	PMB-AD-CH-1	(5.8±0.1)x10 ⁵	(1.6±0.1)x10 ⁰	(5.2±0.3)x10 ⁻¹	(5.6±0.1)x10 ⁰	(1.8±0.1)x10 ⁰
Refere nce	Sandbag zeolite ^{*2} PMB-Ze-01	$(1.1\pm0.1)x10^7$	ND	ND	(2.4±0.6)x10 ⁻¹	ND

*1 Activity concentration was calculated based on the fragmented mass. Activity concentration was corrected on the sampling day (February 27, 2020). The numerical value after  $\pm$  in the analytical value is the error in coefficient value.

*2 Activity concentration value is the value corrected on the sampling day (February 12, 2020)



# (a) Efficient characterization ① Investigation on analytical inventory estimation methods

i. Implementation of analysis (Analysis results Iron co-precipitation slurry)

- 4 samples of the iron co-precipitation slurry from the Advanced Liquid Processing System (ALPS) were analyzed. (Table 1, Figure 1)
- During the analysis, the samples were dissolved in acid (nitric acid) and in alkali fusion (a mixed flux comprising sodium peroxide and sodium carbonate), and the results were compared. Even though the status of dissolution differed, the results of radioactivity analysis by means of dissolution in acid and those in the case of alkali fusion were the same.
- In all samples Sr-90 concentration was dominant, and the analysis results indicated similar tendencies. Pu-239, Pu-239+240 and Am-241 were detected in all samples, and Cm-244 was detected in one of the samples.



Fig. 1 Appearance of the analysis samples

Table 1 Iron co-precipitation slurry analysis samples

		Water treatment results			Sampling		
Sample number	Sample name	Start date of passing water	End date of passing water	(mSv/h)	Sampling date	Sampling locations (HIC number)	Collection location
EAL-S6-1	Existing ALPS C system Iron co-precipitation slurry	July 5, 2014	July 10, 2014	0.785	October 15, 2018	Second facility (PO641180-55)	In the vicinity of the center of HIC (Depth 1000mm)
EAL-S8-1	Existing ALPS B system Iron co-precipitation slurry	October 29, 2014	November 3, 2014	2.093	October 16, 2018	Second facility (PO646393-109)	In the vicinity of the center of HIC (Depth 1000mm)
EAL-S7-1	Existing ALPS B system Iron co-precipitation slurry	March 14, 2014	April 19, 2014	1.0	October 17, 2018	Second facility (PO641180-11)	In the vicinity of the center of HIC (Depth 1000mm)
EAL-S10-1	Existing ALPS A system Iron co-precipitation slurry	November 11, 2013	November 18, 2013	3.612	October 18, 2018	Second facility (PO625899-054)	In the vicinity of the center of HIC (Depth 1000mm)
Reference AL-S1	Existing ALPS Iron co-precipitation slurry	_	June 5, 2014	0.633	June 5, 2014	Existing ALPS (PO641180-96)	In the vicinity of the center of HIC (Depth 1000mm)



### (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis results Iron co-precipitation slurry)

- Based on the transport ratio of Sb-125 using the Sr-90 standard, it was found that there was a strong correlation between the migration behavior of both nuclides. (Fig. 1)
- The transport ratio of Pu-238 and Pu-239+Pu-240 and Cm-244 using the Sr-90 standard suggested that the migration behavior differed.
- The variation in the transport ratios of Sb-125 and α emitting nuclides was smaller in the case of the Sr-90 standard as compared to the Cs-137 standard.
- Fe was the main chemical component of the slurry along with Si, Ca, Zn, etc. (Fig. 2)



Fig. 1 Transport ratios of radionuclides present in the iron co-precipitation slurry

Fig. 2 Chemical composition of the iron co-precipitation slurry (chemical compounds are assum





## (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Analysis results Concentrated liquid waste slurry, carbonate slurry)

- The concentrations of Pu, Am and Cm nuclides in the concentrated liquid waste slurry and carbonate slurry from ALPS have been obtained in the past, and in addition 6 samples were analyzed for U and Np nuclides (Fig. 1).
- ²³⁷Np concentration was in the range of 0.105 0.68 Bg/g. ²³⁶U was detected in all the samples, and thus the samples contained fuel components. With ²³⁶U as reference, when the transport ratio to slurry was calculated, it was found that the transport ratio for ²³⁷Np was about 10 to 300, which was equivalent to or greater than the transport ratio of contaminated water (stagnant water and treated water) (Fig. 2). It became clear that ²³⁷Np which migrated along with the contaminated water migrated to these slurries.



Before heating



After heating

Fig. 1 Appearance of the carbonate slurry sample

1.0E+03



1.0E+02 • 0 0 0 0 0 0 0 0 0 0 0 0 Transport ratio ²³⁶U as reference ^{00+30°} ********************* 0 0 Data on Np obtained this year 8 0 0 0  $\odot$ 0 0 ۲ 1.0E-01  $\circ$ 0 0 0 0 0 ۲ 0 0 1.0E-02 0 0 1.0F-03 Contaminated water Slurry

Fig. 2 Transport ratio of actinide nuclides to the contaminated water and slurries (²³⁶U as reference)

353

237Np

© 238Pu 🛛 © 239Pu

# (a) Efficient characterization ① Investigation on analytical inventory estimation methods

### i. Implementation of analysis (Analysis results Adsorbent)

- In the Advanced Liquid Processing System (ALPS) the adsorbent used in the multi-nuclide removal system (treatment column) becomes secondary waste. Multiple adsorbents are used, and the nuclides that used adsorbents contain different adsorbent depending upon the type. Hence, it is important to analyze the radioactivity for each adsorbent in order to investigate treatment and disposal methods. Samples from resin based adsorbents 1 and 2 collected from the storage container (HIC) and ferrocyanide adsorbent were analyzed.
- The method of analyzing each adsorbent was investigated prior to the analysis (Fig. 1, 2, 3).
- ⁶⁰Co, ¹²⁵Sb, ¹³⁴Cs, ¹³⁷Cs were detected in all the adsorbents. ⁹⁰Sr was detected only in the resin based adsorbent 2. ¹⁰⁶Ru was detected in the resin based adsorbent 1 and 2. α nuclides (Pu, Am, Cm) were not detected.





# (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Database of analysis results)

- The obtained analysis results were recorded in the database of analysis results "FRAnDLi" and thus data was expanded on an ongoing basis.
- The results of radioactivity analysis of the contaminated water, the quantity of treated water and the amount of waste published by TEPCO, as also the data on radioactivity analysis, element analysis, etc. obtained during this project, were recorded. Data on 109 analysis samples was recorded this year. (Table 1)
- FRAnDLi will continue to be released on the Internet so that it will be available for use to the concerned parties. (Fig. 1)

Classifi cation	San	nple	Data obtained from	Number of cases
Analysi	Dobrio	Rooftop rubble	This project	4
S	Deblis	Building sludge	This project	8
			This project	54
	Contaminated	Stagnant water	Tokyo Electric Power Company	24
	water		This project	5
		Treated water	Tokyo Electric Power Company	9
	Secondary waste generated from	Slurry	This project	4
	contaminated water treatment	Adsorbent	This project	1
Amount	Amount of stored treated w	waste, quantity of ater, etc.	Tokyo Electric Power Company	1176

Table 1 Data recorded (As of end of February 2022)

* Fukushima Daiichi Radwaste Analytical Data Library (FRAnDLi) URL:https://frandli-db.jaea.go.jp/FRAnDLi/



database (FRAnDLi) is accessed



- (a) Efficient characterization ① Investigation on analytical inventory estimation methods
   i. Implementation of analysis (Leverage in decommissioning)
- In order to reduce the risks in storage management, measures are being investigated and implemented, and analytical data is being used.





Sludge from the decontamination system

Stagnant water sludge Tokyo Electric Power Company Holdings Inc., "Status of studies aimed at α nuclide removal" Secretariat of the Team for Countermeasures for Decommissioning and Contaminated Water Treatment (86th session), January 28, 2021



Tokyo Electric Power Company Holdings Inc., "Status of studies aimed at stabilization treatment of ALPS slurry" The Commission on Regulation of Radioactive Waste in the Specified Nuclear Facilities (7th session), Document 2, July 23, 2018.



#### Zeolite sandbag

Tokyo Electric Power Company Holdings Inc., "Results of basement environment investigation in the Process Main Building" Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water (93rd session), August 26, 2021

ALPS Slurry Tokyo Electric Power Company Holdings Inc., "Design aimed at slurry stabilization treatment" The Commission on Supervision and Evaluation of the Specified Nuclear Facilities (88th session), Document 1-2-1, February 22, 2021.



# (a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Samples database)

- Data on the source and properties of the samples is important while using the analytical data. Also, information on stock is essential for managing the analysis samples. Hence, a database for managing analysis samples was considered.
- The information recorded in the samples database is closely related to waste management and analysis supply chain in 1F (Fig. 1), and is present in multiple formats (numbers, text, photos, drawings, etc.). The software is expected to be able to manage the history concerning updates to the recorded data. Hence an independent database of analysis results was developed, based on the Wiki software (MediaWiki).



Fig. 1 Relation between waste management activities and information related to analysis



(a) Efficient characterization ① Investigation on analytical inventory estimation methods i. Implementation of analysis (Samples database)

Trial runs of the Wiki database for recording data on samples are being carried out (Fig. 1). At present, data entry is going on internally at JAEA. Data will be expanded gradually for the use of concerned parties who access the database externally (Fig. 2).



Fig. 1 Hierarchy of data in the database (temporarily called as SampleBase)



# (a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods

- Achievements up to FY2020
- Methods for estimating inventory continued to be developed and improved thus making it possible to quantitatively evaluate uncertainties along with accumulation of analytical data, and thus results conforming with the analytical data were obtained (Fig. 1). These results will be made available for future investigations on disposal, etc.
- Promising fundamental measures were investigated regarding the methods for further reducing uncertainties (Fig. 2).
- Goal
- Reduction of uncertainties in inventory estimation so that the estimation matches analytical data.
- Details of implementation
- Conducting investigations on contamination mechanism that contributes to the investigations on contamination composition of nuclides (grouping), and investigations on waste classification (population) that use the status of utilization of newly obtained analytical data and the status of generation and storage of waste as reference, with the purpose of enhancing the accuracy of estimation.
- Investigating measures for enhancing the accuracy during analytical estimation of solid waste inventory based on these, and evaluating the results.
- Indicators for the achievements of goal
- Classification based on the occurrence and storage form, etc. of solid waste is investigated and presented.
- The model of the contamination mechanism investigated last year is reviewed and shown.
- Applicability to the solid waste inventory analytical estimation method is evaluated.



Fig. 1 The estimated ALPS adsorbent inventory and analytical data (yellow mark)



Fig. 2 Investigation of new methods for estimating the inventory of secondary waste generated from contaminated water treatment (Method of estimating the decontamination factor of the previous process based on the water analysis data)


(a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods (Difference in contamination depending on the unit)

- Introduction of the method using Bayesian statistics is being considered in order to quantitatively determine waste classification.
- Parameters of the t distribution of 2 groups are stochastically obtained and the overlap of distribution is evaluated.*
- The uncertainties in distribution parameters of the nuclide migration ratio in each unit were evaluated focusing on the group of elements that are somewhat volatile (semi-volatile), as the nuclide composition of contamination. (Fig. 1)
- While the data distribution is similar in Units 1 and 3, since the distribution range in Unit 2 is wider as compared to Units 1 and 3, classification into Unit 2 and Units 1/3 is considered to be worth investigating.
- However, since the data deviates depending on the Unit from where the analysis samples are obtained, these results are not definitive.
- Waste classification methods that consider difference in contamination depending on the unit have promising prospects for quantitative evaluation.



Fig. 1 Breakdown of the migration percentage of semi-volatile nuclides (H-3) to the air depending on the Unit *The purple section indicates credible interval (in Bayesian statistics). Dark color: 25-75%tile, Light color: 2.5-97.5%tile

* John Kruschke, "Bayesian Estimation Supersedes the t-Test," Journal of Experimental Psychology, 142(2), 573–603 (2013).

# (a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods (Difference in contamination depending on the element)

- The method of waste classification that takes into consideration the difference in contamination depending on the unit as explained in the previous section, can be considered for application with respect to the difference in contamination depending on the element as well.
- Regarding volatile nuclides, since the behavior of C in terms of transport ratio is different as compared to Se and I, the application is being carried out on a trial basis. (Fig. 1)
- Among the volatile nuclides, the distribution of Se and I is similar to each other, however, C has a wider range of distribution and shows an altogether different picture. Hence, it is believed that it would be appropriate to separate C into a different group.
- In this manner, trials are being continued.



Fig. 2 Difference in distribution parameters of transport ratio of volatile nuclides *The purple section indicates credible interval (in Bayesian statistics). Dark color: 25-75%tile, Light color: 2.5-97.5%tile

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volatile nuclides

- (a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods (High radiation rubble)
- In the past, inventory estimation was carried out for the large amount of rubble categorized by dose rate. However, it is desirable to perform estimation for waste that is actually stored. Whenever waste is generated in the form of rubble or felled tree, it is recorded in the Management Form and managed. Considering that these forms would be used, investigations were conducted on development of methods for estimating contamination distribution of high radiation rubble, while referring to data on surface dose rate of the rubble, etc. (metal, concrete) that is stored.
- Rubble that is stored in containers and is categorized under surface dose rate 30 mSv/h or more, is stored in the solid waste storage facility and managed (Fig. 1). It was assumed that the rubble gets mixed and averaged out in the accumulation zone, and since it is likely to have been generated indoors, attempts were made to calculate the nuclide migration using the percentage of emission in air inside the building (Fig. 2).



Fig. 1 Distribution of the surface dose rate of the stored waste

Fig. 2 Approach towards developing the high radiation rubble inventory estimation method





(a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods (Reflection of increase in data)

- The inventory of waste is calculated once again by adding the data obtained last year and obtaining the nuclide migration percentage again.
- Quantity of data is increased by adding data obtained last year (Table 1). By doing so, the parameters (average and dispersed) of nuclide migration percentage improve on the whole.



