

FY2021 Subsidy Project of Decommissioning and Contaminated Water Management Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)

Final Report

(1) Liquid/Gas Phase Systems(2) Criticality Control Technology

June 2023

Technology Research Association International Research Institute for Nuclear Decommissioning (IRID)

IRID

FY2021 Subsidy Project of Decommissioning and Contaminated Water Management Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)

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1. Purpose and goals of "Development of Safety System (Liquid/Gas Phase Systems, Criticality No.2 Control Technology)"

[Purpose of Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)]

At Tokyo Electric Power Company Holdings Inc. Fukushima Daiichi Nuclear Power Station (1F), it is considered that nuclear fuel melted with reactor internals and exists as fuel debris in the reactor pressure vessel (RPV) and primary containment vessel (PCV).

Fuel debris in RPV and PCV is currently considered to be in subcriticality, but since the plant itself is in an unstable state which is different from the initial design, such as the reactor building (R/B), RPV, PCV, etc. being damaged due to the accident, it is necessary to prevent the diffusion of radioactive materials and achieve a stable state by removing fuel debris and maintaining subcriticality of fuel debris.

Against the above background, this project will conduct studies to realize large-scale fuel debris retrieval work under engineering and project management performed by Tokyo Electric Power Company Holdings Inc. (TEPCO) in accordance with the "Mid-and-Long-Term Roadmap towards the Decommissioning of TEPCO's Fukushima Daiichi Nuclear Power Station Units 1-4" (hereinafter Mid-and-Long-Term Roadmap). Development results of this project will be utilized in engineering by TEPCO.

The purpose of this project is to facilitate decommissioning and contaminated water measures of 1F and raise the level of science and technology of Japan, by implementing the project that supports the development of technologies that contribute to decommissioning and contaminated water measures of 1F in accordance with the Mid-and-Long-Term Roadmap and "FY2021 Decommissioning Research and Development Plan" (Decommissioning and Contaminated Water Team Meeting / Secretariat Liaison Meeting (86th)).

In specific, technology related to on-site operation methods shall be developed regarding soluble α nuclide removal technology for α nuclides considered to be eluted in circulation cooling water from fuel debris, RO concentrated water (*) treatment technology, secondary waste treatment technology, as well as criticality approach monitoring technology and neutron absorbent technology. *Liquid waste with high impurity concentration generated by removing impurities from cooling water containing impurities with reverse osmosis membrane (RO membrane)

[Overall development goals]

Perform necessary element technology development and tests regarding system technology and technology for ensuring safety, required to further expand the scale of retrieving fuel debris and reactor internals, in accordance with R&D results obtained up to date.



2. Overview of subsidy project

Here is the system configuration diagram for the liquid/gas phase systems studied in the subsidy project. This term, soluble nuclide removal equipment and sludge dehydration and stabilization treatment system were developed in the subsidy project.



Figure. Liquid/Gas Phase Systems during fuel debris retrieval (subsidy project basis)

Note: This conceptual diagram is an example of equipment configuration (case with no leakage from PCV)



3. Development results up to the previous PJ* and remaining issues ① Development of soluble α nuclide removal technology

: To be studied in this PJ

No.4

No.	ltem	Initiatives and results in the previous PJ	Remaining issues
1	Nuclides to be removed	Particle generation impact of 15 nuclides of 5 elements, U, Np, Pu, Am and Cm, was confirmed through tests. Regarding U, it was confirmed that the need for adsorption removal was low, since solubility near neutrality of pH 5-9 in air atmosphere and concentration in accumulated water in PCV are about two digits lower than the announcement density.	Dissolution behavior and adsorption behavior of α nuclides of U, Np, PU, Am and Cm, respectively, assuming nitrogen environment in PCV
2	Water quality of water to be treated	Evaluation was conducted assuming leaching of seawater components, leaching of concrete components, and injection of sodium pentaborate which is a criticality prevention agent.	Impact of oil, paint components, rust inhibitor components and non-soluble criticality prevention agent
3	Treatment flow rate	Treatment flow rate conditions when installing adsorption removal system inside equipment intended for circulation cooling of accumulated water in PCV, and treatment flow rate conditions when installing adsorption removal system outside equipment intended for circulation cooling of accumulated water in PCV were tentatively set.	Setting of treatment flow rate considering equipment operation schedule
4	α nuclide concentration in assumed water quality	Water quality was evaluated assuming leaching of seawater components and injection of sodium pentaborate which is a criticality prevention agent, and dissolution behavior of each α was grasped. Regarding impact of leaching of concrete, it was confirmed that evaluation in nitrogen atmosphere close to PCV environment is necessary, since α nuclides co- precipitate due to generation of calcium carbonate in atmospheric environment.	Dissolution behavior and adsorption behavior of α nuclides in high pH assuming leaching of concrete components
5	Concentration reduction	Target DF to reduce public exposure impact is set to 100, and announcement density is the concentration reduction target for transport to	Setting of necessary DF considering particulate α nuclide concentration
	request	the water treatment facility. (no progress)	Setting of concentration reduction goal of individual nuclides

Of issues organized in the previous PJ*, No.1 and No.4 were selected as issues to work on assuming control of nitrogen atmosphere. It was suggested that the form of each α nuclide to be removed may be different in nitrogen atmosphere. Therefore, this PJ will evaluate potential impact on solubility and adsorption phenomenon and study soluble nuclide removal system that can remove α nuclides in nitrogen atmosphere.

*FY2018 supplementary budget "Subsidy Project of Decommissioning and Contaminated Water Management (Development of Technology for Retrieval of Fuel Debris and Reactor Internals)" (implemented in FY2019, FY2020)



3. Development results up to the previous PJ and remaining issues ① Development of soluble α nuclide removal technology

No.	ltem	Initiatives and results in the previous PJ	Remaining issues			
6	Applicable adsorbent	As a result of evaluation of α nuclide adsorption performance by an immersion test, active carbon, zirconium phosphate and titanic acid were selected as candidate applicable adsorbents. Since it is at a stage where adsorbents with large equilibrium adsorption were selected through an immersion test, it is an issue to confirm performance when applied to water flow treatment. The evaluation method is an issue since distribution tests handling large amounts of α nuclides cannot be performed with laboratory tests.	Evaluation method of adsorption removal performance when applied to water flow treatment			
		The tower configuration arranges zirconium phosphate, which	Mutual impact of multiple α nuclides			
		in a series with active carbon for removing colloidal α nuclides at the	Impact of adsorption inhibiting components			
7	Adsorption tower configuration	front. Two towers are assumed for each tower assuming a merry-go- round operation. It is necessary to conduct an impact assessment for cases when the respective nuclides exist in the solution together, and also appropriately re-examine the tower configuration based on removal performance and replacement frequency for water flow treatment.	Re-examination of tower configuration based on removal performance and replacement frequency for water flow treatment			
8	Replacement timing / replacement frequency	Since all adsorbents selected as applicable candidates have low Cs adsorption performance, risk of replacement due to increased dose on the tower surface is low. In general, frequency of replacement due to decreased adsorption performance is evaluated with a distribution test, but the evaluation method is an issue since distribution tests handling large amounts of α nuclides cannot be performed with laboratory tests.	Evaluation method of replacement frequency of adsorbent			
		When either replacing the entire adsorption tower or replacing only	Replacement operation			
9	Replacement method	adsorbents, equipment design is required in accordance with the treatment policy of used adsorbents (long-term storage for the time being).	Handling of used adsorbent			
10	Tower size	If the treatment flow rate is 10m ³ /h, it requires the same size as the adsorption tower of the Advanced Liquid Processing System (outer diameter approx. 1m, height approx. 2.5m).	Design of adsorption tower inputting shielding design, replacement methods, etc.			



3. Development results up to the previous PJ and remaining issues

(2) Development of RO concentrated water treatment technology

③ Development of secondary waste treatment technology

: To be studied in this PJ

No.	Item	Initiatives and results in the previous PJ	Remaining issues					
	Water	Assumed water quality of drain water and backwash water dispensed from the particle removal system and input to the sludge collection system was organized based on the element test result and filter test result of the fuel	Select candidate intermediate removal filters based on literature review. Confirm applicability through element tests and acquire properties of generated liquid waste.					
1	quality of water to be treated	debris retrieval metriod PJ. However, since <u>intermediate removal filter has not been selected</u> among particle removal equipment, it is necessary to select candidate equipment and evaluate the properties of generated liquid waste. Additionally, element tests up to the previous fiscal year have only evaluated cases when spherical particles have passed through water, so it is necessary to <u>evaluate properties of filter</u> <u>behavior and generated liquid waste when non-spherical particles pass</u> <u>through water</u> .	Evaluate the impact on the particle removal system when treating non- spherical particles, through element tests. When it is evaluated that there is a predominant influence on behavior of equipment differential pressure increase, recovery rate, and amount of generated liquid waste, incorporate it into the assumed input water quality of the sludge collection system.					
2	Treatment flow rate	Treatment flow rate requirements of the sludge collection system were evaluated based on the operation schedule of the fuel debris retrieval method, filter test results, and the flow rate of the liquid system.	If the assumed input water quality of the liquid system is changed due to future studies, incorporate it into the treatment flow rate requirements of the sludge collection system.					
3	Treatment method	Coagulation-sedimentation treatment was selected as the method to treat filter liquid waste and RO concentrated water, and usable coagulants were selected based on element tests. Additionally, the process of coagulation-sedimentation treatment procedures using the sedimentation separation tank was studied. It is necessary to <u>study applicability to actual equipment regarding</u> <u>coagulation-sedimentation treatment of RO concentrated water</u> using the sedimentation separation tank. Additionally, it is necessary to study reduction of the generated amount of sludge and simplification of operation for the treatment procedures.	Select adsorption treatment using power adsorbents and coagulation- sedimentation treatment using coagulants, as methods to treat RO concentrated water, and select usable powder adsorbents and coagulants.					
4	Properties and generated	Generated amount of sedimentation sludge was estimated based on the coagulation-sedimentation test. As a result, it was evaluated that the generated amount of sedimentation sludge of RO concentrated water is large due to large treatment flow rate and large water content in sedimentation	Consider revising each process of RO concentrated water treatment procedures, in order to reduce the generated amount of sedimentation sludge. Study reduction of added amount of powder adsorbents, reduction of added amount of coagulants and optimization of stirring conditions through element tests.					
4	sedimentati on sludge	sludge. Thus it is necessary to reduce the volume of sedimentation sludge since there are concerns about complication of delivery treatment and increased load on delivery destinations.	Study application of dehydration treatment to sedimentation sludge, select applicable dehydration technology through literature review, and evaluate applicability to actual equipment through element tests.					

In the previous PJ, it was suggested that the amount of sedimentation sludge generated from treatment of RO concentrated water may be large. Thus, this PJ shall conduct studies that contribute to reduction and volume reduction of sludge such as optimization of treatment methods (No.3) and sludge dehydration (No.4). Additionally, dependency on particulate filters (No.1) shall be evaluated in this PJ, as the previous PJ only conducted the filter test using spherical particles and did not conduct an evaluation.



- 3. Development results up to the previous PJ and remaining issues
 - **②** Development of RO concentrated water treatment technology

③ Development of secondary waste treatment technology

No.	ltem	Initiatives and results in the previous PJ	Remaining issues
5	Properties of supernatant	Amount of residual components and SS (suspended solids) in supernatant after solid-liquid separation was organized.	Organize acceptance requirements of soluble nuclide removal equipment and existing water treatment facility, which are delivery destinations, and study necessity of neutralization treatment, removal of neutralizing salts and removal of trace amounts of residual SS components.
6	Operation method	The coagulation-sedimentation treatment process for filter liquid waste and RO concentrated water using the sedimentation separation tank was studied based on the operation schedule of the fuel debris retrieval method as well as element test results. An operation cycle that completes 1 batch treatment in 1 day was studied.	Incorporate the element test results of this term into the coagulation- sedimentation treatment process studied up to the previous PJ.
7	Equipment specifications	Operability of the sedimentation separation tank and each constituent element was verified with an equipment test. Since an event of large particles biting in when opening and closing the gate valve occurred, it is necessary to re-examine the specifications of the gate valve.	Re-evaluate each mechanism of the sedimentation separation tank in element tests using RO concentrated water treatment test equipment to be conducted after this term. Additionally, survey valves applicable to the sedimentation separation tank.
8	Delivery method of sedimentation sludge	Delivery method of sedimentation sludge using the sludge collection container was studied, and it was confirmed with the test equipment that it is applicable to actual equipment. Piping was inserted from the top of the sedimentation separation tank, and it was confirmed that sedimentation sludge can be extracted by pump suction. In the future, acceptance requirements of delivery destinations of sedimentation sludge shall be organized, and size of sludge canister and application of dehydration treatment shall be studied.	When treating and disposing of sedimentation sludge as waste, it is necessary to apply pretreatment in consideration of final disposal of sedimentation sludge as it is delivered to the waste line. If a policy for the final disposal method of sludge is adopted in the future, study necessary pretreatment in the liquid system and incorporate it in the delivery method of sedimentation sludge. When storing sedimentation sludge in storage canisters, it is necessary to apply pretreatment so that the requirements for long-term storage are satisfied as they are delivered to the storage canister line. Organize requirements for long-term storage of sedimentation sludge, and study the shape and moisture content of the sludge collection container.
9	Handling of sedimentation sludge	Assumed properties and generated amount of sedimentation sludge were organized based on the element test result and liquid system requirements.	Share assumed properties of sedimentation sludge with waste PJ and storage canister PJ, and identify issues in each line.



4. Input and output information

This PJ was conducted in coordination with relevant PJ such as the Project of Development of Technology for Containing, Transfer and Storage of Fuel Debris and Project of R&D for Treatment and Disposal of Solid Waste.

ID	Demand-side project	Provider-side project	Description (overview)	Usage of information
1	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Development of fuel debris packaging, transport and storage technology	Acceptance conditions of storage canister	Study of stabilization treatment technology of secondary waste
2	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Research and development of solid waste treatment and disposal technology	Acceptance conditions of waste storage container	Study of stabilization treatment technology of secondary waste
3	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Development of analysis and estimation technology for fuel debris characterization	Study results of formation behavior of fuel debris particles	Study of properties of particles contained in circulation cooling water
4	Development of fuel debris packaging, transport and storage technology	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Properties of sedimentation sludge generated from the liquid system	Identification of issues in handling slurry and sludge fuel debris
5	Research and development of solid waste treatment and disposal technology	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Properties of sedimentation sludge generated from the liquid system	Identification of issues in treatment and disposal of slurry and sludge radioactive waste



5. Implementation schedule

		Implementing operator						FY	2021											FY	2022						
Large category	Small category	(Also list the subcontractor and testing site, if any)	Apr	Мау	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	
Major	milestones		Proje	ct stee	ering n	▲ neeting)	Interim	repor	t F	roject	steeri	ng me	▲ eting	Inte rep	erim port		Projec	t steel mee	ring I	nterim report				Final F	A Report	
(1) Liquid/Gas Phase Systems	a. Conceptual study / test plan	(1) Hitachi GE Subcontractor: None Testing site: Hitachi Research Laboratory	_	Conce	ptual	study /	test p Test	lan prepai	ration	and ar	ranger	nent															: Planned : Actual
 Development of soluble α nuclide removal technology 	c. Preliminary test	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,						Prel	liminar	y test					Ele	ement	test							0			
	e. Summary												Inte	rim su	mmary									Sum	mary		
	a. Conceptual study / test plan	(2) Toshiba ESS Subcontractor: NFD Testing site: Toshiba ESS	-	Conc	eptua	study	/ <u>te</u> st	plan Tes	t prepa	aration	and a	rrange	ement										-				
	 b. Test preparation and arrangement c. Preliminary test 	laboratory, NFD				•				Pre	liminar	ry test	Eleme	nt tes										C F1 F1			
	d. Element test e. Summary											٩	Inte	rim su	mmar	y							``	Sum	nmary	/	
② Development of RO concentrated water treatment technology	a. Conceptual study / test plan	(1) Hitachi GE Subcontractor: Hitachi Plant			Conce	eptual	study	/ test p Test p	lan orepara	ation a	nd tes	t manı	Jfactur	ing of	test e	quipme	ent										
	c. Preliminary test d. Element test	Testing site: Rinkai Plant (Hitachi City)									Pre	limina	ry test	rim su	mman		•		Elerr	ent te	est			Su	mmany		
@ Dovelopment of	e. Summary a. Conceptual study /	(1) Hitachi GE		Conce	ptual	study										, 											
Secondary waste treatment technology	test plan b. Test preparation and arrangement c. Preliminary test d. Element test	Subcontractor: Hitachi Plant Construction Testing site: Rinkai Plant (Hitachi City) Subcontractor: Misuzu Seiko					Tech	nical și	udy / t	test pla	n Test p	repara	ation a	nd tes	t manı	ufactur	ing of	test eo	uipme	nt ent tes	st						
	e. Summary	Testing site: Misuzu Seiko Research Institute										•	Inte	rim sur	mmary									Su	mmary		



6. Implementation framework

Technology Research Association International Research **Tokyo Electric Power Company** Holdings Inc. Institute for Nuclear Decommissioning Formulation of overall plan and coordination of technical supervision Various adjustments from the Coordination of technical management such as progress of technical \geq perspective of applicability in the development field Collaborating development project teams Hitachi-GE Nuclear **Toshiba Energy Systems** Mitsubishi Heavy Development of fuel Japan Atomic Energy Agency debris retrieval method & Solutions Corporation Energy, Ltd. Industries. Ltd. [Element test / technical development] [Element test / technical Development of fuel [Element test / technical development] [Element test / technical (1) Development of Liquid/Gas Phase debris packaging, (1) Development of Liquid/Gas Phase development] development] Systems transport and storage Systems (1) Development of soluble α nuclide (2) On-site operation methods for (2) On-site operation methods for technology (1) Development of soluble α nuclide removal technology • Study of soluble α nuclide removal test removal technology criticality approach monitoring criticality approach monitoring assuming use of actual liquid (accumulated Research and development Study of soluble α nuclide removal test technology and neutron absorber technology and neutron absorber water in reactor building) assuming use of actual liquid (accumulated of solid waste treatment ·Element test assuming fuel debris technology technology water in reactor building) and disposal retrieval work 2 Development of RO concentrated water ·Element test assuming fuel debris treatment technology retrieval work Formulation of criticality Development of solidified Development of analysis Selection of adsorbent and coagulant approach monitoring procedures absorbent technology and estimation Study of applicability to actual equipment (2) On-site operation methods for criticality 3 Development of secondary waste Performance assessment of technology for fuel debris approach monitoring technology and treatment technology Inspection of stabilization treatment characterization neutron absorber technology subcriticality measurement of technology ·Formulation of criticality approach neutron detector Study of applicability to actual equipment monitoring procedures Development of ·Performance assessment of technology for detailed (2) On-site operation methods for criticality subcriticality measurement of neutron approach monitoring technology and investigation inside PCV detector neutron absorber technology ·Formulation of operating procedures for Development of · Formulation of criticality approach absorbents monitoring procedures technology for · Study of impact on debris drying process investigation inside RPV Development of analysis [Details of re-consignment] [Details of re-consignment] [Details of re-consignment] [Details of re-consignment] and estimation ·Subcriticality measurement test (Kyoto ·Subcriticality measurement test (Kyoto ·Subcriticality measurement test (Kyoto ·Structural analysis of solidified absorbents technology for fuel debris University) University) University) (Shizuoka Institute of Science and characterization [Main outsourcing contents] [Main outsourcing contents] [Main outsourcing contents] Technology) ·Operational test of non-soluble neutron •Soluble α nuclide removal test (NFD) Assistance in subcriticality measurement [Main outsourcing contents] Development of technology absorbent (HPC) Corona-type detector (FEO/TENEX-J) test (IPL) ·Contract work for drying tests of test to sort fuel debris and Multicell-type detector (CETD) ·Water flow test of MF membrane and other ·Assistance in study of criticality approach specimens coated with solidified absorbents filters (Misuzu Seiko) monitoring procedures (NS Engineering) radioactive waste (Inspection Development Company Ltd.) ·Cartridge filter test (HPC)



7. Implementation items ① Development of soluble α nuclide removal technology Development background

- There is a plan to build a liquid system as a safety system for fuel debris retrieval. Exposure dose assessment of the liquid system using tentative conditions found that site boundary exposure dose falls below the threshold both during normal times and during an accident but is largely affected by Pu system and its daughter nuclides (Pu-238, Pu-241, Am-241).
- Since α nuclides are not released to the environment at existing nuclear facilities, study and evaluation of effectiveness of the technology to remove α nuclides contained in contaminated water generated during fuel debris retrieval began in FY2017 from the <u>standpoint of keeping</u> exposure caused by α nuclides as low as reasonably achievable.
- "Adsorption" and "RO membrane" are selected as candidate applicable technology to remove soluble α nuclides. "Adsorption" does not require secondary waste treatment as α nuclides are captured with adsorbents, but "RO membrane" requires treatment of concentrated water which is generated by separating α nuclides. This PJ conducted technical development of "adsorption" which requires smaller equipment scale.
- Subsidy projects up to FY2020 conducted adsorption performance assessment of candidate adsorbents using five elements, <u>Pu, U, Am</u>, <u>Cm, and Np, which are α nuclides to be removed</u>. This assessment was conducted under open-air conditions to screen many adsorbents.
- Impact on adsorption performance in the environment of actual equipment has not yet been grasped. Additionally, adsorption performance assessment was conducted using immersion tests up to FY2020, and performance during water flow treatment has not yet been grasped for the study of actual equipment.
- Thus, this PJ studied evaluation methods to grasp the impact on adsorption performance by simulating the environment of actual equipment and grasp performance during water flow treatment for the study of actual equipment.