Fig. 1 Yearly shift in the nuclide migration parameters (average value µ in the case of normal distribution)

#### (a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods (Contamination mechanism)

- Investigations continue to be carried out focusing on the difference in medium (air, water) as the contamination mechanism of radionuclides. Analytical data that serves as reference for the process of settling down and stabilization of nuclides is being accumulated.*
- Data on borings cores or samples that have cracks is available for rubble contamination (Fig. 1). In the case of contamination  $\bigcirc$ through water, the composition of nuclides and elements in the deposits present in stagnant water are reported (Analysis results, stagnant water sludge), and reports related to penetration of  $\alpha$  nuclides such as Pu, etc. to concrete have been made^{*1}. The data suggests stability of the nuclide composition during the process of migration through air, and in the case of contamination through water, the possibility of precipitation after briefly dissolving in water.
- Topics pertaining to the mediums of air and water that need to be handled while further investigating contamination mechanism were clarified and identified (Fig. 2). Correction of data Addition of data obtained through





Summary

experiments

obtained through

experiments

Fig. 2 Correlation diagram of the topics to be investigated related to contamination (Example: contamination through air)

*1 G. Igarashi et al., Journal of Advanced Concrete Technology, 19(9), 950-976 (2021).

likely to be penetrated

# (a) Efficient characterization ① Investigation on analytical inventory estimation methods ii. Investigation on analytical inventory estimation methods (Reflection of improvement measures)

- Element group classification of nuclide migration parameters was carried out on a trial basis, the results were reflected and trial calculation of the inventory was carried out (Fig. 1).
- It is presumed that improved results of the element group are reflected in the estimated value of C, Se and I, and expanded analytical data is reflected in the estimated value of Co, Ni, Ag and Tc.
- The element groups were further broken down based on indoor air emission ratio, outdoor emission ratio and stagnant water migration ratio by determining the uneven distribution using Bayesian estimation focusing on nuclides that were identified as important with respect to disposal safety.
- The results of trial calculation are meant to facilitate imagination of the progress when classification is carried out. It should be noted that they have not been verified by means of benchmarking, etc. of actual waste.



Fig. 1 Results of estimating concrete rubble (obtained rough surface dose rate class < 0.1 mSv/h)





## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan

- Achievements up to FY2020
- As a requirement of the process of waste management, analysis has been carried out mainly for the purpose of establishing waste classification and improving the accuracy of inventory evaluation that contributes to the investigation of treatment and disposal technology (Fig. 1).
- Characterization was carried out with implementation of analysis and estimation of inventory at the core, and in response to the outcome of the investigations on disposal concepts, the nuclides to be analyzed were re-examined (Fig. 2).
- Goal
- To propose the policy for characterization from the FY2021 milestone onwards.
- Details of implementation
- Organization of information related to the analytical capacity (constraints such as evaluation items, volume handled, etc. of waste, nuclides, etc. that can be analyzed) of the facilities located in Ibaraki prefecture that have conducted analysis in the past as also Building 1 of the Analysis and Research Center, as analysis facilities that can be used in the mid-and-long-term.
- Creation of the mid-and-long-term analysis plan considering the status of investigations on the requirements mentioned in ii later in this document, along with waste samples and analytical data obtained so far and the decommissioning process, and using the results of the trial analysis plans mentioned in iii later in this document.
- Indicators for the achievements of goal
- Mid-and-long-term analysis plan is created aiming for characterization of solid waste.



## Fig. 1 Change in the purpose of analysis associated with the progress in decommissioning



Fig. 2 Mid-and-long-term analysis plan during the flow of characterization





## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Method of investigation)

- Analytical data is being accumulated aiming for final disposal of waste. The analytical data will be used gradually while investigating inventory evaluation, waste classification and waste verification methods (Fig. 1 on the previous page).
- The score of the analysis is roughly the product of the scores required until the type of waste and final waste verification method is determined, and forms the foundation for the mid-and-long-term analysis plan (Fig. 1).
- The score of the samples needs to be several 1000 points. For performing radiochemical analysis of the difficult-to-measure Ο nuclides, analysis needs to be continued for the long-term with 10 years as the unit for the duration.*
- Meanwhile, there are uncertainties in the waste classification and the sample score depending on the purpose of analysis. Hence, investigations were carried out to review the mid-and-long-term analysis plan based on the analysis results, information on waste and progress of decommissioning, etc. achieved so far.
- With regards to the analysis score, the purpose of the analysis was specifically investigated (Fig. 2). Also, waste classification was implemented using the analytical inventory estimation method, and quantitative evaluation of the score was implemented using the analysis planning method respectively.



Fig. 1 Calculation of the analysis score at present

*Tokyo Electric Power Company, "Ability to analyze Fukushima Daiichi accident waste"

The Commission on Regulation of Radioactive Waste in the Specified Nuclear Facilities, 2nd session, Document 4, February 12, 2016.



#### (a) Efficient characterization ② Proposal mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Method of investigation)



#### IRID

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## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Purpose of analysis)

There is a gradual shift in the requirements (purpose of analysis) pertaining to characterization while pursuing research & development.

■ The purpose of analysis suitable for waste management is set assuming term 3–① and considering the approach of analysis for each waste.

- A) Estimation of activity concentration distribution and nuclide composition
  - The activity concentration distribution and nuclide composition is estimated based on analytical data, and the nuclide composition is evaluated based on transport ratio.
  - The nuclide migration parameters are set using the analytical inventory estimation method, and the inventory is estimated.
- B) Investigation of applicability of the scaling factor method
  - The migration process of the main nuclides is investigated based on the progression of events at the time of accident and the analytical data, physical and chemical properties, etc.
  - Bearing in mind the relation between the migration process of nuclides and the scaling factor method (SF method), the correlation with key nuclides (¹³⁷Cs and ⁶⁰Co, etc.) is investigated.
  - The inventory is estimated by using the accumulated analytical data to reduce the uncertainties in nuclide migration parameters in the analytical inventory estimation method, and setting the element groups.
- C) Rational grouping
  - Grouping (splitting or combining populations) is investigated using Bayesian statistics and with the nuclide composition, SF applicability and tentative disposal constraints as the indicators.
- D) Selection of the methods for determining activity concentration
  - The applicability of the methods for determining activity concentration is investigated using Bayesian statistics, and methods for determining activity concentration are presented for each main waste.
- E) Determination of waste verification method / Determination of groups
  - The method for determining activity concentration is established, and the process moves on to full-fledged actions towards implementation of disposal.





Key nuclide concentration





Next term

lerm

## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Waste priority indicators)

The purpose of analysis of each waste and indicators related to waste management were scored for each waste, and the product of those scores was considered as the indicator of priority of the wastes.

			<u> </u>	
		Details	Priority and quantification	Setting of indicators (numerical value)
	A) Estimation of activity concentration distribution and nuclide composition	The presence of analytical data (results) to be made available for the analytical method in order to estimate the activity concentration distribution and composition, is considered as the indicator.	Waste that has not been analyzed so far is prioritized. Depends on whether analysis results are available.	1: Analysis results are not available 0: Analysis results are available
Purpose of :	B) Investigation of applicability of the scaling factor method	The applicability of the scaling factor method (SF method) as a method for determining activity concentration is investigated on a priority basis. Applicability of SF method evaluated qualitatively based on existing information on waste, is considered as the indicator.	In order to determine applicability of the SF method, waste that has a high regularity of contamination is prioritized. Depends on the stage at which the SF method is expected to be applicable.	<ol> <li>Waste to which the SF method is considered to be applicable.</li> <li>Waste to which the SF method is considered to be applicable by splitting the population.</li> <li>Waste to which it is difficult to apply the SF method.</li> </ol>
analysis	C) Rational waste	Since the variation in the waste itself and the waste storage container have a strong impact on the evaluation of the population, these variations are considered as indicators.	The regularity of contamination seems to be comparatively higher, but waste with large variations is prioritized. Depends on the necessity to take measures to reduce variation.	<ul><li>2: Waste with large variation, for which measures need to be taken to reduce variation during the treatment process, etc.</li><li>0: Waste that does not require any measures.</li></ul>
	(Grouping)	The uncertainties in inventory estimation have a strong impact on the uncertainties in disposal category. Hence this is considered as an indicator.	Waste that has high uncertainties in disposal category is prioritized. Depends on the fluctuation in disposal category due to inventory reference value.	<ol> <li>Waste in which the disposal category fluctuates due to inventory reference value (50%tile, 95%tile).</li> <li>Waste in which the category remains constant.</li> </ol>
Was	Risk reduction	The necessity to reduce risks during storage in Term 3-① is considered as an indicator.	Waste that is likely to undergo change in properties due to the stabilization treatment, etc. is prioritized. Depends on whether or not measures are taken during Term 3-①.	2: Measures taken 0: Measures not taken
ste managem	Reduction in storage load	The necessity to reduce load by taking measures such as consolidating storage, reduced volume storage, etc. during Term 3-① is considered as an indicator.	Waste that is likely to undergo change in properties due to load reduction measures is prioritized. Depends on whether or not measures are taken during Term 3-(1).	2: Measures taken 0: Measures not taken
hent	Amount	Since waste that is large in quantity has an impact on overall management plan, the amount of waste is considered as an indicator.	Waste that is large in quantity is prioritized, and 3 stages are set.	3: Large (exceeding 10,000 m ³ ) 2: Medium (several 1000 m ³ ) 1: Small (several 100 m ³ )

#### Table Indicators for determining the order of priority for analysis (Proposed)



## (a) Efficient characterization (2) Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Priority related to disposal)

The estimated values of the inventory are statistically distributed. They are used as reference values (50% tile, 95%tile, geometric mean, etc.) for disposal safety evaluation (Table 1). As a result, in the case of waste for which the disposal category fluctuates (yellow portion in Table 1), as those distributions are close to the disposal constraints, reducing uncertainties by means of analysis is strongly required. Hence it is believed that analysis should be conducted on a priority basis.

Type of waste	Amount	Disposal category (50%tile / 95%tile) based on nearing	Disposal category (b0%/ie (Barrier present/absent)/(95%/ie (Barrier present / absent)) based on the reduction factor of the impact of salts as against the distribution coefficient of instant release in a groundwater scenario			Type of waste	Amount	Disposal category (50%tile / 95%tile) based on nearing	Disposal category ([50%ile (Barrier present/absent))/[95%ile (Barrier present / absent]) based on the reduction factor of the impact of salts as against the distribution coefficient of instant release in a groundwater scenario		
	(m)	years, pit 300 years)	Facility	Natural barrier	Facility and natural barrier		(m)	years, pit 300 years)	Facility	Natural barrier	Facility and natural barrier
JRION	2847	L1/L1	[L2/L3]/[L3/L3]	[L3/L3]/[L2/L3]	[L2/L2]/[L2/L2]	Metal inside the building (equivalent to L3 before the accident)	23,677	L1/L1	[L1/L2]/[L1/L2]	[L1/L2]/[L1/L2]	[L1/L2]/[L1/L2]
REVA	597	L1/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L2/L3]	Metal inside the building (non-radioactive waste before the accident)	6,454	L1/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
ARRY	1196	L1/L1	[L2/L3]/[L3/L3]	[L3/L3]/[L2/L3]	[L2/L2]/[L2/L2]	Concrete inside the building (equivalent to L3 before the accident)	8,766	L1/L1	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]
urry 1 Iron co-precipitation	1,833	L1/L1	[L3/L3]/[L1/L2]	[L3/L3]/[L1/L2]	[L2/L3]/[L1/L2]	Concrete inside the building (non-radioactive waste before the accident)	370,586	L1/L1	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L1/L2]
urry 2 Carbonate precipitation	7,887	L1/L1	[L2/L2]/[L1/L2]	[L2/L2]/[L3/L3]	[L2/L2]/[L1/L2]	Waste inside RPV (L1-debris before the accident)	163	Strata/Strata	[L1/L1]/[L1/L1]	[L1/L1]/[L1/L1]	[L1/L1]/[L1/L1]
PS-4 Ag impregnated activated carbon	222	L1/strata	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]	Waste inside RPV (L1-stagnant water before the accident)	67	Strata/Strata	[L1/L1]/[L1/L1]	[L1/L1]/[L1/L1]	[L1/L1]/[L1/L1]
.PS-2 Titanate	332	L2/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	Waste inside RPV (L2-debris before the accident)	131	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L1]/[L1/L1]
PS-3 Ferrocyanide	222	L2/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	Waste inside RPV (L2-stagnant water the accident)	55	L1/strata	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
.PS-6 Chelating resin	443	L2/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	Metal inside PCV (L2-debris before the accident)	176	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]
PS-5 Titanium oxide	222	L3/L2	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	Metal inside PCV (L2-stagnant water before the accident)	988	L1/strata	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]
.PS-7.1	46	L2/strata	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	Metal inside PCV (L3-debris before the accident)	181	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]
ubble 1 (<0.0005mSv/h)	190,000	L2/L2	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	Metal inside PCV (L3-stagnant water before the accident)	1,030	L1/strata	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]
ubble 2 (0.005 to 0.1mSv/h)	48,000	L2/L2	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	Concrete inside PCV (L2-debris before the accident)	66	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]
ubble 3 (0.1 to 1mSv/h)	26,700	L1/L1	[L2/L2]/[L1/L2]	[L2/L2]/[L1/L2]	[L2/L2]/[L1/L2]	Concrete inside PCV (L2-stagnant water before the accident)	365	L1/strata	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
ubble 4 (1 to 30mSv/h)	52,000	L1/L1	[L1/L2]/[L1/L2]	[L1/L2]/[L1/L2]	[L1/L2]/[L1/L2]	Concrete inside PCV (L3-debris before the accident)	68	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]
ubble 5 (>30mSv/h)	36,000	L1/L1	[L1/L2]/[L1/L2]	[L1/L2]/[L1/L2]	[L1/L2]/[L1/L2]	Concrete inside PCV (L3-stagnant water before the accident)	381	L1/strata	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
elled trees	18,000	L1/L1	[L2/L2]/[L1/L2]	[L2/L2]/[L1/L2]	[L2/L2]/[L1/L2]	Metal inside the building (L3 air pollution before the accident)	20,143	L2/L2	[L2/L2]/[L1/L2]	[L2/L2]/[L1/L2]	[L2/L2]/[L1/L2]
aste inside RPV (L1 before the accident)	230	Strata/Strata	[L1/L1]/[L1/L1]	[L1/L1]/[L1/L1]	[L1/L1]/[L1/L1]	Metal inside the building (L3 stagnant water before the accident)	3,534	L1/L1	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]	[L2/L2]/[L2/L2]
aste inside RPV (L2 before the accident)	185	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L1]/[L1/L1]	Metal inside the building (non-radioactive air pollution before the accident)	5,491	L2/L2	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]
etal inside PCV (L2 before the accident)	1,164	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]	Metal inside the building (non-radioactive stagnant water before the accident)	963	L1/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
etal inside PCV (L3 before the accident)	1,211	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]	Concrete inside the building (L3 air pollution before the accident)	7,444	L2/L2	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]	[L3/L3]/[L3/L3]
oncrete inside PCV (L2 before the accident)	431	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]	Concrete inside the building (L3 stagnant water before the accident)	1,322	L1/L1	[L3/L3]/[L3/L3]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
oncrete inside PCV (L3 before the accident)	449	Strata/Strata	[L1/L2]/[L2/L2]	[L2/L2]/[L1/L2]	[L1/L2]/[L1/L2]	Concrete inside the building (non-radioactive air pollution before the	315,069	L2/L2	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]	[L3/L3]/[L2/L2]
FY2020 "Provision of disposal	concept	s and develop	ment of safet	y assessmen	it methods"	Concrete inside the building (non-radioactive stagnant water before	EE E17	14/14	[[ 2/  2]/[] 2/  2]	[[ 2/  2]/[] 2/  2]	[[ 2/[ 2]/[] 1/[ 2]