(1) Development of soluble α nuclide removal technology





Figure Overall flow of development of soluble α nuclide removal technology



7. Implementation items

(1) Development of soluble α nuclide removal technology

· Issues to be addressed in this PJ, and implementation items as well as goals

a. Study of soluble $\boldsymbol{\alpha}$ nuclide removal test assuming use of actual liquid

[Issues]

- There is lack of data such as adsorbent life and impact of competing ion, regarding α nuclide adsorbents selected in the subsidy project conducted in the previous fiscal year (FY2019-FY2020).
- Method of performance assessment test of candidate adsorbents using actual liquid (accumulated water in reactor building) has not been established.

[Implementation items]

- Organization of data required for design and operation of soluble α nuclide removal equipment.
- Study of test methods assuming use of actual liquid (accumulated water in reactor building).
- Study of test equipment and plans assuming implementation of column water flow test with actual equipment.

[Goals]

- Materialization of test methods assuming use of actual liquid (accumulated water in reactor building).
- Materialization of test equipment and plans assuming use of actual liquid (accumulated water in reactor building).



Example of adsorption tower configuration using candidate adsorbents



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(1) Development of soluble α nuclide removal technology

- Issues to be addressed in this PJ, and implementation items as well as goals
- b. Element test assuming fuel debris retrieval work

[Issues]

- Behavior of α nuclides in solution has not been confirmed under conditions simulating the environment inside PCV during fuel debris retrieval.
- Impact of changes in solution water quality on soluble α nuclide removal performance has not been evaluated.

[Implementation items]

- Fabrication of test equipment to simulate the environment during fuel debris retrieval.
- Preliminary test to confirm dissolution behavior of α nuclides under conditions simulating the environment during fuel debris retrieval.
- Selection of applicable adsorbents upon conducting adsorption tests to confirm α nuclide removal performance under conditions simulating the environment during fuel debris retrieval. At the same time, also consider cases of changes in water quality.

[Goals]

- Selection of candidate adsorbents upon acquiring α nuclide adsorption performance data under an environment assuming fuel debris retrieval.
- Setting of water quality adjustment policy of soluble α nuclide removal equipment.

<Image of test equipment to simulate nitrogen injection environment during fuel debris retrieval work>



Simplified glove box





(1) Development of soluble α nuclide removal technology

- Development schedule of this PJ

Items to be			_			F	Y20	21									_	FY2	022		_		_	
studied		5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3
Major milestones			Ir	nterir	n rep	oort V	/			I	nterim	report	t V			Inte	rim re	port 🔻	,			Final ı	eport	▼
a. Study of tests using actual liquid																								
b. Element test 1. Test plan																								
2. Test preparation (preparation of test supplies)								Prepai	ration	of tes	t equip	oment,	etc.											
3. Pre-test									1															
4. Preliminary test / adsorption test (Am, Cm, Np) (U, Pu)	FU	Prelin	ninar Cm, F	y tes Am, Prelim est us	t anc Np ninar sing	l ads y tes [.] U, Pi	orptio t and	n test adsorp	otion															
4. Summary																								
Remarks																								



① Development of soluble α nuclide removal technology

Setting of prerequisite system (Liquid/Gas Phase Systems)

- The liquid system takes water from D/W (drywell) or S/C (suppression chamber) and water level is controlled to confine contaminated water in PCV, and performs circulation cooling, which cools and injects water taken from D/W or S/C, to cool fuel debris.
- In order to limit the range of α nuclides that migrate from fuel debris to liquid phase, α nuclides are removed according to its form (particle removal filter / soluble nuclide removal adsorption tower) from water taken from PCV and reactor building basement.
- The gas system controls negative pressure to confine dust, etc. in PCV, and injects nitrogen to scavenge hydrogen.
- Although air flows in due to negative pressure control, gas phase inside PCV is a nitrogen environment consisting mostly of nitrogen.





1 Development of soluble α nuclide removal technology

- Assumptions and action policy for soluble α nuclide removal system configuration
- The previous PJ studied equipment configuration consisting of receiving tank, adsorption tower, monitor tank, sampling equipment, etc., as a soluble α nuclide removal system when adsorption removal is applied.
- Additionally, adsorption tests were conducted using α nuclide elements to be removed in air atmosphere, and adsorption tower configuration was proposed with impregnated active carbon expected to mainly remove colloidal α nuclides at the front and zirconium and titanic acid expected to remove soluble α nuclides at the rear.
- This PJ shall conduct similar adsorption tests in nitrogen atmosphere simulating gas phase environment in PCV and evaluate solubility of α nuclides and impact on adsorption performance, thereby confirming whether tower configuration and applicable adsorbents set in the previous PJ need to be revised.



Figure Schematic system diagram and proposed tower configuration* of the soluble α nuclide removal system

*Supplemental information on tower configuration

- The number of towers in each adsorption tower is tentatively set at 2 to allow merry-go-round operations. The necessary number of towers of each adsorption tower needs to be evaluated in the future.
- In order to reduce the number of adsorption towers as much as possible, adsorption tower of impregnated active carbon which has high removal performance of colloidal α
 nuclides in addition to soluble α nuclides and adsorption tower of zirconium which has high adsorption performance for multiple soluble α nuclides shall be installed. However,
 since Np adsorption performance of impregnated active carbon and zirconium is relatively high and concentration reduction during water flow treatment is insufficient, titanic
 acid adsorption tower shall be installed at the rear of zirconium.



7. Implementation items

(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Study of environmental conditions of actual equipment and policy on incorporation to environmental conditions of tests

- Proposed management policy and proposed control values of PCV gas phase environment were organized in the below table based on the status of operator engineering study.
- Oxygen concentration is an item related to the degree of air inflow. Since it is considered to set the control value below 4%, it shall be incorporated in the conditions in the test evaluation. (However, 4% is a tentative value and not a definitive value)
- Since oxygen concentration in air is approx. 20%, atmospheric components in gas phase in PCV are estimated at concentrations less than 1/5 of that of air.

Table Proposed management policy and proposed control value of environment items during fuel debris retrieval (gas system)

No.	En	vironment items	Management requirement	Proposed management policy	Proposed control value	Remarks
1		PCV gas phase pressure	Gas phase confinement (leakage prevention)	Manage at negative pressure by managing exhaust flow rate	Undetermined *1	*1: Positive pressure management and equal pressure management are options when negative pressure management is not feasible
2	Ga	Nitrogen concentration	None	(Management of nitrogen supply amount)	_	Nitrogen concentration itself is not subject to management
3	as phase env	Hydrogen concentration	Gas phase confinement (prevention of fire and explosion)	Manage below the lower explosive limit of hydrogen by managing nitrogen supply amount and exhaust flow rate	Less than 2% *2	*2: Not a definitive value as it is a tentative value
4	ironment	Oxygen concentration	Gas phase confinement (prevention of fire and explosion)	Manage below Zr fire conditions by managing nitrogen supply amount and exhaust flow rate	Less than 4% *3	Concentration increases due to inflow of air during negative pressure management *3: Not a definitive value as it is a tentative value
5		Carbon dioxide concentration	None	In the course of events	_	Concentration increases due to inflow of air during negative pressure management
6		Dust concentration	Gas phase confinement (leakage prevention)	Manage below the prescribed activity concentration by managing exhaust flow rate	Undetermined *4	*4: Assuming below concentration equivalent to public exposure of 5mSv at total release

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(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

- Study of water quality conditions assumed for actual equipment

- Gradual changes in water quality is expected before fuel debris retrieval work starts, due to circulation cooling operation with the liquid system and PCV negative pressure management with the gas system.
- When fuel debris retrieval work starts, pH in liquid phase is expected to increase due to leaching of concrete components during cutting.
- Additionally, pH increase is expected to increase the amount of carbon dioxide in gas phase that dissolves in liquid phase.

Table Water quality of accumulated water in PCV and status of evaluation of α nuclide adsorbent performance in operational states of liquid system / gas system and fuel debris retrieval

Oper	rational state	STEP1	STEP2	STEP3	STEP4
Circu	lation cooling	×	о	o	0
PCV negative pressure management		×	×	o	ο
Fuel debris retrieval work		×	×	×	0
Water quality of accumulated	Outline	Basic water quality is the same as that of fresh water, since fresh water is injected from outside and injected water leaks outside PCV.	Circulation of fresh water could cause leaching of concrete components, etc. Since gas phase is nitrogen atmosphere, there is no atmospheric influence.	Negative pressure management of PCV increases carbon dioxide concentration of gas phase due to inflow of air into PCV, which increases inorganic carbonic acid concentration underwater.	pH is increased by increase of eluted amount of concrete components due to fuel debris retrieval work. pH increase increases inorganic carbonic acid concentration. Injection of boric acid water is also assumed to prevent criticality.
water in PCV	Estimated pH	Around 7	Around 7-9	Around 6-9	Around 6-11
	Inorganic carbonic acid* concentration (estimated)	Around 1ppm	Around 1ppm	Several ppm (influence of air inflow)	Several to several dozen ppm (influence of air inflow + pH increase)
Status of evaluation of α nuclide adsorbent performance		(Not applicable)	(Not applicable)	Candidate adsorbents were selected from adsorption tests up to FY2020	Not evaluated



7. Implementation items

① Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Inspection and evaluation regarding dissolved form of α nuclides

- In the previous PJ, thermodynamic equilibrium calculations were conducted for systems with carbon dioxide in the gas phase. It was evaluated that under alkaline conditions, abundance ratio of dissolved α nuclides in forms of carbonate ion coordination and hydroxide ion coordination increases compared to neutral conditions.
- In nitrogen environment and atmospheric environment, chemical forms of α nuclides may be affected under alkaline conditions since carbon dioxide concentration changes.
- Additionally, under acidic conditions, abundance ratio of dissolved α nuclides in metal ion forms (such as Pu³⁺) may increase compared to neural and alkaline conditions.



Figure 1 Evaluation result of Pu solubility by thermodynamic equilibrium calculations using carbon dioxide concentration conditions in the gas phase as parameters (50 times diluted seawater) (Result of thermodynamic equilibrium calculations of previous PJ)



(2) Carbon dioxide concentration in gas phase: 350ppm (simulated atmospheric environment)



Figure 2 Evaluation result of Pu dissolved forms by thermodynamic equilibrium calculations using carbon dioxide concentration conditions in the gas phase as parameters (50 times diluted seawater) (Result of thermodynamic equilibrium calculations of previous PJ)



7. Implementation items

(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Setting of element test conditions

- Assumed water quality conditions of accumulated water in PCV were divided into patterns and organized in the following table from three perspectives: ①management policy of each environment item during fuel debris retrieval, ②study result of water quality conditions considered to predominantly change due to fuel debris processing, and ③analysis result of accumulated water in PCV conducted to date.
- For the evaluation assuming increase in pH due to elution of concrete components, the evaluation was conducted with around pH9, 10 which assumes impact of pH decrease due to dissolution of carbonic acid to be minimal and sedimentation of carbonate not to be generated.
- Additionally, evaluation was also conducted assuming water quality adjustment (pH adjustment), as adsorption performance could be reduced if water quality of water to be treated changes in actual equipment.

No.	Purpose	Liquid conditions	рН⁺	Gas phase conditions	Remarks							
1		1000 times diluted seawater	6-7	Ambient air /	Regarding conditions that were evaluated up to the previous PJ, the policy is to diver the results of the							
2		200 times diluted seawater	6-7	nitrogen	previous PJ, and tests shall be conducted with conditions whose results have not been acquired.							
3	Water	1000 times diluted seawater	Around 9		Assume operation near the analyzed value of chloride ion concentration in accumulated water to date							
4	quality conditions	+ Ca(OH)2	Around 10	N litera era er	 Assume increase in pH due to leaching of concrete components 							
5	for actual	200 times diluted seawater +	Around 9	Nitrogen	Assume operation near the upper control limit (100ppm), due to the condition of high leaching of seawater							
6	equipment	Ca(OH)2	Around 10		Assume increase in pH due to leaching of concrete components							
7		Sodium pentaborate water (7000ppm as B)	8-9	Ambient air / nitrogen	Apply for impact assessment of injection of sodium pentaborate.							
8	Conditions assuming	Around 3		Ambient air /	 In order to grasp the trend of which range, neutral, alkaline or acidity, is appropriate to be adjusted to, 							
9	quality adjustment		Around 5	nitrogen	conduct an evaluation under acidic conditions (pH3, 5) whose data has not been acquired to date. •Tentatively set hydrochloric acid as the pH adjuster, in consideration of impact on rear equipment							

Table Element test conditions in this PJ

Scope of this PJ

*: Set with initial pH



(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Adsorption test procedures

- Figure 1 shows the test system for evaluation simulating the environment inside PCV (nitrogen atmosphere).
- Simplified glove box (simplified GB) is installed in the hood of the laboratory from the perspective of radiation control, and nitrogen injection line is connected from the N2 cylinder to create nitrogen atmosphere inside GB. Oxygen concentration inside the simplified GB is regularly checked, and nitrogen injection operation is accordingly performed so that oxygen concentration does not exceed 4%.
- Temperature control inside the simplified GB is difficult due to space limitations. So, temperature is measured and recorded during tests to collect data to consider the impact of differences in temperature if it emerges.
- Figure 2 shows the image of adsorption test procedures. Amount of adsorption on the adsorbent is evaluated by shaking the test solution containing adsorbent and α nuclide tracer (test container is PP (polypropylene) container) in the simplified GB (nitrogen atmosphere) (21 days) and analyzing the α nuclide concentration in the solution after 0.1µm filtering.





(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Adsorption test result: Alkaline conditions simulating concrete leached components

- Immersion tests were conducted for zirconium and impregnated active carbon using Pu under alkaline conditions (initial pH adjusted to 9 or 10 for 1000 times diluted seawater) and nitrogen conditions.
- Although zirconium showed relatively high distribution coefficient (Kd), it was lower than that of impregnated active carbon, which made it
 difficult to determine as an applicable adsorbent in alkaline conditions. There is possibility that adsorption performance may decrease under
 alkaline conditions.
- Kd of impregnated active carbon after 21 days of immersion is about the same as that of the previous PJ (liquid phase: around pH 6-7, 1000 times diluted seawater; gas phase: atmospheric environment), which indicates robustness to pH. However, pH decreased to 7 after immersion, which shows the possibility that adsorption performance may decrease during water flow treatment with alkaline conditioned solution.
- Based on this result, pretreatment to adjust pH upstream adsorption tower to a constant level could be effective, in order to demonstrate stable removal performance even when the quality of water to be treated fluctuates in actual equipment.



(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Adsorption test result: Range of pH adjustment upstream adsorption tower

- To grasp the appropriate pH range for adjusting pH upstream adsorption tower at a constant level (Figure 1), Figure 2 shows the result of distribution coefficient (Kd) of Pu and Cm to zirconium adsorbent in nitrogen environment at each pH.
- The adsorption test (immersion test) with solutions of initial pH 3 and 5 (1000 times diluted seawater added with hydrochloric acid) showed that Kd decreased by one order of magnitude than the condition with initial pH 9 and 10. Thus, it is possible that adjusting the water quality to the acidic side could reduce adsorption performance.
- To summarize the results of immersion tests for initial pH to date, Kd is relatively high when the pH after immersion from initial pH is ranged around pH 6.5-9. Since there is concern of decrease in Cm adsorption performance when pH becomes lower than 6, the pH adjustment range was tentatively set around pH 7-9.



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(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Selection of candidate adsorbents

- Below figure shows the distribution coefficient (Kd) of candidate adsorbents subject to evaluation in this PJ, obtained from adsorption tests (immersion tests) using Np, Pu, Am and Cm in nitrogen atmosphere around initial pH 6-9.
- Pu, Am and Cm, which are considered to be contained in large amounts in water to be treated with actual equipment, showed high distribution coefficient (Kd) with titanic acid, zirconium and impregnated active carbon. On the other hand, silicotitanate was eliminated as a candidate since it may have poor Pu adsorption performance.
- Therefore, titanic acid, zirconium and impregnated active carbon shall be set as candidate adsorbents, and Np, Pu, Am and Cm shall be removed with combinations of titanic acid, zirconium and impregnated active carbon to reduce α nuclide concentration of water to be treated.



IRID

7. Implementation items

(1) Development of soluble α nuclide removal technology (b. Element test assuming fuel debris retrieval work)

Result of study of schematic system and identification of issues for application to actual equipment

- Below figure shows the proposed schematic system configuration when assuming adjustment of water quality upstream adsorption tower, which was newly added to the development goal in addition to the conventional adsorption tower system configuration (areas revised from the previous PJ is indicated in blue). A system that measures pH (assuming pH 5.5-11) of water to be treated in advance, and feeds water to the adsorption tower after adjusting pH to 7-9 with the pH adjustment tank if water is alkaline (pH 10 or higher) shall be assumed.
- Three issues up to application of the system that assumes adjustment of water quality upstream adsorption tower to actual equipment were identified.

- Issue ②: Setting of range of pH adjustment (setting of target pH to be adjusted by chemical injection / setting of action policy for deviation from target pH)
- Issue ③: Formation of precipitates due to pH adjustment (determining whether filter is needed)



Figure Proposed schematic system configuration for soluble nuclide removal (when assuming water quality adjustment)



Issue ①: Selection of pH adjuster (hydrochloric acid was tentatively set for the test, but pH adjuster shall be selected including other means)

7. Implementation items

(1) Development of soluble α nuclide removal technology (a. Study of soluble α nuclide removal test assuming use of actual liquid)

Necessity of water flow test using actual liquid

- By conducting immersion tests with conditions assuming changes in water quality during fuel debris retrieval, adsorbents with large equilibrium adsorption capacity for α nuclides are evaluated, and impregnated active carbon and zirconium are selected as candidate adsorbents. However, since performance of candidate adsorbents during water flow has not been evaluated, it is necessary to conduct water flow tests to evaluate applicability of the process to actual equipment and acquire necessary data for design and operation.
- Water flow tests require much α nuclides, but it is realistic to conduct water flow tests using actual liquid (hereinafter actual contaminated water) due to limitations of α nuclide tracer amount that can be handled at the laboratory. Therefore, the plan up to the water flow test using actual contaminated water was studied.



Figure Flow up to selection of process applicable to actual equipment

Target α nuclide concentration of outlet water: Three conditions (tentative) of

A Bq/L, 40 Bq/L and 400 Bq/L were set in operator engineering

Target replacement frequency of adsorption tower: Tentatively set at 1 month (30 days) based on the replacement frequency of existing water treatment facility



7. Implementation items

(1) Development of soluble α nuclide removal technology (a. Study of soluble α nuclide removal test assuming use of actual liquid)

· How to proceed with water flow test

- It is necessary to verify removal performance of adsorption tower during water flow treatment and its adsorption performance maintenance period, before starting operation of actual equipment. It was confirmed that such verification requires water flow test (verification test) in 1/10 scale of actual equipment through literature review.
- Since it is necessary to set adsorption tower configuration in order to set the specifications of verification test equipment, water flow test
 shall be conducted using column of actual equipment length with the same water flow conditions (SV^{*1} or LV^{*2}) as those of actual
 equipment to confirm that the target removal performance and performance maintenance period of the set adsorption tower is satisfied.
- While water flow conditions can be matched with those of actual equipment for the test with column of actual equipment length, it requires a
 large volume of test solution. Thus, water flow test shall be conducted matching SV of water flow conditions with that of actual equipment
 and using mini column which requires a small amount of test solution, in order to confirm feasibility of adsorption tower configuration
 selected in this PJ.
- Based on the above, a plan was developed to proceed with the water flow test up to application to actual equipment in the following order: water flow test with mini column, water flow test with column of actual equipment length, water flow test (verification test) in 1/10 scale.
- The following page shows the study on the materialization of the plan of water flow test with mini column to be conducted in the first step.



Figure Proposed way to proceed with water flow test up to application to actual equipment



*1 SV: Space velocity *2 LV: Linear velocity

(1) Development of soluble α nuclide removal technology (a. Study of soluble α nuclide removal test assuming use of actual liquid)

- Study of test equipment configuration and test conditions of water flow test with mini column
- Test equipment configuration and test conditions of water flow test with mini column were studied.
- Water flow test solution is prepared in the inlet tank and fed to the test column. A filter is installed at the front of the column to capture solids. Sample collection points are also secured to evaluate changes in concentration and pH before and after water flow.
- In order to secure a certain period of water flow, several dozen liters (target) of test solution for several thousand BV* is secured.
- Two cases, 1E+03BQ/L (same level as current actual contaminated water) and 1E+05Bq/L (assuming two-digit increase in concentration compared to the current actual contaminated water), are set for gross α concentration of water flow test solution to prepare test solutions containing the same amount for the water flow test.
- Currently, no actual contaminated water with gross α concentration exceeding 1E+05Bq/L after filter filtering has been confirmed. Thus, it is necessary to study solution preparation methods to secure appropriate concentration for the water flow test.