#### Table 1: Impact of inventory reference value (50%tile / 95%ile) on disposal category*



(a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Priority pertaining to waste management)

High priority waste was set based on the prospects of the status of management in Term 3-① in light of the indicators (proposed) for determining the order of priority of analysis.



considered as a pre-condition (Example) Likelihood of Waste change in Indicators of waste management in Waste nuclide Term 3-(1) management composition Volume reduction Reduction in storage load Waste stored outdoors treatment Yes (volume reduction) Indoor storage Waste stored in solid waste Continuation of storage facility No storage Incineration Reduction in storage load Flammable waste disposal Yes (volume reduction) Indoor storage Contaminated soil Indoor storage No Stabilization Risk reduction Spent adsorbent slurry, etc. treatment Reduction in storage load Yes Volume reduction (volume reduction) treatment Stabilization Risk reduction treatment Waste sludge Reduction in storage load Yes Volume reduction (volume reduction) treatment Stabilization Risk reduction treatment Concentrated liquid waste Reduction in storage load Yes slurry Volume reduction (volume reduction) treatment

Table 1 Status of waste management that must be

(*1 Tokyo Electric Power Company Holdings Inc., "Tokyo Electric Power Company Holdings Inc. Fukushima Daiichi Nuclear Power Station solid waste storage management plan, July 2021 edition", July 29, 2021)

	-	-	-	-	
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,				-	



(a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Example of trial run of the priority investigation)

- When evaluation was carried out on a trial basis, the priority level of KURION, SARRY, ALPS slurry / adsorbent, felled trees, etc. turned out to be high.
- In order to steadily continue with the 1F waste stream, the stakeholders need to examine all aspects including the indicators and confirm the priority.

			Demolition waste inside the containme nt vessel	Waste incidental to debris retrieval	Building concrete waste	Building metal waste	Rubble (stored in containers)	Rubble (accumulat ed / low contaminati on)	Protective clothing	Felled trees	KURION	SARRY	AREVA sludge	ALPS slurry	ALPS adsorbent	Concentrat ed liquid waste slurry	Activated carbon zeolite sandbag
P	A) Estimation of activity concentration distribution and nuclide composition	Actual results of analysis	Not available (1)	Not available (1)	Available (0)	Available (0)	Not available (1)	Not available (1)	Available (0)	Not available (1)	Not available (1)	Not available (1)	Available (0)	Available (0)	Available (0)	Available (0)	Available (0)
urpose of analysis	<ul> <li>B) Investigation on applicability of SF method</li> </ul>	Applicability of SF method	Difficult (0)	Difficult (0)	Splitting (1)	Splitting (1)	Splitting (1)	Applicable (2)	Applicable (2)	Applicable (2)	Splitting (1)	Splitting (1)	Applicable (2)	Splitting (1)	Splitting (1)	Applicable (2)	Applicable (2)
	C) Rational waste	Measures for reducing variation	Required (2)	Required (2)	Required (2)	Required (2)	Not required (0)	Required (2)	Not required (0)	Required (2)	Required (2)	Required (2)	Not required (0)	Required (2)	Required (2)	Not required (0)	Not required (0)
	(Grouping)	Change in disposal category	Present (1)	Present (1)	Present (1)	Present (1)	Absent (0)	Absent (0)	Absent (0)	Present (1)	Present (1)	Present (1)	Present (1)	Present (1)	Present (1)	Present (1)	Present (1)
		Sub-total	4	4	4	4	2	5	2	6	5	5	3	4	4	3	3
W	Risk reduction		Absent (0)	Absent (0)	Absent (0)	Absent (0)	Present (2)	Absent (0)	Absent (0)	Absent (0)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)
aste ma indic	Reduction in storage I	load	Absent (0)	Absent (0)	Absent (0)	Absent (0)	Present (2)	Absent (0)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)	Present (2)	Absent (0)
anagem ators	Amount		Medium (2)	Small (1)	Large (3)	Large (3)	Large (3)	Large (3)	Large (3)	Medium (2)	Medium (2)	Medium (2)	Small (1)	Medium (2)	Medium (2)	Small (1)	Small (1)
lent		Sub-total	2	1	3	3	7	3	5	4	6	6	5	6	6	5	3
		Total	8	4	12	12	14	15	10	24	30	30	15	24	24	15	9

#### Table 1 Investigation of the order of priority for analysis (Example)



## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Plan for construction works, etc.)

- In this project, sampling of rubble and secondary waste generated from contaminated water treatment was planned and carried out in coordination with construction works (List 1) and equipment operation (① Investigation on analytical inventory estimation methods, i. Implementation of analysis).
- Coordination with construction works and equipment operation is essential for acquiring samples in an efficient manner. Hence a short term plan will be put in place.
- The plan for the mid- and long-term will be investigated using the plans for construction works, research & development, etc. as reference.
- Specific construction methods, schedules related to generation of waste will be considered using the mid-and-long-term plans as reference (Table 1).
- A sampling plan will be investigated in conjunction with TEPCO.
- List 1 Works during which samples, etc. were acquired.

•Units 1 to 3

- Environment improvement around the nitrogen gas loading facility
- RCW system dose reduction measures
- Debris removal from the southern side of Unit 3
- Flange tank decontamination / storage
- Collection of flammable materials / hazardous materials

•Temporary storage area C

- New construction of water discharge channel
- Construction of access roads within the premises
- New installation of communication lines within the premises
- Repairs of outer weir drain valve control panel



* Tokyo Electric Power Company Holdings, Inc., :FY2021 Mid-and-long-term Decommissioning Plan", March 25, 2021





Table 1 Plan for the mid- to long-term when various types of waste will be generated*

#### (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Actual results of analysis and resources)

- The rubble samples for analysis are obtained from the reactor building mainly from tasks such as construction works, etc.. Planned sampling is limited to low dose rate areas (Unit 4). Coordination with the decommissioning plan is important for full-fledged sampling from the high-dose rate areas.
- In Term 3-① from FY2022, in addition to the analyses conducted at facilities in the Ibaraki area (Table 1), analysis will start being conducted at the Radioactive Substances Analysis and Research Facility Building 1. This is considered to be the precondition for the analytical capabilities for the time being.



Fig. 1 Actual results of analysis of samples collected from inside the reactor building



### (a) Efficient characterization ② Proposal mid-and-long-term analysis plan i. Development of mid-and-long-term analysis plan (Analysis plan)

- The waste anticipated to be generated based on the mid-and-long-term plan will be investigated, and a specific analysis plan will be developed.
- Since fuel debris retrieval will gain momentum, specific sampling methods, number of samples, etc. will be investigated pertaining to sampling during the process of construction (Improvement of environment within and outside the buildings) which will be carried out assuming the samples will be acquired from high-dose rate regions.
- The ballpark analysis scores will be kept under control as before (waste classification x standard scores), and they will be concretized and quantified by means of the DQO process.
- It is desirable for concerned parties to carry out investigations by bringing together specific information that would contribute to planning.

#### FY2021 FY2022 FY2023 FY2032 Manufacturing, installation and retrieval of collection device for floor surface sludge, etc Investigation of methods for collecting floor surface sludge Waste from contaminated water countermeasures DQO "Floor surface sludge" Designing measures such as for zeolite sandbag ring & Retrieval Waste from contaminated water DQO countermeasures "Zeolite sandbag" Additional miscellaneous solids waste incineration facility Incineration of temporarily stored waste Rubble, etc. processing waste DQQ "Incineration processing waste" Volume reduction treatment facility Installation work Volume reduction treatment facility Volume reduction of temporarily stored waste Rubble, etc. processing waste DQQ "Volume reduction processing waste" Slurry stabilization treatment Installation of slurry stabilization treatment facility Secondary waste from contaminated DQO water treatment "Slurry stabilization processing waste" nit 3 High radiation equipment retrieva Debris retrieval Unit 2 High radiation equipment retrieval Unit 4 High radiation equipment Unit 1 High radiation equipment retrieval DQO Pool fuel removal waste "High radiation equipment, etc." Improvement of environment within and outside the buildings Inside the buildings: Dose reduction / Interference removal, etc. Debris retrieval waste "Waste from improvement of the

#### Table 1 Example of development of mid- and long-term analysis plan

FY2026

DOO

IDID

environment within and outside the

buildings" "Interference removal waste"

## (a) Efficient characterization 2 Proposal for mid-and-long-term analysis plan ii. Investigation of analytical goals based on requirements

- Achievements up to FY2020
- The important nuclides (Table 1) identified while Ο investigating the disposal concepts were used as reference and reflected in the analysis plan.
- The estimated inventory and analytical data was Ο compared, and along with investigating the certainty of estimation, information contributing to setting specific goals of analysis was obtained (Fig. 1).
- Goal
- To set specific goals of analysis
- Details of implementation
- Consolidating the analytical data accumulated so far, Ο the level of difficulty of the analysis methods, likelihood of alternative means of calculation, etc. along with the contamination mechanisms investigated in the previous section (1)ii and the requirements of multifaceted analyses that is considered in the next section iii.
- Identifying nuclides for which there is a stronger need Ο for analysis in the mid-and-long-term based on those results, and deriving targeted concentration, accuracy, etc. as quantitatively as possible.
- Indicators for the achievements of goal

Analytical goals based on the analytical requirements associated with decommissioning are investigated.

#### Table 1 Examples of exposure dose affecting nuclides identified while investigating disposal concepts (Cited from the FY2020 Report)

		Dose rate*		Ratio of	Ratio of	Ratio of
	Type of waste	µSv/y	Dominant nuclides	to dominant nuclides 1/10<	to dominant nuclides 1/100<	to dominant nuclides 1/1000<
Secondary waste	KURION	1.4E+02	C-14		I-129	
contaminated water	AREVA	9.1E-01	C-14		I-129	
treatment	Slurry 2 Carbonate precipitation	3.6E+01	C-14		I-129	
Rubble	Rubble 1 (0-0.005mSv/h)	5.1E+00	C-14		I-129	
	Rubble 2 (0.005 - 1mSv/h)	+01.7E+1	C-14		I-129	
Dismantling waste	RPV waste (L1 before the accident)	7.3E+03	C-14			
	Metal inside PCV (L2 before the accident)	6.5E+00	C-14	Pu-239	Tc-99, Po-210, Np-237, U-234, I-129	U-238, Pb-210, Ra-226, Pu-242, U-236
	Concrete inside the building (Non-radioactive waste before the accident)	7.5E+00	C-14		I-129	



Fig. 1 Comparison between the results of inventory estimation of ALPS carbonate slurry and analytical data

(a) Efficient characterization ② Proposal for mid-and-long-term analysis plan ii. Investigation of analytical goals based on requirements (Method of investigation)

- The nuclides to be analyzed for the representative / high priority waste and the target concentration during analysis were set as the goals of analysis.
- In the past, analysis was conducted by selecting 38 nuclides using the domestic track record as reference.
- This is assumed in the design and operation of the Radioactive Substances Analysis and Research Facility Building 1 as well. For inventory estimation, nuclide selection was more extensive. 134 nuclides were selected and evaluated.
- In FY2016, nuclides were selected on a trial basis. The number of target nuclides was reduced from 38 to 30 and it was proposed that 23 nuclides be selected based on calculations.
- The outcome of (i) was used as reference for the purpose of analysis, nuclides were selected, and their respective target concentration was quantitatively investigated. (Fig. 1)



Waste		Nuclides						
А	$^{14}\mathrm{C}$	$^{90}\mathrm{Sr}$	$^{125}\mathrm{Sb}$	$^{129}\mathrm{I}$	$^{137}\mathrm{Cs}$	$^{236}\mathrm{U}$	$^{237}\mathrm{N}$	²³⁸ Pu
							р	
В	xxx	XXX	xxx		_			XXX
С	xxx	XXX		XXX	—			XXX
D	xxx	XXX	XXX	xxx	_	XXX	xxx	xxx
E	XXX	XXX		XXX	_		xxx	XXX

Fig. 1 Flow of the Investigation of analysis goals

#### IRID

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## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan ii. Investigation of analytical goals based on requirements (Selection of nuclides)

- The nuclides to be analyzed were selected once again using the methods investigated previously and the results of those investigations as reference (Fig. 1).
- As a result 47 nuclides were selected for analysis (for inventory estimation), and upon considering the results of detection and future possibilities it was thought that it would be appropriate to select 17 out of those nuclides using calculations (Table 1).