*BV: Bed Volume; water flow rate per adsorbent filling volume

Figure Schematic system configuration of test equipment of water flow test with mini column (draft)

(1) Development of soluble α nuclide removal technology (a. Study of soluble α nuclide removal test assuming use of actual liquid)

- Study of method to secure soluble α nuclide concentration in test solution

- According to the disclosed analysis result of actual contaminated water, most α nuclides contained in contaminated water are present in non-soluble form and the percentage of soluble form is small. Thus, it is necessary to perform acid dissolution of non-soluble α nuclides in order to secure ionic α nuclide concentration. Dissolution behavior when adjusting to the water quality assumed for actual equipment shall also be confirmed. (No.3-8 in below figure)
- However, if acid dissolution and water quality adjustment are performed, ion amount in the solution may drastically differ from liquid conditions for treatment with actual equipment and preparation operation itself may be difficult. Thus, solution preparation methods with no acid dissolution and water quality adjustment shall be assumed. (No.1-2 in below figure)
- Solution preparation methods for water flow test solutions shall be set by testing such solution preparation methods using actual contaminated water and confirming soluble α nuclide concentration in the solution.

Figure Proposal of securing water flow test solution

(1) Development of soluble α nuclide removal technology (a. Study of soluble α nuclide removal test assuming use of actual liquid)

Identification of issues for water flow tests using actual contaminated water

- Below figure shows how to proceed with the water flow test before operation of actual soluble α nuclide removal system assuming the adsorption tower method, based on the study results so far.
- Simulated demonstration is required before operation of actual equipment, in which water flow test equipment in 1/10 scale of actual
 equipment is required. As the application phase to set adsorbent / adsorption tower configuration and number of adsorption towers, water
 flow test with column of actual equipment length and water flow test with mini column shall be conducted. Such tests shall be conducted
 upon confirming whether the test solution can be secured, as securing the test solution is an issue.
- Future issues of water flow tests using actual contaminated water were organized as follows.
 - Issue ①: Setting of policy to acquire data for setting conditions for operation of actual equipment
 - Issue ②: Setting of sampling specifications (target, location, method, etc.) for actual contaminated water
 - Issue ③: Materialization of water quality adjustment method of actual contaminated water (including whether it can be implemented at the site)
 - Issue ④: Study of alternative means for cases when α nuclide concentration suitable for water flow tests cannot be stably secured
 - Issue (5): Study of materialization of water flow test equipment
 - Issue (6): Selection of areas to implement water flow test
 - Issue ⑦: Setting of analysis specifications (measurement/analysis items, analysis framework) for water flow test solution

(1) Development of soluble α nuclide removal technology

- Summary of issues of soluble α nuclide removal equipment using adsorption tower method
- Below table shows the status of efforts and progress on issues identified at the start of this PJ as well as remaining issues, toward application of soluble α nuclide removal equipment applied with adsorption tower method to actual equipment. (Issues requiring test evaluation are indicated in red)

No.	ltem	Status of efforts and progress on issues	Remaining issues
1	Nuclides to be removed	At the initial stage of this PJ, 15 nuclides of 5 elements,U, Np, Pu, Am and Cm, were tentatively set as nuclides to be used in exposure assessment and nuclides to be removed with the Advanced Liquid Processing System. Analysis results of contaminated water in different PJ found that soluble α nuclides hardly exist in accumulated water in the building and that soluble α nuclides, mostly consisting of Pu-238, 239, 240 and Am-241, exist in accumulated water in PCV. Additionally, tests conducted in this PJ simulating the environment and water quality of actual equipment found that it is highly possible that U can be eliminated from nuclides to be removed, as filtrate after 0.1 μ m filter filtering hardly contained any U.	•Whether U (U-234, U-235, U-236, U- 238) can be eliminated from elements to be removed
2	Water quality of water to be treated	At the initial stage of this PJ, regarding water quality of water to be treated, increase in Ca concentration and pH was assumed due to elution of seawater components and concrete components with chloride ion concentration below 100ppm. The test simulating the environment of actual equipment, conducted in this PJ, found that pH is maintained at a somewhat high level and pH tends to decrease due to inflow of air in nitrogen environment assuming actual equipment. It was also confirmed that there is no extreme decrease in oxidation-reduction potential.	• Impact assessment with water quality conditions (e.g. case when liquid is returned from the secondary waste treatment facility) that have not been studied
3	Treatment flow rate	The range of treatment flow rate was tentatively set to 2-10m ³ /h, assuming operation of actual equipment. •2m ³ /h: Assuming installation of the system outside the circulation cooling equipment and treatment of 22m ³ /day •10m ³ /h: Assuming installation of the system inside the circulation cooling equipment	Setting of treatment flow rate considering nuclide removal efficiency
4	α nuclide concentration in assumed water quality	In the initial stage of this PJ, α nuclide concentration of water to be treated was not set and the policy was to conduct adsorption tests under the concentration condition of 10Bq/ml from the perspective of analytical precision during tests. Assumed conditions in operator engineering were discussed, and a policy to set 100Bq/ml as the maximum value for concentration condition of gross α was established, which was incorporated in the study and plan.	 Setting of concentration conditions for each nuclide Confirmation of α nuclide concentration with draft procedures for water flow test solution preparation (test using actual contaminated water)

Table Status of efforts on issues for application of soluble α nuclide adsorption removal system to actual equipment (1/3)

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(1) Development of soluble α nuclide removal technology

- Summary of issues of soluble α nuclide removal equipment using adsorption tower method
- Below table shows the status of efforts and progress on each issue as well as remaining issues. (Issues requiring test evaluation are indicated in red)

Table Status of efforts on issues for application of soluble α nuclide adsorption removal system to actual equipment (2/3)

No.	ltem	Status of efforts and progress on issues	Remaining issues
5	Concentration reduction request	At the initial stage of this PJ, target DF to reduce public exposure impact was set to 100, and announcement density was the concentration reduction target for transport to the water treatment facility. Assumed conditions in operator engineering were discussed, and a policy to set DF of gross α to 10, 100 or 1000 according to inlet conditions was established, which was incorporated in the study and plan. Additionally, the test plan was studied since water flow tests using actual contaminated water are required to verify DF.	 Setting of removal requirements for each nuclide Materialization of test plan using actual contaminated water
6	Applicable adsorbent	Immersion tests were conducted using U, Np, Pu, Am and Cm, which are elements to be removed, to evaluate equilibrium adsorption of candidate adsorbents, and impregnated active carbon, zirconium and titanic acid were selected as adsorbents with large equilibrium adsorption capacity and high prospects for application. Such adsorbents have large equilibrium adsorption under neutral conditions, but since equilibrium adsorption relatively decreased under alkaline conditions assuming concrete leached components, it is inferred that appropriate water quality adjustment is necessary.	 Setting of adsorbents that satisfy removal requirements (including new candidate adsorbents) Setting of test methods that reduce the impact of fluctuations in water quality during the test period Evaluation of generated amount of secondary waste Investigation/evaluation of impact of disposal of used adsorbents
7	Adsorption tower configuration	Confirmation by water flow tests is required to set the adsorption tower configuration, but since small- scale tests in the laboratory is not sufficient for confirmation, confirmation methods with water flow tests using actual contaminated water were studied.	 Materialization of test plan using actual contaminated water
8	Replacement timing / replacement frequency	Target replacement frequency was set at once a month, and a policy to confirm in the water flow test using actual contaminated water that removal performance is maintained after feeding water equivalent to one month of treatment was studied.	 Evaluation of removal performance maintenance period Materialization of test plan using actual contaminated water
9	Replacement method	A replacement policy was set to replace the entire adsorption tower from the perspective of confinement.	None
10	Tower size	As the adsorption tower assuming treatment flow rate of 2-10m ³ /h, the same size as that of adsorption tower of Advanced Liquid Processing System with system flow rate of 250m ³ /day was tentatively set as the study conditions for the water flow test using actual contaminated water.	None
	RD	Sinternatione	

7. Implementation items

(1) Development of soluble α nuclide removal technology

- Summary of issues of soluble α nuclide removal equipment for treatment process other than adsorption tower method

• Additional issues were organized based on results of study in this PJ.

Table Status of efforts on issues for application of soluble α nuclide adsorption removal system to actual equipment (3/3)

No.	ltem	Status of efforts and progress on issues	Remaining issues
11	Water quality adjustment	Performance assessment has been conducted up to the previous PJ in neutral conditions. In this PJ, adsorption tests were conducted in acidic and alkaline conditions under nitrogen environment. Equilibrium adsorption showed a decreasing trend in some conditions compared to neutral conditions. It was inferred that pretreatment is effective so that pH of adsorption tower inlet water is constant near neutral. Accordingly, system configuration was studied with the assumption of water quality adjustment.	 Selection of pH adjuster Setting of range of pH adjustment Formation of precipitates due to pH adjustment
12	Colloidal measures	In July 2021, MSIV leakage water from PCV accumulated water was sampled and analyzed at Unit 3, and it was confirmed that α nuclides in contaminated water are not only in ion forms but also include particle and colloidal forms that remain after filter filtering (blue area in below figure). Since colloidal α nuclides may not be removed with the adsorption tower method, depending on the amount and water quality, it was judged that it is necessary to set measures to remove colloids (water quality adjustment, coagulation-sedimentation, batch treatment, etc.).	•Setting of policy to remove colloidal α nuclides

[Reference] E 3 MSIV room	Evaluation and	analys	is of α	nuclide	s of sa	mples o	of Unit	TER	200	
 Stepwise f filter were Even with 	filters were instal analyzed. 0.02µm filtrate, /	led for t Am and	he test v Pu are l	vater, ar nigher th	nd collec nan the a	ted mate	erials and ement de	l filtrate a nsity.	at each	
Test water	Destinite sites	Topic State Stat								
1000	Particle size	Q-235	Np-237	0-238	Am-241	Cm-244	Cm-242	Pu-238	Pu-239+24	
filter	> 10 µm	1.3E+01	2.7E+01	1.16+02	4.4E+05	3.1E+05	7.1E+03	6.9E+05	2.2E+05	
Filtrate	103 hw	3.36-08	1.76-01	2.76-02	1.1E409	9.56+02	1.76=01	1.96+03	6.3E+02	
<u> </u>	1~0.1 µm	1.18-02	1.85-01	8.56-02	4.96+01	1.52+01	<2.0E+00	8.75+01	2.98+01	
1µm	<0.02µm (filtrate)	<5.5E-04	6.4E-01	5.66-04	1.6E+01	<3.0E+00	<5.0E+00	3.5E+00 1.9E+02	5.85+01	
Filtrate	[Reference]									

0.1µm		Ba/L						
filter	Particle size	Gross a	Cp-134	Cs-157	Co-63	3b-125	Eu-154	
	> 20 µm	2.18+06	3.26+04	7.8E+05	5.5£+05	1.8E+05	6.4E+05	
Itrate	10~1 µm	1.36+01	2.6E=0.2	6.5E+03	1.96+03	6.0E+02	1.5€+03	
No.	1~0.1 µm	1.05+01	<1.06+02	1,78+03	7,85+01	<2.05+03	<2.01+03	
0.02µm	0.1~0.02 µm	3.76+01	<8.0E+62	1.18+04	<4.0E+03	<1.00+03	<2.06+03	
filter	<0.02µm (filtrate)	2.45+03	1.32+05	3.12+06	9.75+03	<9.02+03	<2.0E+03	

Source: TEPCO HD "Progress of treatment of accumulated water from buildings March 31, 2022"

(1) Development of soluble α nuclide removal technology

Summary

[Results to date]

No.	Implemented item	Goal	Development results		
a-1	Study of soluble αMaterialization of test methods assuming u of actual liquidnuclideof actual liquidremoval test 		Regarding the development goal of soluble α nuclide removal equipment using the adsorption tower method, a plan was developed to study steps for evaluating α nuclide adsorption removal performance and performance maintenance period before application to actual equipment and proceed with water flow test with mini column, water flow test with column of actual equipment length and water flow test in 1/10 scale.		
a-2		Materialization of test equipment and plans assuming use of actual liquid (accumulated water in reactor building)	Of water flow tests to be conducted before application to actual equipment, test equipment configuration was materialized for the water flow test with mini column and the test solution preparation method using actual contaminated water was studied. Since the solution preparation method needs to be confirmed with actual contaminated water in order to secure necessary α nuclide concentration for the water flow test, test details using actual contaminated water water water in order to secure studied and the test plan up to start of water flow test was materialized.		
b-1	Element test assuming fuel debris retrieval work Element test adsorbents upon acquiring α nuclide adsorption performance data under an environment assuming fuel debris retrieval		Adsorption test (immersion test) was conducted for candidate adsorbents (impregnated active carbon, zirconium, etc.) identified in the previous PJ, in nitrogen environment and alkaline conditions. It was confirmed that impregnated active carbon achieves the same level of Pu removal rate in alkaline conditions as that in neutral conditions in the previous PJ. On the other hand, zirconium showed slightly lower Pu and Cm removal rate in alkaline conditions, which confirmed decreasing trend of adsorption performance in alkaline conditions for some adsorbents. Thus, adsorption test assuming pH adjustment and adsorption tower system configuration was conducted, and the same adsorbents as candidate adsorbents selected thus far were selected as candidates.		
b-2		Setting of water quality adjustment policy of soluble α nuclide removal equipment	In adjusting pH at the front of the adsorption tower, adsorption tests were conducted in acidic conditions as evaluation to select the appropriate pH range. It was confirmed that α nuclide adsorption performance becomes low in acidic conditions, and the policy to adjust pH near neutral was tentatively set.		



① Development of soluble α nuclide removal technology Summary

[Future issues to be studied]

- Adsorption evaluation with water quality conditions that have not been studied thus far (impact assessment with return water from secondary waste treatment facility) shall be conducted based on studies conducted in other PJ.
- It is necessary to acquire necessary data for evaluation of applicability of the process to actual equipment as well as design and operation (removal performance and adsorption performance maintenance period of candidate adsorbents during water flow), in future water flow tests, and it is an issue to prepare a large amount of test solutions while securing the prescribed α nuclide concentration. It shall be evaluated whether requirements of water flow test solutions can be satisfied by using the actual contaminated water of Fukushima Daiichi.
- Additionally, materialization of the test plan using actual contaminated water shall be studied. It shall also be studied whether necessary data can be acquired without using actual contaminated water, in case tests using actual contaminated water cannot be established.
- New candidate adsorbents that have not been evaluated thus far shall be evaluated, and disposal impact of used adsorbents shall be investigated and evaluated regarding the candidate adsorbents.
- In addition to technical development with the adsorption tower method, which had been the development goal, applicability of treatment processes such as water quality adjustment, coagulation-sedimentation and batch treatment shall be evaluated as measures against decreased treatment performance due to changes in water quality and measures for colloid components that could not be removed with adsorption removal.

[Summary]

Since the possibility of changes in treatment performance due to changes in water quality was suggested from the element test results, evaluation was conducted assuming water quality adjustment equipment, candidate adsorbents were selected on the premise of pH adjustment, and schematic system configuration was tentatively set. A phased water flow test plan from small scale to scale of verification of actual equipment shall be materialized in order to acquire necessary data for evaluation of applicability of the process to actual equipment as well as design and operation.



No.36

7. Implementation items ② Development of RO concentrated water treatment technology

Study flow





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7. Implementation items ② Development of RO concentrated water treatment technology

[Issues]

- Nuclide removal by coagulation-sedimentation is studied as a treatment method of RO concentrated water generated by RO membrane, and it is necessary to establish treatment procedures.
- As powder adsorbents and coagulants that can be used for RO concentrated water treatment have not been selected, its data on nuclide adsorption performance and coagulation-sedimentation performance is insufficient.

[Implementation items]

- Candidate powder adsorbents and coagulants shall be selected based on literature review and actual use at existing water treatment facility.
- Nuclide removal performance of powder adsorbents shall be evaluated with adsorption tests.
- Coagulation performance of coagulants on powder adsorbents shall be evaluated with coagulation-sedimentation tests.
- Applicability of the developed RO concentrated water treatment procedures shall be evaluated with simulated RO concentrated water tests in beaker scale and equipment scale.

[Goals]

- Evaluate nuclide removal performance of powder adsorbents and coagulation-sedimentation performance of coagulants.
- Select applicable candidates of powder adsorbents and coagulants.
- Draft RO concentrated water treatment procedures.





7. Implementation items ② Development of RO concentrated water treatment technology

Sludge collection system configuration

Collect solid components from liquid waste generated from non-soluble nuclide removal equipment and RO membrane equipment



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7. Implementation items ② Development of RO concentrated water treatment technology

Requirements of sludge collection system configuration

Liquids to be treated at each sedimentation separator and treatment requirements are as follows.

*Estimated with water quality conditions set in the subsidy project. Underlined parts were revised incorporating the results of this PJ.

No.	ltem	Roughly removed liquid waste treatment facility	Intermediate removal / final liquid waste treatment facility	RO concentrated water treatment facility	
1	Water to be treated	Drain water / backwash water generated from rough removal equipment	Backwash water generated from intermediate removal equipment and final treatment equipment	RO concentrated water generated from RO membrane equipment	
2	Contained particle components (subject to coagulation- sedimentation)	Molten fuel, reactor internals, concrete components, other solid components (particle size 100 to several dozen µm)	Molten fuel, reactor internals, concrete components, other solid components (particle size several dozen to 0.1µm)	Powder adsorbent for nuclide adsorption	
3	Particle concentration	Several thousand to 10000 ppm From filter element test results	Several thousand to 20000 ppm From filter element test results	When adsorbent is added: Several hundred ppm After CF concentration: Approx. 10000 ppm From adsorbent element test results	
4	Treatment flow rate	Intermittent drain: <u>100 m3/year</u> From filter element test results	1.4μm MF membrane: <u>30 m3/year</u> 0.1μm UF membrane: 10 m3/year 0.05μm UF membrane: <u>50 m3/year</u> From filter element test results	8-11 m3/day Calculated assuming RO membrane concentration factor of 2 to 3 times for debris retrieval work water (22 m3/day)	
5	Treatment flow rate requirement	Intermittent drain: <u>More than 0.7</u> <u>m3/day</u> Flow rate to enable treatment at 80% equipment availability	1.4μm MF membrane: <u>More than 0.2</u> <u>m3/day</u> 0.1μm UF membrane: More than 0.063 m3/day 0.05μm UF membrane: <u>More than 0.4</u> <u>m3/day</u> Flow rate to enable treatment at 80% equipment availability	More than 14 m3/day Flow rate to enable treatment at 80% equipment availability	
6	Operation method	Batch treatment Assuming to complete treatment of 1 batch in one to several days			

Table System requirements of each sedimentation separator



7. Implementation items ② Development of RO concentrated water treatment technology

[Issue] Amount of coagulating sediment (hereinafter sedimentation sludge) generated with the sedimentation separation tank is large.

When assuming the sludge container container is the unit can size $(\varphi 200 \times H400 \text{mm})$, the necessary number of times of discharge is estimated as shown in the below table.

Table Number of times of discharge of sludge collection container

Liquid waste to be treated	Quantity of collection container	Generated amount of sedimentation sludge
Roughly removed liquid waste	1 container/day	3m3/year
Final treatment liquid waste	<10 containers/day	<30m3/year
RO concentrated water	<120 containers/day	<300m3/year

Sedimentation sludge properties

- Generated amount is approx. 10-15 vol% of initial liquid volume
- Moisture content is around 90%
- High liquidity



Filter liquid waste coagulation-

RO concentrated water treatment test *Magnetite 1000ppm

sedimentation test *SiO₂ 10000ppm

Figure Appearance of liquid waste after coagulation-sedimentation treatment

- The amount of sedimentation sludge generated from the sedimentation separation tank for filter liquid waste treatment is relatively small, and it is inferred that the number of times of discharge is an order applicable to actual equipment
- The amount of sedimentation sludge generated from the sedimentation separation tank for RO concentrated water treatment is massive, and the number of times of discharge needs to be revised as it is also massive

Items to be studied

- ① Selection of adsorbents with higher removal performance
- 2 Study of optimization of addition amount of powder adsorbent
- ③ Study of optimization of addition amount of coagulant
- (4) Re-examination of sedimentation separation treatment procedures and equipment
- 5 Study of volume reduction treatment of sedimentation sludge due to dehydration

①-④ ⇒ Study with element tests
 ⑤ ⇒ Implement element test after literature review and desktop study

*Study in item 3

② Development of RO concentrated water treatment technology

Adsorbent test

[Purpose] Select adsorbents applicable to RO concentrated water treatment [Implementation items] Evaluate adsorption performance of powder adsorbents with stirring-type batch test

Study of test system

In general, immersion-type batch test is conducted when evaluating adsorption performance of adsorbents. However, since stirring treatment is performed for RO concentrated water in the sedimentation tank, the test system^{*1} simulates the system of actual equipment.