#### (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan ii. Investigation of analytical goals based on requirements (Investigation of quantitative of goals)

- The results of analysis (quantitative value, limit value) of the selected nuclides carried out so far were consolidated. These values were compared with the indicators.
- Here, during the analysis of many nuclides, as an indicator, the concentration limit in water was assumed to be 1/100, considering that solid samples were subjected to chemical separation in the form of an aqueous solution.
- As a result of organization, the nuclides for which the indicator was not achieved were identified as nuclides for which it was desirable to investigate analysis methods. (Table 1)
- O It is desirable to continue investigating the indicators based on disposal safety.

	Actual result	s of analysis		Comparison		Actual result	s of analysis		Comparison
Nuclides	Lower detection limit	Quantitative value	Underwater concentration limit / 100 (Bq/cm ³ )	between the results and concentration in the left column*	Nuclides	Lower detection limit	Quantitative value	Underwater concentration limit / 100 (Bq/cm ³ )	between the results and concentration in the left column*
H-3	3.0E-02	5.2E-02	6E-01	0.1	Cs-137	7.0E-03	1.0E-03	9E-04	1.1
C-14	2.0E-02	3.3E-02	2E-02	1.0	Eu-152	3.0E-02		6E-03	5.0
CI-36	0.004		9E-03	0.4	Eu-154	2.0E-02	3.2E-01	4E-03	5.0
Ca-41	3.0E-02	5.2E-02	4E-02	0.8	U-233	4.0E-04		2E-04	2.0
Co-58	3.0E-02	5.2E-02	1E-02	3.0	U-234	1.6E-06	8.7E-06	2E-04	0.0
Co-60	9.0E-03	7.8E-03	2E-03	3.9	U-235	7.5E-10	3.7E-09	2E-04	0.0
Ni-63	5.0E-02	5.5E-02	6E-02	0.8	U-236	7.2E-09	7.8E-09	2E-04	0.0
Se-79	4.0E-02	5.2E-02	2E-03	20	U-238	4.0E-09	6.3E-10	2E-04	0.0
Sr-89	1.3E-01	6.9E+00	3E-03	43	Np-237	5.0E-05	2.0E-05	9E-05	0.2
Sr-90	2.0E-03	4.2E-03	3E-04	6.7	Pu-238	9.0E-05	1.5E-04	4E-05	2.3
Nb-94	8.0E-03	3.4E-01	4E-03	2.0	Pu-239	1.0E-02	1.4E-02	4E-05	250
Nb-95		4.3E+02	1E-02	43000	Pu-240	1.0E-02	5.0E-03	4E-05	125
Tc-99	6.0E-03	6.4E-03	1E-02	0.6	Pu-241	5.0E-02	1.8E+02	2E-03	25
Ru-103		5.0E-01	1E-02	50	Pu-242	2.0E-06	2.0E-03	4E-05	0.1
Ru-106	4.0E+00	6.8E-02	1E-03	68	Am-241	9.0E-05	2.7E-04	5E-05	1.8
Ag-110m		1.1E+02	3E-03	36667	Am-242m	1.4E+00		5E-05	28000
Sn-126	5.0E-02	5.2E-01	2E-03	25	Am-243	8.0E-04		5E-05	16
Sb-125	7.0E-02	3.9E-03	8E-03	0.5	Cm-242	2.0E-03	-03.9E+2	6E-04	3.3
I-129	2.0E-02	2.0E-02	9E-04	22	Cm-244	9.0E-05	1.1E-04	7E-05	1.3
Cs-134	6.0E-01	1.3E-03	6E-04	2.2	Cm-245	6.8E-03		5E-05	136
Cs-136		2.5E+01	3E-03	8333	Cm-246	6.7E-03		5E-05	134

#### Table 1 Flow of the investigation of quantitative goals

* Compared with the lower detection limit or quantitative value, whichever is lower. Font color is changed to indicate that the value has exceeded 1.





## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan iii. Investigation of analysis planning methods

- Achievements up to FY2020
- The usefulness of the analysis planning method in which the DQO process considered to be the standard method in the environment restoration domain is used as the base (Fig. 1), and the method of calculating the required analysis score using the Bayesian statistical method is used in combination, was verified (Fig. 2).
- Goal

- To establish a method for quantitatively calculating the required analysis score.
- Details of implementation
- Investigating an analysis planning method in which the DQO process developed by the US and Bayesian statistical method are used in combination, considering that the population of waste to be investigated is tentative, considering the multifaceted requirements of disposal, etc. and that the statistically required analysis score is calculated.
- Selecting several representative solid wastes, and conducting trial runs with existing analytical data and the most recent analytical plan as reference. And consolidating the appropriateness for future use, and issues with respect to practical application based on the results.
- Indicators for the achievements of goal
- For waste with a tentative population, analysis planning methods using statistical techniques are investigated and their appropriateness is evaluated.







Fig. 2 Calculation of the analysis score required for verifying whether the expected criteria is met (Example)

## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan iii. Investigation of analysis planning methods (Calculation techniques to be considered)

- The method of combining the Bayesian statistical method with the DQO process continues to be considered for the analysis planning method.
- As a condition for evaluating the analysis score, preparations are underway for investigating 3 purposes (Fig. 1) in accordance with the purpose of analysis investigated as part of the mid-and-long-term analysis plan.





## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan iii. Investigation of analysis planning methods (Method of analysis)

- 3 methods were investigated related to Step 6 of evaluation of analysis score during the DQO process. The incinerated ash, carbonate slurry, and rubble were respectively investigated with respect to the concentration distribution, scaling factor method, and grouping (waste classification).
- The DQO process was implemented in a workshop format with participation by the concerned parties (TEPCO, NDF, IRID) (Fig. 1). The third session is planned to be held shortly. The progress of events is reported in this report.
- Applying Bayesian statistics for evaluating the analysis score in Step 6 was considered with the cooperation of UK's NNL. As a result of preliminary research, it was believed that it would be appropriate to carry out grouping upon comprehensive examination. With the alternative method of Kruschke's t test as the basis, the Bayesian statistical method investigates the uncertainties in the parameters of t distribution (Fig. 2) ^{*1} Calculated with the statistical computing environment R using the BEST package ^{*2}



Fig. 1 DQO process for the waste

RII



Fig. 2 Prior distribution and parameters handled by means of Kruschke's method^{*2} (Average  $\mu$  of t distribution, standard deviation  $\sigma$ , shape v)

- *1 J. Kruschke, "Bayesian estimation supersedes the t test," *J. Experimental Psychology*, 142, 573–603 (2013).
- *2 J. Kruschke, et al. "Package 'BEST, (May 18, 2020.



## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan iii. Investigation of analysis planning methods (Trial run of the DQO process)

- Calculating the analysis score for estimating the activity concentration distribution of incinerated ash discharged from the miscellaneous solid waste incineration facility" was considered as the purpose of Steps 1 and 2.
- In Steps 3 and 4, information on the incinerated ash and the incineration facility that was the source of its generation was consolidated, and the target was specified.
- O The miscellaneous solid waste incineration facility is the target. The additional facility where other objects are incinerated is not be considered as the target. The ash (approx. 2000 units) generated so far is considered for the population, and the 25 samples collected during this project will be analyzed.
- There is 10% (target value) volume reduction by means of incineration. The activity concentration and rate of generation (4, 20 kg/h) of the bottom ash and the fly ash are different. As ash gets stored in drums in the order of generation, there is variation in the ash between drums and inside the drums.



In Step 5, it was found that further analysis is required.

Fig. 1 Overview of miscellaneous solid waste incineration facility

* Tokyo Electric Power Company, "Results of the hot run during the Fukushima Daiichi Nuclear Power Station Miscellaneous Solid Waste Incineration Facility installation work" The Commission on Regulation of Radioactive Waste in the Specified Nuclear Facilities (3rd session), March 17, 2016.



#### (a) Efficient characterization (2) Proposal for mid-and-long-term analysis plan iii. Investigation of analysis planning methods (Calculation using Bayesian statistics)

- In Step 6, the analysis score was probabilistically investigated. Ascertaining the 95% tile value was considered as a parameter of concentration distribution, but it was unable to be calculated using BEST package, and needed to be programmed. Hence the Highest Density Interval (HDI) at 95% was obtained instead (Fig. 1).
- Investigation of the 95% tile (Fig. 1 top) will be considered next year.
- The prior distribution in the case of Bayesian statistics was calculated using the BEST package by referring to the dose rate of the drums and the samples collected, converting it to ¹³⁷Cs concentration and thereafter using it as the input condition (Table 1) for the calculation (Fig. 2).



Fig. 1 Modification of calculation conditions

*1 J. Kruschke, et al. "Package 'BEST, (May 18, 2020.



the BEST package (n = 2,000)

## (a) Efficient characterization ② Proposal for mid-and-long-term analysis plan iii. Investigation of analysis planning methods (Creating the analysis plan)

- In Step 6, further, the posterior distribution was calculated as the distribution of 25 samples most likely to be collected (Fig. 1). The statistical power (analysis score) was calculated while letting the expected range (95%HDI) to be 10² to 10⁴ (Fig. 2). The distribution of the samples assumed to be in the drums was wider than the collected samples. Hence many samples are required in order to obtain 95%HDI with certainty.
- Realistically speaking, calculation that assumes the current number of stored samples to be the population seems to be effective. The examination will be continued.
- In Step 7, the stakeholders carried out discussions based on this information and determined the analysis score.
- Since incineration continues, continuing sampling while the process of analysis begins, increasing the number of analysis samples and conducting evaluation once again based on the DQO process is presumable.
- The slurry and rubble will be investigated and the results will be consolidated.



Fig. 1 Posterior distribution of ¹³⁷Cs calculated by means of the BESTmcmc of the BEST package



Fig. 2 The probability of 10² to 10⁴ Bq/g calculated by means of BESTpower of BEST package, entering 95%HDI



## (a) Efficient characterization Summary

Current results

With regards to implementing the analyses, the analytical data on samples (activated carbon sandbag, iron co-precipitation slurry, ⁷⁹Se in contaminated water) for which analysis has been completed, was consolidated, it was sequentially stored in a database and released.

- With regards to inventory estimation, the likelihood of a method of classifying (grouping) elements and waste based on the concentration distribution of nuclides using Bayesian statistics was found out.
- ✓ With regards to the mid-and-long-term analysis plan, the purpose of the analysis was concretized assuming Term 3–① starting from next year.

Issues, etc.

- Continuing consolidation of analytical data
- Finalizing the mid-and-long-term analysis plan
- Completing the 3 case studies on analysis planning methods



#### Achievements up to FY2020

- Separate preliminary tests were conducted on elemental technologies (making an opening, sampling, closing an opening) that are necessary for sampling from the adsorption tower, and the design specifications of the sampling equipment were finalized.
- Simplified Integrated Sampling Module (hereinafter, S-ISM), Simplified Working Dock (hereinafter, S-WD), simulated adsorption tower, Simplified Working Stand (hereinafter, S-WS), etc. were fabricated as elemental test apparatus (Fig. 1).
- Elemental test plan was drafted, and the elemental tests were conducted using the simulated adsorption tower simulating the top portion of the adsorption tower.
- It was confirmed by means of the elemental test using S-ISM, that the series of tasks including making an opening, sampling and closing an opening can be competed in the case of the KURION adsorption tower and the SARRY adsorption tower.



Fig. 1 FY2020 elemental test equipment and overview



#### FY2021 Goals

To design and manufacture sampling system capable of being used for sampling from the 1F used adsorption towers and the required incidental facilities based on the results of the element tests, and to verify their performance. To draft a sampling plan (including work area, and items to be collected) for collecting actual samples from the 1F used adsorption towers.

#### FY2020 Details of implementation

- Fabrication of element test apparatus was completed, and element tests on making an opening in the adsorption tower, collecting samples, and closing the site of the opening were implemented.
- The results of the element tests showed prospects for sampling methods and equipment that can be used on the site.
- FY2021 Details of implementation
  - The design of the sampling system was investigated based on the results of the element tests and the investigations on conditions during sampling carried out so far.
  - Parts of the sampling system were fabricated.
  - Investigations on the sampling work flow and work area and selection of items to be collected was started.
  - Fabrication of the sampling system and the incidental facilities was continued and performance verification tests were implemented.
  - A plan (including selection of work area, selection of items to be collected) for collection from actual adsorption towers, etc. in order to verify the sampling technology was drafted.



#### Indicators for the achievements of goal

FY2021• The sampling system and the required incidental facilities are investigated,<br/>designed and manufactured, and performance tests, etc. are conducted.<br/>• Sampling plan is drafted and presented.<br/>[Target at completion: TRL: Level  $4 \rightarrow 5$ ]

TRL level 4	Stage at which functional tests are implemented at the test manufacturing level as a development and engineering process.
TRL level 5	Stage at which a prototype is manufactured based on the actual equipment and verified in a simulated environment at the plant, etc.

[Excerpts from the general information manual on subsidized projects]

#### Goals and results

	•Investigating the design of the sampling system $\Rightarrow$ Completed
	•Manufacturing the sampling system $\Rightarrow$ Completed
	•Performance verification tests of the sampling system $\Rightarrow$ Manufacturer tests
FY2021	completed. Verification tests are planned to be conducted after delivery
	•Designing the incidental facilities $\Rightarrow$ Planned to be completed
	•Investigating and drafting the sampling plan $\Rightarrow$ Investigation of work area and
	selection of items to be collected planned to be completed



- Development process so far -

The development process aimed at on-site introduction is indicated in Fig. 1.





Fig. 1 Sampling system development process

#### Sampling system [Manufactured in FY2021]

- Outcome of element tests and reflection of issues
- Implementation of the functions and safety measures required for hot testing (Radioactive materials leakage prevention and exposure countermeasures)



## (b) Development of sampling technology Basic conditions for sampling

#### > The basic conditions for sampling are indicated in Table 1.

Table 1 Basic conditions for sampling

Items			Basic conditions
Sampling target		KURION	All adsorption towers
		SARRY	Cesium adsorption towers with a laminated flange type adsorbent filling part (simultaneous adsorption tower is outside the scope)
	Maki ope	KURION	Adsorption tower top board surface
	ng an ning	SARRY	Flange with opening for filling adsorbent
	Sampli	Amount collected	3 or 10 to 20 mL / sample *The amount to be collected is selected depending on the anticipated activity concentration of the sample
Sa		Collection depth	Up to about 100mm from the surface
mpling	Вu	Number of samples collected	1 sample in the circumferential direction x 1 sample in the depth direction (Total 1 sample / Adsorption tower)
	Closing th	Installation / removal	It should be ensured that the opening is properly sealed and that the seal can be removed even after the opening has been closed.
	e opening	Pressure resistance	Pressure resistance during operation should be ensured assuming removal of adsorbent, etc.







Fig. 2 SARRY adsorption tower top part



## (b) Development of sampling technology Components and functions of the sampling system (1/3)

- Overall composition
  - Work area is set up in an existing building that has a crane that meets the required specifications.
  - Fig. 1 illustrates an image of the overall composition of the sampling system.





## (b) Development of sampling technology Components and functions of the sampling system (2/3)

#### Table 1 Components of the sampling system and their overview

Composition	Overview
Sampling system [Integrated Sampling Module (ISM)]	Has the functionality for a series of tasks including "Making an opening", "Sampling" and "Closing the opening". Used in common for KURION and SARRY
Working dock [Working Dock (WD)]	Connection device for installing the ISM on the adsorption tower. Has the functionality to shield the radiation source at the top part of the adsorption tower.
Working stand [Working Stand (WS)]	Ensures that the adsorption tower is stably fixed, and secures a work floor in the area around the lateral shielding and sampling system.

 WS
 Fig. 1.00







Laser range finder

Endoscope

Cleaning buff

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#### Fig. 2 Dedicated tools for ISM



#### (b) Development of sampling technology Components and functions of the sampling system (3/3)

- Sampling system (ISM)
  - The shielding lid and canister are activated by means of the motor operated sliding unit, and the shielding at the access port at the bottom of the ISM and the opening / closing are controlled automatically.
  - > Packing is added to the opening/closing panel for improving air-tightness.
  - Leakage of contamination outside the system is prevented by means of the cut pieces suction recovery unit and the ventilation unit inside the system.
  - > The main body frame is made up of stainless steel. Hence it is difficult to get contaminated and even if it does, decontamination is easy.




#### (b) Development of sampling technology

#### Items from the outcome of the element tests to be reflected in the sampling system (1/4)

#### Technology for making an opening

- Functions and methods established by means of element tests (Fig. 1, Fig. 2, Fig. 3 (c))
  - Making a stepped hole of the size through which sampling can be performed, and which can be closed
  - Making a high precision opening by means of the guide mechanism
  - Integrated structure for spraying cutting lubricant and recovering cut pieces by suction
  - Drill bit structure such that the resulting cut pieces can be easily sucked out
- Rotating motor Suction recovery Coupling Hole making rod Cutting lubricant The quide mechanism Drill bit supports the outer periphery of the drill bit The hole itself supports the outer Guide mechanisr periphery of the drill bit (a) At the time of starting the process (b) At the time of drilling the hole of making a Φ60 hole Φ60 deep Fig. 1 Schematic diagram of the work of making an opening Suction port Lubrication hole

- Improvements
  - The entire structure was reviewed and as a result rods were shortened by 300 mm
  - A center run-out control function was added to the Φ40 hole making rod
  - In order to increase the efficiency of recovering cut pieces, a skirt covering the outer periphery of the Φ40 drill bit was added (Fig. 3)
  - > In order to improve strength, the shape of the tip of the  $\Phi$ 40 drill bit was improved
  - In order to review the internal structure of the WD and to improve the installation precision, the guide mechanism installation location was changed from the WD to the bottom of ISM







(a) Before measures

(b) After measures

- (c) Schematic diagram of skirt
- Fig. 3 Φ40 skirt
- Functional test (Fig. 4)
  - > All aspects were appropriately reflected and it was verified that the predetermined opening can be made.



Fine grooves in the cut piece





- (c) Data log
- Fig. 4 Status during the functional test







# (b) Development of sampling technology Items from the outcome of the element tests to be reflected in the sampling system (2/4)

# Sampling technology

Functions and methods established by means of element tests

The physical properties of the actual adsorbent obtained are measured, and the simulated adsorbent that has physical properties similar to those of the actual adsorbent is selected.

- ≻ Establishment of the procedure for collecting adsorbent (Fig. 1)
- $\triangleright$ Development of 2 types of sampling heads (SH), a small vane opening/closing type sampling head and a bottom opening/closing type sampling head, which has a dual cylindrical structure and in which the sampling port opens and closes by means of clockwise and anti-clockwise rotation (Fig. 2).
- $\triangleright$ The predetermined amount of adsorbent can be collected by running the program (Fig. 3)



Fig. 2 External appearance and structure of SH



#### Fig. 3 Results of adsorbent collection tests

- Improvements
  - The program was improved so that the sampling location gets set automatically when the value measured at the surface is entered.
  - The S-ISM sampling program was available only for lateral sampling. The option of bottom sampling was added.
  - The function of automatic insertion of the SH into the  $\triangleright$ canister after sampling was added (Fig. 4).





# (b) Development of sampling technology

Items from the outcome of the element tests to be reflected in the sampling system (3/4)

# Technology for closing the opening

- Functions and methods established by means of element tests
  - The developed closing plug can be installed remotely into the opening by running the program
  - $\geq$ The pressure resistance test is implemented so that the required pressure resistance performance is attained (Fig. 1)



Fig. 1 Pressure resistance test apparatus during the element test on closing

Improvements

RI

- In the FY2019-2020 version of the closing plug, the frictional force  $\geq$ of the seal alone was counteracting against the internal pressure. Hence the event of the plug rising to the surface occurred during the pressure resistance test. Therefore, a new closing plug was developed which is equipped with a wire net that prevents the plug from rising to the surface so that seal is not subject to excess sheer force (Fig. 2).
- $\geq$ The installation position for appropriately installing the closing plug was determined and the program pertaining to the conditions for determining proper closing was improved.





Fig. 3 Expansion of ring spring









### (b) Development of sampling technology

Items from the outcome of the element tests to be reflected in the sampling system (4/4)

# Other

- Functions and methods established by means of element tests
  - It was evaluated that the design concept of carrying out the series of operations including making an opening, sampling and closing the opening by changing the tools with the help of the vertical sliding mechanism and the rotating motor is reasonable.
  - It was confirmed that the difference in the structure of the top portion of KURION and SARRY is taken care of by the WD and that both can be handled with a single ISM.
  - It was confirmed that the safety function of automatic shutdown if a person opens the panel while any of the motors are in operation, and the emergency shutdown button functioned properly (Fig. 1).

- Improvements
  - Along with placing the guide mechanism in the ISM rather than the WD, measures were taken to ensure that the guide mechanism does not come in contact with the WD when the ISM is installed on the WD.
    - Since the ISM is suspended and lowered rectilinearly, the guide pin was installed on the WD (Fig. 2).
    - A clinometer that displays the direction in which the crane should move so that the ISM is in a straight line while the guide pin is being inserted, was developed and installed on the ISM (Fig. 3).



(a) Door open/close sensor



(b) Main body control panel emergency shutdown

Fig. 1 Safety function



Fig. 2 ISM installation with the help of the positioning pin

#### Guidance about direction in which crane should be operated



(a) Clinometer (test manufactured)



(b) Clinometer (incorporated in the main body control panel)

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Fig. 3 Crane operation guidance clinometer (test manufactured)



# (b) Development of sampling technology Items to be reflected in the sampling system for hot testing (1/3)

## Leakage prevention

The functions that the sampling system is provided with for preventing leakage of radioactive  $\geq$ substances during hot testing, based on the investigations directed towards on-site introduction during the conceptual designing, are indicated in Table 1.

Table 1 Radioactive substance leakage prevention functions (1/2)

Risk of leakage	Function		
Inflow of dust from inside the adsorption tower into the ISM	Assumption: Although the adsorption tower is stored with the hydrogen vent, etc. open, dust is not detected and excessive dust is not afloat.		
	Hole making program that makes a through hole without suction by stopping suction right before boring		
	Shielding lid with seal that opens and closes the seal insertion opening at the bottom of ISM, and its motor operated sliding mechanism		
Diffusion of dust from inside the adsorption tower to the outside	Sealing mechanism at the region of contact between the guide mechanism and the upper surface of the adsorption tower (Fig. 2), and its pushing mechanism (Fig. 3)		



# (b) Development of sampling technology Items to be reflected in the sampling system for hot testing (2/3)

#### Leakage prevention

Table 1 Radioactive substance leakage prevention functions (2/2)

Risk of leakage	Function		
Contamination of the inside of the ISM due to the collected samples	Assumption: According to the results of the element tests, there is no visible dispersion of the collected samples.		
	The flow of air is reduced by shutting down the ventilation system during the process from collecting samples to storing them in canisters.		
	The collected samples are stored in the canisters inside the ISM while they are still enclosed in the sampling head.		
Diffusion of dust from inside the ISM to the outside	The ISM door, panel, cable penetration section have seals and are air tight (Fig. 1).		
	As there are HEPA filters respectively in the air supply and exhaust systems, the air inside the ISM passes through filters even after the ventilation is shutdown (Fig. 2).		
	Dust is measured before the ISM door is opened (Fig. 3)		
	If dust is detected inside the ISM, the air is replaced by means of the ventilation unit (Fig. 3).		
	The ISM is enclosed inside a house (optional).		

filter





(b) Cable port

Fig. 1 Improved air-tightness





#### (b) Development of sampling technology Items to be reflected in the sampling system for hot testing (3/3)

# Reduction of radiation exposure

Functions provided for reducing exposure during hot testing are indicated in Table 1.

Table 1 Exposure risk reduction function

Exposure risk	Function		
Exposure during preparations and tidying up	Installation is carried out remotely using a crane.		
Exposure from the upper surface of	Shielded by means of the WD that is filled with lead particles (Fig. 1).		
the adsorption tower	A remote control panel separated from the ISM main body is provided in order to minimize the time for which one stays on the WS scaffolding.		
Exposure from the sides of the	Shielded by suspending and lowering a lead shielding mat on the WS.		
adsorption tower	Remote control panel is installed at a distance from the adsorption tower.		
Even over the beam abarred	Device is remotely operated so that people can keep a distance.		
radiation from the penetration section	When the tool rises up back to its original position, the shielding lid automatically closes the penetration section by means of the motor operated slide (Fig. 2).		
	The process from sampling to inserting the sampling head (hereinafter, SH) into the canister is carried out automatically (Fig. 3).		
Exposure around the collected	The area around the SH is shielded by the canister and the canister shield.		
samples	The SH and the rod are separated using a dedicated long reach spanner through the glove port and hence the hands can be kept away.		







Fig. 3 Automatic insertion of SH into the canister



# (b) Development of sampling technology Specifications of the sampling system (1/2)

The points in which the results of the element tests were reflected and the points in which items investigated for hot testing were reflected while developing the sampling system are indicated in Table 1.

Table 1 The points reflected from the results of the element tests and the points reflected from the items investigated for hot testing

Procedure	Reflection of element test results	Reflection of investigations conducted aiming for hot testing
1. Preparati on	Enhancement of the ease of remote installation of ISM with the help of the positioning pin and clinometer	<ul> <li>Exposure reduction through addition of shielding around the adsorption tower</li> <li>Exposure reduction by carrying out work remotely</li> <li>Prevention of the flow of dust from inside the adsorption tower into the ISM and shielding of the beam shaped radiation from the through hole by means of the automatic sliding of the shielding lid</li> <li>Prevention of diffusion of dust from inside the adsorption tower to the outside and shielding of radiation by means of the air-tight structure of the ISM itself and the seal under the</li> </ul>
2. Making an opening	<ul> <li>Reliance on the process of making a stepped hole for putting in the closing plug</li> <li>Prevention of center run-out by means of the guide mechanism and by shortening the rod</li> <li>Continuation of spraying of cutting lubricant and of the cut pieces suction recovery function</li> <li>Addition of a skirt to the Φ40 hole making rod</li> <li>Reliance on the design of the end milling drill bit and its strength enhancement</li> </ul>	
3. Sampling	<ul> <li>Setting of sampling procedure in which the internal verification is carried out first</li> <li>Reliance on the SH sampling port opening/closing mechanism that has a dual cylindrical structure</li> <li>Setting of separate procedures for lateral sampling and bottom sampling according to the adsorbent</li> <li>Provision of automatic sampling program corresponding to the lateral sampling SH and bottom sampling SH respectively</li> </ul>	<ul> <li>guide mechanism</li> <li>Prevention of contamination of the inside of the ISM through automatic insertion of the collected samples into the canister</li> </ul>
4. Closing the opening	<ul> <li>Reliance on the basic design for remotely closing the opening by means of the extendable closing plug</li> <li>Securing of stable pressure resistance by reviewing the structure</li> <li>Provision of the function to determine the position and fastening state by improving the hole closing program</li> </ul>	



# (b) Development of sampling technology Specifications of the sampling system (2/2)

The main specifications of the sampling system are given in Table 1, and photos taken when the functional tests were conducted by the manufacturer are shown in Fig. 1.