Selection of powder adsorbent

Adsorbents to be used in this test were selected as shown in the following table, with reference to actual usage at 1F and adsorption rate data acquired through tests up to FY2020, based on literature review. Adsorbents reported to have high adsorption performance for α nuclides to be removed (Pu, Am, etc.) were selected.

No.	o. Adsorbent type Particle size (average)		Remarks	
1	Titanic acid	4.08µm	Actual usage at the US Savannah River Site (batch treatment)	
2	Titanium silicate	8.55µm	Actual use at Fukushima Daiichi	
3	Active carbon	48.5µm	Actual use at Fukushima Daiichi	
4	Hematite	1.32µm	Iron oxide adsorbent. Has been reported of high Kd for α nuclides suc as Pu.	
5	Magnetite	1.96µm	Iron oxide adsorbent similar to hematite.	

Table Result of selection of powder adsorbent*2



Figure State of adsorbent test (titanic acid)



Figure Image of stirring-type adsorption test

- *1 Since hot test is difficult with this test method, all tests are conducted as cold test.
 For α nuclides (Pu, etc.) that are difficult to simulate with the cold tracer, item ① soluble adsorption rate data was used as reference.
 - *2 Selected from α nuclide adsorbents.

It was also selected from adsorbents that cannot take granular form and is difficult to fill adsorption towers.





7. Implementation items

② Development of RO concentrated water treatment technology

Coagulant test

[Purpose] Select coagulants applicable to RO concentrated water treatment [Implementation items] Conduct coagulation-sedimentation test using simulated liquid added with powder adsorbent





Figure Appearance of coagulant test equipment

*1 Tracer removal rate is not evaluated, since whether coagulation sedimentation of powder adsorbents can be performed is the main item to be confirmed. Thus, tracers have not been added.

*2 Coagulants selected in this test were all acidic reagents, and since it needs to be adjusted to the neutral range after addition, sodium hydroxide solution shall be used.

Study of test system

Stirring-type jar test similar to the adsorbent test shall be adopted. A scaled-down system of treatment with an actual coagulation sedimentation tank shall be simulated.

Selection of coagulant

Coagulants were selected as shown in the right table based on results of literature review and FY2020 test results.

In addition to coagulation sedimentation performance, redundancy for ionic strength of liquid waste was also taken into consideration.

Table Result of selection of coagulant

No.	Coagulant	Remarks			
1	High base PAC ^{*3}	High treatment performance has been reported for liquid waste with low ionic strength			
2	Aluminum sulfate	High treatment performance for liquid waste containing boric acid was confirmed in tests of the previous PJ			
*3 PAC: Polyaluminum chloride					

7. Implementation items

② Development of RO concentrated water treatment technology

Result of selection of adsorbent and coagulant

Reagents were selected based on the results of adsorbent test and coagulant test. Reagents for the RO concentrated water treatment test were set based on the below selection result.

Powder adsorbent

Titanium silicate, impregnated active carbon, (titanic acid)

Powder adsorbents to be tests shall be titanium silicate and active carbon among adsorbents that were evaluated to have high α nuclide removal performance. One of each type was selected from adsorbents for selective α nuclide removal and adsorbents for gross activity removal.

*High α nuclide removal performance was also confirmed with titanic acid, but it is not used in this test since the sample amount usable for this test is small.

Coagulant

Aluminum sulfate, (high base PAC)

Based on the coagulant test result, aluminum sulfate shall be selected which showed good results with redundancy to water and relatively low amount of generated amount of coagulating sediments.

Tests using high base PAC were also conducted only under representative conditions.



Figure Distribution coefficient in adsorbent test result



Figure Sedimentation sludge volume in adsorbent test result

(2) Development of RO concentrated water treatment technology

Selection of powder adsorbent and coagulant

Addition amount (concentration) of each powder adsorbent and coagulant was set as follows.

Powder adsorbent

Table Evaluation of required addition amount of powder adsorbent

		Distributi	Distribution coefficient [ml/g]				
No	Adsorbent	Cs	Sr	Eu (Am)	Additive concentrati on		
1	Titonia agid	0.2 E 102	105,00		-50ppm		
I	manic acid	9.2.0+03	1.0.2+06	6.U.E+06	<200ppm		
2	Titanium silicate	1.6.E+06	1.3.E+05	1.1.E+06	50ppm		
3	Impregnated active carbon	1.2.E+03	2.6.E+02	5.9.E+04	200ppm		

Coagulant

Table Evaluation of required addition amount of coagulant

No		Coagulant additive concentration [ppm]				
	Adsorbent	Titanic acid	Titanium silicate	Active carbon		
1	High base PAC	100	1000	1000		
2	Aluminum sulfate	20	200	50		

Setting of adsorbent additive concentration

Value that satisfied DF100 was set based on the distribution coefficient obtained from the adsorbent test. For the convenience of the test, margin that does not fall under the lower limit of detection was considered for analysis.

Setting of coagulant additive concentration

In the coagulant test, additive concentration that confirmed that SS concentration of supernatant can be reduced below 20ppm was set.



7. Implementation items

(2) Development of RO concentrated water treatment technology

Proposed RO concentrated water treatment procedures



(1) Adsorption treatment

Adsorb dissolved nuclides by adding powder adsorbent and stirring for a certain period of time.

(2) Concentration treatment

Concentrate solid components of liquid waste after adsorption treatment, with the cross flow filter (CF). *Repeat the process of reception of RO concentrated water \rightarrow (1) \rightarrow (2) to secure a certain amount of CF concentrate.

(3) Coagulation-sedimentation treatment

Perform coagulation sedimentation of powder adsorbents by adding coagulant to CF concentrate, adjusting to the neutral range with pH adjustment reagent and stirring for a certain period of time. (generation of RO concentrated water sludge)

(4) Delivery of supernatant and sedimentation sludge Supernatant: Residual particles filtered and removed after extraction.

Sedimentation sludge: Collected with sludge collection container.

(5) Sludge dehydration treatment

Perform dehydration treatment on sedimentation sludge with the cartridge filter. Reduce moisture content to around 60%.



② Development of RO concentrated water treatment technology

RO concentrated water treatment test

Treatment test of simulated liquid of RO concentrated water was conducted using sedimentation separation tank. Test conditions were set based on the beaker-scale test result. This test confirmed feasibility of the developed RO concentrated water treatment method.

Simulated target process

Powder adsorption additive concentration was set in two patterns, low concentration and high concentration, considering whether concentration treatment is performed.



Data on nuclide removal performance with this treatment method as well as operability of each mechanism of the sedimentation separation tank and treatment time required for each process shall be acquired to be incorporated in the study of operation of actual equipment.



7. Implementation items ② Development of RO concentrated water treatment technology

> Setting of test parameters

Parameters of the RO concentrated water treatment test were set as shown in the below table.

Table Test parameters in RO concentrated water treatment result

No.48

No.	o. Item Setting		etting	Study policy	
		Nuclide	Cs	0.1 ppm	Cs, Sr and Am are selected as representative nuclides contained in circulation cooling water. Tracer concentration is the initial concentration that can evaluate DF100 (lower limit of detection: 1ppb).
1	S	Nuclide دoncentration	Sr	0.1 ppm	However, Eu is set to 0.5ppm based on evaluation with pre-tests, since it has low stability unless it has certain high concentration and significantly decreases due to precipitation.
	ditior	(tracer concentration)	Eu (Am)	0.5 ppm	Since Am is an α nuclide that is difficult to be handled, tests are conducted using Eu as a substitute due to its chemical similarity.
2	ty cor	Simulated liquidity	Equivalent to 10 seawater	00 times diluted	Chlorine concentration equivalent to accumulated water in PCV was set.
3	r quali	рН	7 (neutral range)	Initial value is neutral. In pH adjustment in coagulation-sedimentation treatment, pH is adjusted to pH recommended for each coagulant.
4	Wate	Water temperature	25°C		Water temperature is expected to be around 10-40°C with actual equipment due to seasonal fluctuations, but since the preliminary test of the previous subsidy project confirmed that temperature dependence is small, tests are conducted at 25° C normal temperature as a representative condition.
5	vder rbent itions	Туре	 Titanium silicate (8.55µm) Active carbon (48.5µm) 		Adsorbents evaluated to have high α nuclide removal performance based on adsorption tests are selected. Particle size is measured with the laser diffraction method. *Titanic acid is not studied as only a small amount of samples for testing could be collected and a large amount of industrial samples could not be collected. There is actual industrial usage at overseas sites.
6	Pov adso cond	Additive concentration	50-100ppm (no concentration treatment), 10000ppm (with concentration treatment)		Two conditions of low concentration and high concentration were set, assuming two patterns of case when implementing concentration treatment with filter filtering and case without such concentration treatment.
7		Туре	 High base PAC Aluminum sulfate 		Selected from the viewpoints of coagulation performance for target particles and redundancy for ionic strength. Since there is concern of organic coagulants having an adverse effect on the treatment disposal process if coagulating sediments contain organic materials, inorganic coagulants are selected to be studied.
8	agulant	Additive concentration	20-100ppm (no concentration treatment), <5000ppm (with concentration treatment) Strong stirring: 157rpm/5min Weak stirring: 52rpm/15min		For liquid waste with SS concentration around 10000ppm, coagulant concentration of around 2000-5000ppm is set as it was evaluated to require such value based on additional test results.
9	ပိ	Stirring speed/time			Necessary stirring speed to achieve stirring capacity obtained under stirring conditions with 500mL beaker- scale tests with the equipment scale (600L) is converted, evaluated and set. Two-stage paddle type structure is adopted for the stirring blade.
10		Settling time	60min-24h		Set at 24h at maximum since 1 batch treatment in 1 day is the basis in the study
1	R				

② Development of RO concentrated water treatment technology

Adsorption performance assessment result

Test conditions

Test solution: 1000 times diluted seawater, pH: 7, tracer: 0.1ppm (Cs, Sr), 0.5ppm (Eu), liquid volume: 600L, stirring condition: 157rpm × 10-120min

Coagulant: Aluminum sulfate (200ppm/50ppm), adsorbent: titanium silicate (50ppm), impregnated active carbon (200ppm)



Figure Tracer in adsorption performance assessment result (removal rate)

- Titanium silicate can remove Cs, Sr and Eu, and is superior in reducing gross activity concentration.
- Impregnated active carbon does not remove Cs and Sr but can selectively remove Eu, and it is superior in reducing only α activity concentration.
- Both tracers have fast adsorption speed, and are considered possible to satisfy the prescribed removal performance (DF100) in a very short treatment time. However, the adsorption test result shows that adsorption speed tends to slow down as addition amount decreases, and a trend with longer required treatment time was confirmed.
- Coagulation-sedimentation treatment did not show any influence such as breakaway of tracers adsorbed to adsorbent particles.