Table 1 Basic specifications of sampling system

Items		Specifications	
External dimensions		L1.420xW1, 206xH3,604 mm	
Mass		Approx. 1,600kg	
Powe	r supply	3Ф200V, 50Hz, 50A	
Vertical drive	Drive system	Ball screw vertical drive	
	Motor specifications	Output / torque: 0.75kW / 20Nm Vertical speed: up to 25 mm/s	
	Stroke	1,900mm Sampling depth equivalent to maximum 200 mm* *SARRY standards	
Rotational drive	Motor specifications	Output / torque: 5kW / 300Nm Rotational speed: up to 450 rpm	
Making an opening		Hole making rod, drill bit (Ø60, 50, 40mm) Cut pieces suction function Cutting lubricant spraying function	
Sampling		Endoscope rod, laser range finder, endoscope sampling rod, sampling head	
Closing the opening		Perforated part cleaning rod, cleaning buff Closing plug rod, extendable closing plug	
Data logger		Recording and verification of the status of movement Recording of the motor load	



Fig. 1 External appearance of the sampling system during the functional test



# (b) Development of sampling technology

Sampling plan for verification of sampling technology

# Objective

To plan sampling from actual adsorption towers, etc. for verifying the sampling technology that uses the sampling system.

# Overview

Sampling tests (cold testing and hot testing) were conducted using actual adsorption tower through which contaminated water has not passed (cold) and a used adsorption tower (hot) for verifying the sampling technology.

The following items were investigated for conducting cold and hot testing.

- Selection of the target adsorption tower
- Selection of the sampling work area
- Details of work procedures
- Handling of collected samples
- Calculation of time required for sample collection
- Investigation of overall schedule
- Estimation of total exposure dose



(b) Development of sampling technology Selection of the target adsorption tower (1/3)

- Adsorbent to be sampled from KURION
  - KURION system configuration is shown in Fig. 1. (Number of systems and number of adsorption towers have been reduced)



Fig. 1 KURION system configuration

- Silica sand has been used as the adsorbent in SMZ skid on an ongoing basis except in the initial stage.
- H and EH have similar properties. Note that, after the H and EH adsorption tower is used in the AGH skid, it is moved to the H skid and used.
- TSG, which is granular titanosilicate, is a mineral and its properties are likely to be different than other adsorbents.
- It appears that the AGH adsorbent used for I-131 has adsorbed I-129 which is an important nuclide in treatment and disposal. Hence the facts pertaining to its collection need to be ascertained.
  - Silica sand, H, TSG and AGH are sampled from KURION during the sampling technology verification test.



# (b) Development of sampling technology Selection of the target adsorption tower (2/3)

- Types of SARRY adsorption towers and sampling target
  - There are various types of SARRY adsorption towers as indicated in Fig. 1 based on the manufacturer, date of manufacturing, and application.
  - In this research, Cs adsorption towers pertaining to which information has been obtained, are investigated.
  - The lead particle type adsorption towers are excluded as there are uncertainties such as sticking, deformation, etc. of lead due to long term store Classification based on the shielding



- Fig. 1 Types of SARRY adsorption towers
- The SARRY adsorption towers from which sampling is carried out during the verification test is selected from the flange type adsorption towers.



(b) Development of sampling technology Selection of the target adsorption tower (3/3)

- Adsorbent to be sampled from SARRY
  - SARRY system configuration during the period when flange type adsorption towers were used, is shown in Fig. 1.

(Number of systems and number of adsorption towers have been reduced)



Fig. 1 SARRY system configuration when flange type adsorption towers were used

• There are 4 types of adsorbents used in the SARRY flange type adsorption towers, namely, coarse sand, fine sand, IE96, and IE911.

(The shape of the media filter is different than those used in other SARRY adsorption towers. Media filters did not have any track record as far as it was checked. Hence these filters have been excluded from the very beginning.)

 A maximum of 4 adsorption towers from those containing coarse sand, fine sand, IE96 and IE911 are sampled from SARRY.



# (b) Development of sampling technology Selection of the sampling work area (1/2)

- Conditions for selecting the sampling work area
  - Availability of the required lifting equipment
    - Maximum crane load: Should be able to lift the KURION and SARRY adsorption towers.
    - Crane lifting height: KURION 7.9 m or more, SARRY 8.0 m or more
  - The impact of risks of natural disasters such as typhoon, etc. should be sufficiently small.
- Contents of investigations
  - The following work locations have been selected based on the condition of availability of required lifting equipment.
    - Working Machine Building (Indoor work)
    - High Performance ALPS Building (Indoor work)
    - Second temporary storage facility (Outdoor work)
  - Due to the condition that the impact of risks of natural disasters such as typhoon, etc. should be sufficiently small, the second temporary storage facility where work would be carried out outdoors was excluded.
  - Considering the efficiency of on-site work, the Working Machine Building was excluded.
- Investigation results
  - It was evaluated that the High Performance ALPS Building would be the most suitable as the sampling work area.
  - Dedicated working stands are built inside the building and the sampling technology is verified for adsorption towers of both KURION and SARRY.



# (b) Development of sampling technology Selection of the sampling work area (2/2)

- Building the work area in the High Performance ALPS Building
- WS and maintenance stand (hereinafter, MS) are installed at the back side of the truck bay entrance.
- > A generator and compressor are installed outdoors.
- After installation from adsorption tower to ISM, the ventilation unit is connected to the ISM.
- Thereafter, a house is installed on the WS.



Fig. 1 Image illustrating construction of the work area in the High Performance ALPS Building

- Facilities related to sampling work and details of implementation
- The layout diagram of the main facilities related to sampling work is shown in Fig. 2.
- The adsorption towers are temporarily placed in the second temporary storage facility, or are temporarily stored in the temporary storage facility.
- In the following circumstances, the required measures are taken in advance at the second temporary storage facility.
  - If the adsorption towers need to be decontaminated or covered.
  - If the adsorption towers need to be rotated 90° so that they face in the direction in which WS has been installed.
- While sampling, the adsorption towers are transported to the High Performance ALPS Building one at a time.
- Collected samples are transported on-site to the solid radioactive waste storage facility.
- After sampling the adsorption towers are planned to be returned to the original storage facility.
- > On-site investigation of the second temporary storage facility is carried out.



Fig. 2 The layout of the main facilities related to sampling work



# (b) Development of sampling technology Details of work procedures (1/2)

### Main work procedures when the sampling system is used



#### IRID

# (b) Development of sampling technology Details of work procedures (2/2)

#### Main work procedures when the sampling system is used





# (b) Development of sampling technology Handling of collected samples

- An image of the canister in which the sampling head (hereinafter, SH) is placed, is shown in Fig. 1.
- The flow from collecting the samples to analysis is shown in Fig. 2.



Fig. 2 The flow from collecting the samples to analysis (Image)



# (b) Development of sampling technology Calculation of the time required for sample collection

Table 1 Work procedures and expected number of days

Nie	Werkeente	Expected number of days		
NO.	work contents	KURION	SARRY	Sub-total
1	Preparation for sampling			1 day
2	Adsorption tower transportation and installation			1 day
3	Series of tasks using the sampling system	Total 244 minutes	Total 677 minutes	
	• Measurement of the distance of the surface where the opening is to be made (adsorption tower top board location)	1 minutes		
	<ul> <li>Φ60 deep hole drilling</li> </ul>	54 minutes	424 minutes	
	<ul> <li>Measurement of the depth of the Φ60 perforated hole and checking of the inner surface of the perforated hole</li> </ul>	19 mi	nutes	
	• Φ50 through hole	44 minutes		
	<ul> <li>Measurement of the distance up to the welded bottom and checking of the inner surface of the perforated hole</li> </ul>	-	19 minutes	1 day * *The functioning of the system can be arbitrarily
	• Φ40 through hole	-	44 minutes	discontinued and resumed, but the series of tasks
	<ul> <li>Measurement of the distance up to the surface of the adsorbent and checking of the inside of the adsorption tower</li> </ul>	19 mi	nutes	in 3 shifts so that the adsorption towers are not left unattended at any time without closing their openings.
	Sampling, insertion of the sampling head into the canister	11 minutes		
	Measurement of the radioactive dust concentration inside ISM	30 minutes		
	Removal of the canister containing the sampling head	3 minutes		
	Cleaning of the perforated hole and checking of the inner surface of the perforated hole      41 minutes			
	Placement of the closing plug	6 minutes		
	Checking of the status of placement of the closing plug	16 minutes		
4	Sampling system removal / tidying up			1 day
(5)	Adsorption tower transfer	1 day		1 day
6	Sampling system maintenance / decontamination / transportation of samples	1 day		
$\overline{\mathcal{O}}$	Extra days 1 day		1 day	

• Collecting samples from 1 adsorption tower requires 1 day only for the series of tasks and 7 days including preparation and tidying up.



# (b) Development of sampling technology Investigation of overall schedule

- Reflection of work procedures and expected number of days
  - According to the results of the element tests and the investigations on work procedures, it was calculated that the series of tasks using the sampling system would require 224 minutes (approx. 5 hours) in the case of KURION, and 677 minutes (12 hours) in the case of SARRY.
  - However, the plan is to complete the series of tasks within the same day so that work is not discontinued while the opening in the adsorption tower is yet to be closed.
  - It was calculated that it would take 7 days to collect samples from 1 adsorption tower considering transportation of the adsorption tower and installation of the sampling system.
- Implementation items
  - Along with arranging for the materials and equipment, equipment testing and trainings for the personnel involved in testing are conducted at the in-house facilities.
  - Before collecting hot samples, the functioning of the equipment is verified and training is conducted by carrying out cold testing using adsorption towers through which contaminated water has not passed.
- Preconditions of the overall schedule
  - It is assumed that the time and work area required for testing is available.
  - It is assumed that there would be no additional regulatory requirements besides those that are anticipated at present.
  - It is assumed that sampling is carried out from a maximum of 8 adsorption towers. Although it takes 7 days to collect samples from 1 adsorption tower, an evaluation period is set between every 2 collections.
  - The proposed overall schedule for collecting samples from 4 adsorption towers each from KURION and SARRY, is shown in Table 1.



#### Table 1 Proposed overall schedule



# (b) Development of sampling technology Estimation of total exposure dose (1/3)

1. Method of calculating the total exposure dose



-ig. 1	Sample collection work location
(High	performance ALPS Building)

- The number of workers assigned and the duration of work is set based on the investigations on schedule and work steps, and the results of the element tests
- The radiation source, etc. that is likely to affect work exposure is set, and the atmosphere dose rate is calculated according to the work steps (Table 1, Table 1 on next page)
- The work classification is set depending on the contents of the target work (Table 2), and accordingly the work coefficient is set
- The exposure dose in each work step is calculated with the following formula and aggregated

#### Work exposure dose (person/mSv)

- = Work coefficient
  - x Number of workers (persons)
  - x Duration of work (h)
  - x Atmosphere dose rate (mSv/h)

Table 1 Target dose rate and calculation method	b
-------------------------------------------------	---

Dose contribution	Target dose rate	Calculation method	
Impact of work area	BG value of work building	Set based on survey data	
Impact of adsorption tower	Around the adsorption tower while preparing for work		
	Around the adsorption tower after removing the shielding lid and while installing WD (Only KURION)		
	Around the adsorption tower after installing WD	Calculated using the computing code	
	Around the adsorption tower while installing the sampling system (ISM)	QAD-000F2N	
Impact of collected samples	Surface of canister and the area in its vicinity		
	Area in the vicinity of sampling head		

#### Table 2 Work classification

Work classification	Target Work	
Low	Work that requires multitude of trips outside the work area (adsorption tower transportation, preparatory work, etc.) Work during which it is possible to retreat to a low radiation area (dust measurement, etc.)	
Medium	Work that requires constant presence in the work area Short duration work while operating the sampling system (ISM)	
High	Work of manually operating the sampling system (ISM) Work for which the time set is as short as several minutes and hence is considered as a margin	



## (b) Development of sampling technology Estimation of total exposure dose (2/3)

#### 2. Estimation of atmosphere dose rate

• The work steps for which the atmosphere dose rate is calculated are indicated in Table 1, and an example of the dose rate distribution based on the results of analysis is shown in Fig. 1.

Table 1 Work steps for which the atmosphere dose rate will be calculated

Case	Work step	
1	Inside the High Performance ALPS Building	
2	Around the adsorption tower	
3	While installing WD	
4	While installing ISM	
5	Work on the working stand such as tool change, etc.	
6	Inside ISM	
7	While inserting sampling head into canister	
8	Canister handling inside ISM	
9	Removing the canister	
10	Before transporting canister	



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# (b) Development of sampling technology Estimation of total exposure dose (3/3)

#### Estimation of total exposure dose and evaluation of feasibility of work

- > Setting the allowable total exposure dose
- The number of work teams and the number of members in each team are set based on the work procedures and the content of work
- Considering that the work is assigned in rotation, it is assumed that all workers are uniformly exposed.
- Assuming that the workers are involved in other tasks as well during the year, the upper exposure limit per person for this task is set.
- ⇒ The allowable total exposure dose for this task is set based on the upper exposure limit per person and the total number of workers.
- Calculating the total exposure dose evaluation value
- The radiation for evaluating the exposure is set considering the radiation from the adsorption tower mentioned in the implementation plan and the attenuation due to time elapsed up to the day on which exposure is evaluated.
- It is assumed that the 4 adsorption towers from SARRY and KURION have the same radiation.
- ⇒ The total exposure dose for this task which includes preparatory work, sampling from each adsorption tower and the work of tidying up, is evaluated.

Evaluation results
 The following results were obtained.

#### <u>Allowable exposure dose > Total exposure dose evaluation value</u>

It was verified that the task is feasible on site.

- Issues
  - Re-evaluation will be carried out based on the latest dose rate of the adsorption tower from which samples are collected, in order to improve accuracy.
  - Since working on the KURION adsorption towers involves high level of exposure, enhancement of shielding performance will continue to be investigated.



# (b) Development of sampling technology Summary

### Current results

- The sampling system was designed and manufactured based on the results of investigations directed towards hot testing and results of the element tests during the FY2019-2020 research, and functional tests were completed.
- The incidental facilities required for the series of tests using the sampling system were designed and manufactured.
- Site investigation of the facilities was carried out in relation to verifying sampling technology, and the required peripheral equipment such as ventilation unit, etc. were investigated and designed.
- Considering the designed peripheral equipment, the sampling plan for verifying the sampling technology was drafted.
- The dose around the adsorption towers under certain radiation conditions was evaluated, and the total exposure dose was estimated.

#### Issues, etc.

- Manufacturing and functional verification of peripheral equipment such as ventilation unit, etc. required for verifying the sampling technology
- Verification of the layout of cameras, etc. in accordance with the on-site environment for ensuring ease of remote operation
- Investigation of the arrangement of the dosimeter for monitoring changes in air dose rate and of the methods of evaluating the readings
- > Dose evaluation considering the conditions of the actual adsorbent to be sampled as the model conditions
- Consideration of additional shielding, etc. based on the results of dose evaluation for further reducing exposure



2. Project details (No. 4)

d. Integration of R&D results





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- Achievements up to FY2020
- By FY2020, a system was developed for comprehensively managing and evaluating the direction of research and development related to treatment and disposal of solid waste, its progress, results and pending issues (Fig. 1).
- Goal
- To put forth view points for selecting waste streams when multiple options are left to choose from, and to provide the indicators for making a decision.
- To comprehensively summarize the research results including results from previous years.
- Details of implementation (Details are provided from next page onwards)
- Investigation of view points for narrowing down waste streams when multiple streams are left to choose from, based on the waste stream investigated up to FY2020, and of the indicators for making a decision.
- Conducting of case studies using specific wastes as examples, with respect to the viewpoints for narrowing down the investigated waste stream options and the indicators for making a decision, and consolidation of issues such as too much or too little information that is required while selecting the option.
- Indicators for the achievements of goal
- The viewpoints for narrowing down the options and the indicators for making a decision are provided.
- Case studies related to narrowing down options with respect to representative wastes are conducted, and issues in narrowing down the options are consolidated (Example: Information required for selection, etc.).

(Target at the completion: TRL:  $4 \rightarrow 4$ )



Fig. 1 Details of investigations conducted up to FY2020 and their output

(Flow of integration of research & development results)



Role of "Integration of R&D results" and its correspondence to the contents of investigations

Purpose of investigating "Integration of R&D results"

To consolidate the flow (= waste stream) from generation of waste to storage, treatment such as segregation, volume reduction, stabilization, etc. and then long term storage or disposal based on the uncertain character of information (limited information) related to the properties of solid waste generated in 1F.

And in doing so, to carry out research & development rationally and efficiently by getting an overall bird's-eye view, and in addition by mutually providing information during every stage of the waste stream as feedback.



Matters to be investigated aiming at "Integration of R&D results" and the output goals

No.	Items to be investigated	Output goals (Example)
1	Presentation of the direction of the research & development directed towards treatment and disposal of waste and the goals	Provision of a mechanism (example: waste stream) for sharing with the concerned parties, who finally utilize the research & development results.
2	Provision of a mechanism for investigating the sufficiency of the R&D topics concerning treatment and disposal of waste	Provision of a mechanism (example: input consolidation chart) for sharing and making visible the domains of research & development at each stage to the concerned parties.
3	Provision of a mechanism for sharing and making holistically visible the status of progress of the research & development topics	Provision of a mechanism (example: temporal axis organization chart) for sharing and making visible the progress of research & development and the pending issues to the concerned parties.
4	Provision of items that need to be considered while selecting the treatment and disposal options	Consolidation of information for making decisions (perspective for narrowing down the options and the indicators for making decisions) when multiple waste stream options are left to choose from



#### (d) Integration of R&D results Overview of the investigations





#### (d) Integration of R&D results Contents of investigations and procedures

Investigations were conducted in accordance with the following procedures directed towards "Research Goal 1 of the research: To put forth view points for selecting waste streams when multiple options are left to choose from, and to provide the indicators for making a decision" and "Research Goal 2 of the research: To comprehensively summarize the research results"



development, are investigated.

# (d) Integration of R&D results Contents of investigations: ① Investigation of the indicators for narrowing down

[Details of examination]

# options (1/3)

>The indicators for selection and narrowing down when multiple waste stream options are left to choose from were investigated from the viewpoint of "safe", "reliable", "rational", "quick", "field oriented", while referring to the Five Guiding Principles for reducing risks indicated in the "Technical Strategic Plan^[1]".

From the viewpoint of "safe", the IAEA safety standards "Management of radioactive waste before disposal" (GSR-Part 5) and related separate safety requirements were investigated, and were reflected while setting the indicators.
 With regards to "reliable", "rational", "quick" and "field oriented", the indicators of each viewpoint were investigated while referring to the "Technical Strategic Plan" and the results of brainstorming implemented while investigating options for treatment and disposal of waste ^[2].





#### (d) Integration of R&D results Contents of investigations: ① Investigation of the indicators for narrowing down options (2/3)

[Investigation results: Output]

> The "indicators for narrowing down", "determining factors", "information required while narrowing down", and "proposed methods for narrowing down" were consolidated with respect to 5 viewpoints (Safe, reliable, rational, quick, field-oriented).





Contents of investigations: ① Investigation of the indicators for narrowing down options (3/3)

[Investigation of the methods for leveraging the Investigation results]



# Contents of investigations: 2 Investigation of information required for the task of narrowing down (1/2)

>The source from where information required for narrowing down is acquired is classified into the following 3 categories based on the results of Contents of investigations ①.

- From the object to be treated (waste): Inventory, quantity, properties
- From the treatment technology and method: Treatment speed, TRL, etc.
- From the storage and disposal facilities / plans: Storage capacity, acceptance criteria, etc.

>Assuming that efforts will be made to further enrich information in accordance with the stage such as research & development progress, etc., the processes concerning "objects to be treated (waste)", "treatment technology and method", and "storage and disposal facilities/plans" are set.

>Based on the set processes, the stages at which information required for narrowing down options can be obtained are investigated and consolidated.



are classified into "objects to be treated (waste)", "treatment technology and methods", and "storage and disposal facilities/plans" *In reality, processes are likely to be implemented in parallel or are likely to be iterated, but the purpose here is to indicate the stage at which information required for narrowing down options can be acquired. Hence the processes indicated here have been simplified.



# Contents of investigations: 2 Investigation of information required for the task of narrowing down (2/2)

The required Information, the source of information, and the stage at which the information can be obtained are investigated and consolidated in accordance with the 5 viewpoints and narrowing down indicators consolidated in Contents of investigations ①.



: Information that needs to be collected from objects to be treated (waste)

: Information that needs to be collected form the treatment technology/methods and the treatment plan

: Information that needs to be collected form the storage/disposal facilities and related work plans





#### (d) Integration of R&D results Contents of investigations: ③ Narrowing down on a trial basis using the [Contents of investigations] representative waste (Case study) (1/3)

>Specific wastes and options (prospective applicable technologies) are narrowed down on a trial basis based on Contents of investigations ① and Contents of investigations ②,

Note: As limited information can be provided at the current point in time, <u>illustrating the task of narrowing down options with</u> <u>examples</u> and <u>identifying issues in the viewpoints for selecting and narrowing down options</u> are considered as the purpose of the case study rather than trying to determine the superiority/inferiority of the options.

[Object of the investigations]

Target waste: ALPS slurry*

(Reason for selection)

•Waste that has been selected as a representative waste for investigations conducted until last year as well, and the results of investigations conducted during the subsidized projects can be leveraged.

•Waste for which treatment and disposal work is planned to be implemented on a priority basis in the solid waste storage management plan^[1].

•Waste for which multiple waste stream options are available in the waste conditioning treatment process

*For carbonate slurry and iron co-precipitation slurry to which application of low temperature solidification treatment is being considered.

•Target process and options: Waste conditioning treatment process / low temperature solidification treatment (cement solidification/AAM solidification)

(Reason for selection)

• Even though options have been narrowed down till the storage management process based on the dehydration treatment, multiple treatment technologies remain as options during the waste conditioning process.



#### <Status of investigating the ALPS streams>

Contents of investigations: ③ Narrowing down on a trial basis using the representative waste (Case

[Investigation results]

study) (2/3)

>Information that can be gathered at the current point in time on representative waste was provided leveraging Contents of investigations (2).

>Using this information, the approach towards evaluating the narrowing down indicators for which the method for comparative evaluation of the options was not clear, from among those set in Contents of investigations ①, was illustrated with examples.

No. 絞り込み時に必要とする情報	関連する評価指標	情報の入手元	情報が取得可能となる段階(赤枠内)		in time	e was carried out, or indicators that can be considered w	vere set.
1 処理対象物のインペントリデータ	Al	処理対象物	<u>発生</u> → 性材理 → 処理/炭型体化 → 保證/期2		Indicator A2: Controlling amount generated		
2 処理対象物の緑量データ	A1	処理対象物	死生 + 性状記録 → 処型/疾薬体化 → 保管/期段			cator A4: Form that has good confinement performance	(stable/difficult t
3 作業計画線量	A1	処理技術・方法・作業計画		307BRNS	aet dif	fused)	
4 処理作業の内容(作業中における飛歌リスク)	(作業中における限制リスク) AL EL 処期技術・方法・作業計画 要素は56月7日 - E2時代1時の対 適用活用の定 通 E2時代1時、 第月時の日		適用開始	Indicator B4: Elexibility (Scope of application of technology)			
環後の状態(後段プロセスへ引渡す際の鉱款リスク) A1 処理技術・		処理技術・方法・作業計画	技術・方法・作業計画 要素技術研究 → 120m/1484対 → 初用計画設定 → 120m/361+38/作 → 39		Bearing in mind the use of the evaluation indicators that have been applied for 1F the		
6 処理対象物の物量	A2	処理対象物		TUTANA	appro	ach of quantifying the qualitative indicators was investigated a	nd illustrated with
7 Q項により完全する初量(減裕本寺に基づく評価値) (協会になった)もの量(減裕本寺に基づく評価値) (協会になった)もの量(協会になった。	A2	处理技術·方法·作業計画	SCRUCKINGAL AND ADDREED ADDREE	2000000	exam	nles	
	A3, B2	保管、環辺施設、保管/環設計画		THE CONTRACT OF	Fxam	ble: Quantification of "physical stability"	
Results of sorting an gathered for the putter temper	nd investi urpose of rature so	gating the in f evaluating lidification to	nformation that can be the ALPS slurry low reatment	1990) 1990) 1990) 1990) 1990) 1990)	perspe ≻The (Exam applic	applicable Form Factor was set and compared based on the p ple: uniform solidified substances / brittle solidified substance ation of the selected option (treatment technology) Form Factor indicator ^{*2, Note}	properties of matters) resulting from the
				9 <b>7</b> 9784	#	Form	FF
Grav batching: Items pertaining to which it is difficult to provide					1	Gas, liquid, sludge containing lots of moisture* and condensed particles*	1
information at this point in time			研開始	2	Other sludge	1/10=0.1	
21 保管				FERRIC	3	Powder and loose contaminants (surface contamination, etc.)*	1/10=0.1
22 処理				研開始	4	Adherent* or penetrating contaminants (surface penetration contamination)*	1/100=0.01
<ol> <li>23 処理技術の週用までに掛かる工程情報(開発期間)設備設計・製作期間)</li> <li>24 適用決断の適用までに増える情報</li> </ol>	D2	処理技術・方法・作業計画		·通用用始	5	Brittle, easy to break solids (very porous MCCI)*	1/10,000=1E-4
25 約2時代の第4年4月19日 10月1日     25 約2時代の第4年4月19日 (10月1日日日日日日日日日日日日日日日日日日日日日日日日日日日日日日日日日日日	E1	A. HELE HALL AND A HELE HELE HELE HELE HELE HELE HELE HE	EVER-SHIPPINE EVERTSERVEY 20101118322 ZOBIERT 2011 STRESHERR STRESHERR SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERSER SOBIELERS	IDHINK IDHINK	6	Discontinuous solids (size and weight such that manual transportation is possible, such as pellets, etc.)	1/100,000=1E-5
			· · · · · · · · · · · · · · · · · · ·		7	Continuous solids	1/1.000.000=1E-6

*Form that was added to the NDA definition in order to increase its relevance to the Fukushima Daiichi Nuclear Power Station

Based on the information provided, comparative evaluation at the current point

*1: Safety and Environmental Detriment

*2: Cited from the FY2021 Technical Strategic Plan for Decommissioning of the Fukushima Daiichi Nuclear Power Station of Tokyo Electric Power Company Holdings, Inc. (October 29, 2021, Nuclear Damage Compensation and Decommissioning Facilitation Corporation)

Note) The SED evaluation indicators (Form Factor, etc.) are not necessarily set in stone, but can be modified so as to make the definition more specific, etc. in accordance with the items to be evaluated, just like the UK NDA definitions were modified to increase their relevance to 1F.

Information related to the subsequent stages (example:							
acceptance criteria), and information related to evaluation based							
on the equipment designing and operation plan (example:							
treatment cost) is difficult to gather.							
## (d) Integration of R&D results

### Contents of investigations: ③ Narrowing down on a trial basis using the representative waste (Case study) (3/3)

[Investigation results]

>ALPS slurry / Low temperature solidification treatment technology (Cement solidification / AAM solidification) were subject to comparative evaluation based on the information that can be currently gathered and on the results of investigations on the narrowing down indicators and proposed methods.

Note: Since the results are based on information that can currently be gathered, the following results do not indicate the superiority or inferiority of the options, but indicate the information that is insufficient for selection and narrowing down.

Viewpoints for narrowing down options	Narrowing down indicators	Determining factor		Example case study (ALPS slurry)	
				Cement solidification	AAM solidification
		Impact on the site boundary dose			
Safe	A1. Radiation protection	Worker exposure impact during treatment, etc.		(Same)	
		Preventing spread of contamination			
	A2: Controlling amount generated	Amount when the selected treatment is used (storage	e and disposal capacity)	0	
		Amount of secondary waste when the selected treatment is used		(Difficult to evaluate at this point in time)	
		Applicability to the acceptance capacity in subsequent processes (storage and disposal, etc.) (Difficult to evaluate at this po		this point in time)	
	A3. Acquisition of information on characteristics that contributes to determining acceptance in subsequent processes	Conformance with the acceptance criteria of the storage facility (Example: Surface dose rate, etc.)		(Difficult to evaluate at this point in time)	
		Conformance with the acceptance criteria (WAC) of the disposal facility		(Difficult to evaluate at this point in time)	
	A4: Form that has good confinement performance ( = stable/difficult to get diffused)	Stability of the waste during treatment when the selected treatment is used (Presence of significant change in state)		- (Same)	
		Properties of waste in the state after the selected treat	atment is used (Physical properties)	O (FF:1E-6)	(FF:1E-4)
		Properties of waste in the state after the selected tree	atment is used (Chemical properties)	O (CF:1,000)	(CF:100)
	A5. Measures for maintaining good confinement performance	Form in which waste is transferred to the subsequent processes (Boundary structure)		(Difficult to evaluate at this point in time)	
		Performance of the boundary structure		(Difficult to evaluate at this point in time)	
	A6. Form suitable for the work in subsequent processes (Considering the requirements of the work carried out during subsequent processes such as inspection, removal, etc.)	Form in which waste is transferred to the subsequent processes (Ease of handling)		(Difficult to evaluate at this point in time)	
		Consideration of the requirements of subsequent processes (Example: Inspection, monitoring, etc.)		(Difficult to evaluate at this point in time)	
Reliable	B1. Technology Readiness Level of the	Application track record			
	(Applicability of technology)	Progress of research & development		-(Same)	′
	B2. Presence of clear judgment criteria (≒ Determining suitability to the requirements)	Quantitative criteria for determining the suitability of treatment results		(Difficult to evaluate at	this point in time)
	B3. Flexibility (Extent to which changes in the properties resulting from target treatment can be handled)	Conditions for accepting the technology to be used (Scope of applicability)			0
		Cost for developing the technology to be applied		(Difficult to evaluate at this point in time)	
	C1. Costs involved in using the option	Cost of equipment used for the technology to be applied		(Difficult to evaluate at this point in time)	
Rational		Running costs when the technology is applied		(Difficult to evaluate at this point in time)	
		Cost impact on the subsequent processes when the applicable technology is applied		(Difficult to evaluate at this point in time)	
	C2. Versatility of the option	Scope of application of the selected technology (Waste to which the technology can be applied)		(Difficult to evaluate at	this point in time)
Quick	D1. Time used for obtaining understanding with respect to application of the option (Social acceptability)	Track record of application of the technology		(Difficult to evaluate at	this point in time)
		Explainability related to safety (at each stage of trea	Results of case	e studies	point in time)
	D2. Time required for applying the technology	Time required for developing the technology			point in time)
		Time required until on-site application including equ	related to com	parative	point in time)
	D3. Time required from application until goal achievement (treatment completion, etc.)	Treatment speed of the technology to be applied	evaluation of	options	point in time)
		Securing of work area and site		(Difficult to evaluate at	this point in time)
Field oriented	E1.1 Feasibility in the site environment	Work efficiency (Environment accessibility ease of	operation)	(Difficult to evaluate at	this point in time)

: Items that were rated to be highly relevant to the indicators during the evaluation using information that can be currently gathered. : Items that were excluded from the evaluation as only limited information can be currently gathered or it

is difficult to gather any information.

#### <Consideration from the perspective of providing information used for comparative evaluation>

#### Narrowing down from the viewpoint of "Safe"

There is a wide variety of sources of information (from waste / from treatment technology / from storage and disposal facilities). Information needs to be provided based on the following investigations in particular for enhancing the evaluation.

- · Ascertaining the characteristics of treatment based on the conditions set for the actual treatment (container specifications, mixing or filling & solidification, etc.)
- Design conditions of the storage and disposal facilities (acceptance criteria, capacity, etc.)

#### Narrowing down from the viewpoint of "Reliable"

Mainly, information related to the performance of treatment technologies is required.

The acceptance criteria for the subsequent processes (storage) and disposal) needs to be set for evaluating the applicability of the final technological capabilities.

#### Narrowing down from the viewpoint of "Rational", "Quick", "Field oriented"

Information from the treatment technology/storage and disposal facilities is required. And, for acquiring the required information, actual designing (example: designing the treatment equipment, designing the storage and disposal facilities) based on research & development needs to be implemented.



The level of priority of providing information required for the work of narrowing down from the viewpoint of "Safe" and "Reliable" is evaluated to be high considering the conditions under which the necessary information is acquired (stage of investigation).





## (d) Integration of R&D results

Contents of investigations: ④ Investigation of procedures for integration of R&D results (1/3)

[Contents of investigations]

>The following points are investigated along with re-consolidating the roles and applications of the main outputs related to the integration of research results.

- (1) Correlation of the outputs during the entire process while integrating the research results
- (2) The flow of utilization of the outputs while integrating research results

		Output	Overview/Applications	
The section in which investigatio ns were conducted in previous years		Waste management sheet	Provides information on each waste from various sources, status of investigations at various stages from temporary storage to disposal/reuse, and research results.	
		Stream (Treatment flow)	Shows the flow from generation to disposal/reuse of each waste including the options available at each s Uses for indicating the direction and goals of research & development	
		Input consolidation chart Consolidates input from the previous processes and input (requirements) of the subsequent processes the main contents of investigations at each stage (generation to treatment/reuse) of the waste stream		
		Temporal axis organization chart	Makes the progress of research & development related to solid waste visible by consolidating the status of investigations (whether or not investigations need to be conducted and their status) at each stage from generation to treatment/reuse of each waste.	
		Dashboard	Summarizes information so that the status of progress of research & development related to solid waste generated at 1F (① Properties of solid waste, ② Direction of management based on the properties (goals), ③ Issues in achieving the goals) can be shared with concerned persons.	
The section in which investigatio ns will be conducted in FY2021	List of viewpoints and narrowing down indicators for narrowing down options		Indicates examples of viewpoints, indicators and narrowing down methods to be considered while selecting and narrowing down options available at each stage of the waste streams.	
		List of information required for narrowing down options	Indicates the information that is believed to be necessary for the task of selecting and narrowing down options including the source of information and the phase at which the information can be obtained.	
			*Difference with the input consolidation chart Along with indicating information focused on while selecting and narrowing down options, the following points are added which are not expressed in the input consolidation chart. •Linking the viewpoints for narrowing down options/narrowing down indicators and the required information •Reinforcing the information from the temporal axis related to the treatment technology development process and the process of investigating storage and disposal facilities during which information can be obtained	





## (d) Integration of R&D results

Contents of investigations: ④ Investigation of procedures for integration of R&D results (2/3) [Investigation results: (1) Correlation of the outputs during the entire process while integrating the research results]



Setting the entire process involved in integration of R&D results till the stage when it helps in establishing the issues to be addressed, by indicating the waste streams and research & development status at a specific point in time, and consolidating the correlation with the phase in which the outputs are utilized



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(d) Integration of R&D results
Contents of investigations: ④ Investigation of procedures for integration of R&D results (3/3)
[Investigation results: (2) The flow of utilization of the outputs while integrating research results]



→ Consolidation of information required for the evaluation while selecting and narrowing down options

The procedures related to the method of updating associated with the manner in which output will be utilized or the progress of research & development, during the cycle of investigating ⇒ Identifying issues ⇒ Planning and execution of research & development, are provided.



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## (d) Integration of R&D results Summary

## **Current results**

- The functions and mechanisms for rationally and efficiently carrying out research & development related to treatment and disposal of solid waste were investigated, and until previous years the waste management sheet was provided from the perspective of understanding the current situation, the treatment flows (streams) were provided from the perspective of sharing the direction of the research & development, and the input consolidation chart and temporal axis organization chart were provided from the perspective of ascertaining the correlation between the various types of information and of understanding the progress made.
- This year, the idea behind the criteria for selecting and narrowing down waste streams from the multiple options that remain and the indicators for making that decision were investigated.
- ✓ During the investigations, the viewpoints and indicators that are believed necessary to be considered while selecting and narrowing down based on the investigations of international standards, etc. were consolidated by classifying the viewpoints into 5 categories, namely, "Safe", "Reliable", "Rational", "Quick" and "Field oriented". And, along with that, while actually carrying out the task of selection and narrowing down, the information that needed to be gathered for each indicator was investigated and consolidated including the source of information and the conditions under which the information was obtained (stage of investigation).
- ✓ Further, the roles and applications of each year's output were re-organized while integrating the research results, and the correlation with the method of utilizing the outputs in the entire process of integrating the research results was clarified.

## Issues, etc.

- ✓ It is believed that during research & development related to treatment and disposal of solid waste, it is necessary to repeatedly implement the cycle of establishing issues to be addressed based on the status of investigations at a specific stage and planning and implementing further research & development.
- ✓ While implementing this cycle, during "Integration of research results", the functions and mechanisms believed to be necessary at each stage of the cycle were investigated and the entire process of integrating the research results was clarified based on the results of investigations conducted this year.
- Since the output of integration of research results that was investigated will be utilized and the research progress will be reflected accordingly, and since a variety of information will be required for selecting and narrowing down options, it is believed that the results of clarifying the required information will need to be utilized and inter-disciplinary information will need to be provided.



## 3. R & D management





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## - Knowledge gathering, decommissioning industrial cluster, human resource development -

- Expertise from within Japan as well as overseas is being called upon
- Since FY2014, along with continued cooperation with research institutions within Japan and overseas (CRIEPI, Corporate research institutions, UK's NNL, etc.), useful information was gathered from experts and experienced individuals.
- Realization of the concept of Fukushima Innovation Coast and forming decommissioning industrial clusters in Fukushima
- In order to facilitate participation of local companies in the decommissioning related industry, collaboration with local companies such as those in the Fukushima Prefecture Hamadori region, etc. was considered and some of the trial manufacturing of test equipment related to the development of the sampling system was carried out in collaboration with the local companies.
- Mid-and-long-term human resource development
- The IRID symposium was held and explanatory videos on the results of research & development were played on the IRID website to promote understanding and awareness by presenting the research results and future plans.
- Along with research result presentations including 18 items during the "FY2021 Fall Meeting" of the AESJ, and 2 items (planned) during the "FY2022 Spring Meeting" of the AESJ, reports on treatment technology were presented during the seminar of the back-end task force to promote understanding and awareness.
- Some of the results of characterization were presented during the intensive courses held at the University of Tsukuba (held in July) and the National Institute of Technology, Fukushima College (held in December) as educational activities for the students.
- Some of the results of characterization were presented during the decommissioning human resource development training (held in January 2022) in an effort to promote participation by young researchers.



## - Clarification of conditions and specifications, setting of indicators, coordination with other research & development -

- Clarification of test conditions or development specifications
- Efforts were made to clarify the test conditions and specifications by bringing about exchange of opinions between concerned parties on the details of the outputs of the project.
- Setting of indicators for the achievements of goals
- In the beginning, the indicators that are supposed to serve as the criteria for determining achievement of project goals were investigated, and the Technology Readiness Level (TRL) that was considered to be the goal was set.
- Coordination with decommissioning work and other research and development
- Information on the status of progress of the projects, the issues, etc. was shared by holding regular meetings between related projects. Also, the common system for managing information on project plans and results, which has been developed so far, continued to be utilized.
- Efforts were made for opinion exchange and information sharing with the Canister Project and the Fuel Debris Retrieval Project^{*}, etc. in order to exhaustively promote research & development related to treatment and disposal of solid waste generated in association with fuel debris retrieval.
- O Information was shared with partial proposers (IHI, KURION), and based on that the test conditions were set.
- In connection with the "Project for the Promotion of Nuclear Energy Science & Technology and Human Resource Development through Concentrating Wisdom", information was exchanged between Hokkaido University and The University of Sheffield (Investigation on rational treatment and disposal of contaminated concrete waste considering change in properties due to leaching), and the symposium on the same topic was participated in (lectures were given and young researchers were dispatched).
- Technical information was provided for research at Hokkaido University and University of Fukui (Quantitative evaluation of the changes in long-term state of contaminated reinforced concrete that considers actual equipment environment for rational disposal).

*Canister Project: "Development of technology for collection, transfer and storage of fuel debris" Project

Fuel Debris Retrieval Project: "Development of technology for further increasing the scale of retrieval of fuel debris and reactor internals" Project



# - Research management, project reports, information communication, alternative plans -

## Research management

O A meeting of all concerned parties of this project was regularly held every month (Waste treatment and disposal project coordination meeting), to share the schedule of research & development and the status of progress, and in addition, to determine the policy for resolving issues and implementing measures.

FY2021: April 6, May 7, June 4, July 2, August 6, September 3, October 1, November 5 December 3

FY2022: January 11, February 4, March 4

### Project reports

- With regards to status of progress, the actual schedule chart was submitted to the MRI Secretariat at the end of every month. Also, the status of progress was reported during the Interim Meeting on October 25 - 26.
- Enhancing communication of information
- O The results of this project were posted on the IRID website and information was communicated such that general public can easily understand it. Also, information about the results of this project was communicated by presenting it at academic conferences, etc. held within Japan and overseas by AESJ or other foreign institutions.
- Advanced preparation of alternative plans
- Throughout the duration of the project, alternative plans had been considered in the event that the project does not progress as planned. However, alternative plans did not need to be implemented during this project.



End