Removal performance and appropriate addition amount of adsorbents are observed from the viewpoints of equilibrium adsorption and adsorption isotherm.



7. Implementation items

② Development of RO concentrated water treatment technology

> Equilibrium adsorption evaluation

Eu equilibrium adsorption of each adsorbent was evaluated based on the adsorption test result. Adsorption amount at 120 min was used, assuming that adsorption equilibrium was reached at 120 min.



Figure additive concentration dependence of Eu equilibrium adsorption

- Equilibrium adsorption of each adsorbent is considered to be in the range indicated in red. However, it is possible that low values were calculated in this test due to dependence on adsorption speed and additive concentration.
- Equilibrium adsorption showed little dependence on adsorbent concentration, and it was suggested that it may be nearly constant in a wide concentration range.
- Evaluation result of adsorption isotherm indicated the possibility of adsorption performance being maintained even when nuclide concentration changes.

It was suggested that saturated adsorption of each adsorbent may be around the same level as equilibrium adsorption confirmed in this test result.



② Development of RO concentrated water treatment technology

Summary of adsorption performance assessment result

Eu removal performance of each adsorbent was evaluated as shown in the below table, based on this test result.

Adsorbent	Eu distribution coefficient ^{*1}	Necessary additive concentration ^{*2}	Eu equilibrium adsorption	Treatment time ^{*3}
Titanium silicate	8.4E+04-3.8E+06ml/g	26mg/L (<50 mg/L)	10-8.0mg/g	<30 min
Impregnated active carbon	2.7E+03-5.3E+05ml/g	190mg/L (<200mg/L)	2.5-2.0mg/g	<30 min

Table Evaluation result of Eu removal performance of each adsorbent

*1 Evaluation result in cases when lower limit may not have reached adsorption equilibrium.

*2 Evaluation using Kd on the upper limit side

*3 However, longer treatment time is required as adsorbent addition amount becomes smaller.

- Eu distribution coefficient was evaluated as 1.0+E04-06 for titanium silicate, and 1.0+E03-05 for impregnated active carbon. According to Eu distribution coefficient, it was evaluated that addition amount of more than 50ppm for titanium silicate and 200ppm for impregnated active carbon is required to meet DF100.
- Additionally, if adsorbent addition amount is sufficiently large, adsorption speed is extremely fast, and it is thought to meet or exceed DF100 in a short treatment time. In this case, the <u>required adsorption treatment time</u> is considered to be within 30min.
- However, preliminary tests found that the time required to reach adsorption equilibrium tends to be longer if adsorbent additive concentration is smaller. Regarding impregnated active carbon in particular, it was suggested that treatment time of more than 120min. may be required if additive concentration is small.

*Since powder materials with small particle size were used, it is considered that surface area is large and adsorption speed is fast.

• It was suggested that equilibrium adsorption may be nearly constant within the concentration range in this test condition. However, dilute concentration range equivalent to actual liquid needs to be separately evaluated since equilibrium adsorption may decrease.



No.52

2 Development of RO concentrated water treatment technology

Coagulant performance assessment result (low SS concentration condition) \triangleright

Here is the coagulant performance assessment result when coagulation-sedimentation treatment is performed for liquid waste after adsorption treatment.

Test conditions (common)

Test solution: 1000 times diluted seawater, pH: 7, tracer: 0.1ppm (Cs, Sr), 0.5ppm (Eu), liquid volume: 600L, stirring condition: 157rpm × 10-120min



*Similar trend as that confirmed in beaker-scale test

- It was confirmed that concentration of liquid, whose initial SS concentration of adsorbent was 50-200ppm, was reduced to 10-٠ 20ppm with treatment using coagulants. Based on the reduction rate of SS concentration, it was suggested that several ppm of particles may remain in the supernatant after treatment.
- If SS concentration requires more than DF100 (removal rate 99%), it is necessary to remove residual particles in supernatant ٠ with filtration.
- It was confirmed that sludge (sediment) generated from coagulation-sedimentation treatment has more than 90% moisture content and its generated amount is around 0.1-0.4%. Since there is much water content, it is thought that sludge amount has increased.



2 Development of RO concentrated water treatment technology

Coagulant performance assessment result (high SS concentration condition)

Here is the coagulant performance assessment result when coagulation-sedimentation treatment is performed for liquid waste that had high SS concentration after CF concentration treatment.

Test conditions (common) Test solution: 1000 times diluted seawater, pH: 7, tracer: not added, liquid volume: 600L, stirring condition: 157rpm×10-120min



*Similar trend as that confirmed in beaker-scale test

- <u>It was confirmed that concentration of liquid, whose initial SS concentration of adsorbent was 10000ppm, was reduced to around 20ppm with treatment using coagulants.</u> Sedimentation behavior immediately after starting treatment is especially noticeable, which decreased below 50ppm within 60min. Thereafter, sedimentation behavior hit the ceiling, and it was confirmed to asymptotically approach around 10ppm.
- Similar to the test with low SS concentration conditions, it was confirmed that particles of around 10-20ppm remain in supernatant. It was suggested that it may be difficult to reduce SS concentration below several ppm with coagulants independent of initial SS concentration.
- It was confirmed that sludge (sediment) generated from coagulation-sedimentation treatment has more than 90% moisture content and its
 generated amount is around 4%. It is considered that moisture content is slightly lower than that of sludge generated from treatment under low
 SS conditions due to the consolidation effect.



2 Development of RO concentrated water treatment technology

Observations on removal performance of coagulants

① SS concentration may be reduced to approx. 10ppm independent of initial SS concentration

② Sedimentation performance of coagulants stabilizes as SS concentration becomes higher, and appropriate treatment is possible within a somewhat wide range of additive concentration.



③ Amount of sludge generated from coagulation-sedimentation treatment tends to increase due to its extremely high moisture



considered more reasonable.

Coagulation-sedimentation treatment using coagulant may reduce sludge moisture content, but 80% is considered to be the lower limit, and it is expected to be accompanied by some amount of supernatant during discharge.

Necessity of sludge dehydration treatment is high.

(Developed in development item ③ (Development of secondary waste treatment technology))

It is considered necessary to implement sludge dehydration treatment to reduce its volume.



(2) Development of RO concentrated water treatment technology

Observations on removal performance of coagulants

The cause of coagulation-sedimentation treatment resulting in SS concentration converging to around 10-20ppm independent of initial SS concentration was observed.

Since a clear mechanism is unknown, the studied hypotheses is as follows.

When the number of coagulating particles and number of naturally dispersing particles become balanced, growth of agglomerates apparently stops and decrease in SS concentration due to coagulation sedimentation no longer occurs.

Coagulant neutralizes particle surface charges and cancels electrostatic repulsion between particles, which causes coagulation of particles. However, the formed agglomerates are not bound together by strong forces, and <u>certain</u> <u>particles are expected to disperse over time</u>. In order words, coagulation and dispersion occur in parallel, and if coagulation has larger contribution rate, agglomerates grow large and sediment.

Particle coagulation occurs when particles collide with each other in liquid, and the probability of collision of particles depend on the number of particles existing in liquid, or SS concentration. Therefore, contribution rate of coagulation becomes smaller as SS concentration decreases, and <u>it is assumed that coagulation and dispersion eventually</u> becomes balanced (equilibrium state).

The aforementioned state where coagulation and dispersion are balanced is considered to be 10-20ppm which is the lower limit of SS concentration that can be reduced as confirmed in this test.



Figure Image of coagulation and natural dispersion of particles due to coagulant



(2) Development of RO concentrated water treatment technology

> Analysis of properties of residual components after treatment

Assumed properties of supernatant and sedimentation sludge generated after RO concentrated water treatment were organized based on analysis data of each test.

Table Assumed properties of supernatant / sedimentation sludge generated after RO concentrated water treatment

	Component / water quality item	Initial concentration [ppm]	Concentration after treatment [ppm]	Residual rate [%]	Remarks
	CI	17	17	—	Seawater component. Not measured in the test. Almost all amount is assumed to remain in liquid.
	AI	9.21	0.067	0.7%	Coagulant component. Almost all amount is assumed to sediment as aluminum hydroxide, and Al concentration in liquid was less than 0.1ppm.
atant	SO ₄	53	45	85%	Coagulant component. Sulfuric acid components tend to remain in liquid, but concentration decreases by 10-20%.
Superr	Са	2.72	2.32	85%	Concrete leached component. Concentration decreases by 10-20% due to precipitation and coprecipitation, but it hardly changes from initial concentration.
	В	201	199	99%	Boric acid solution component. Almost all amount is assumed to remain in liquid. (In the case of 7000ppmasB, 5% at maximum precipitates; FY2019 element test result)
	SS concentration	<200	<20	<10%	It was confirmed that concentration may be reduced to approx. 20ppm independent of initial SS concentration
	рН	(5-9)	7±0.5	_	It is confirmed that there is no significant change from the pH adjustment range (pH7) after coagulation-sedimentation treatment.
ge	Item	ļ	Assumed properties		Remarks
Sedimentation sludç	Generated amount	In the case of low SS concentration: Approx. 1 vol.% In the case of high SS concentration: Approx. 4 vol.%			It is assumed that around 1 vol.% of sludge is generated against the treatment liquid amount. However, approx. 4 vol.% is assumed for high SS concentration liquid waste.
	Moisture content	90-99% (lower limit approx. 80%)			Many test results show moisture content of 80-90% or higher, independent of treatment methods and water quality conditions, and the same is assumed with actual equipment.
	Components	Powder adsorbent, Al component (coagulant)			Powder adsorbent is dominant, which contains aluminum hydroxide derived from coagulants. Coexisting ions contained in treatment liquid are assumed to remain in the liquid.

Concentration of Ca and B decreases due to precipitation and coprecipitation, but the amount of decrease is small, and almost all amount is delivered to the rear. Additionally, AI components remain in sedimentation sludge and sulfuric acid components remaining in supernatant due to addition of coagulant, which is assumed to remain in liquid at SS concentration of around 20ppm.



② Development of RO concentrated water treatment technology

> Study on delivery of supernatant to existing water treatment facility

Reception requirements of existing water treatment facility were studied based on past records of reception of accumulated water of process main building (PMB).

Reception requirements of existing water treatment facility during the fuel debris retrieval work period is a future issue to be studied

Table Tentatively set values* of reception requirements of existing water treatment facility and assumed properties of supernatant

Item	Set value	Test result	Remarks		
CI	<700 ppm	<20 ppm	If each ion concentration of circulation cooling water is below the assumed water quality condition, it is		
Ca	<50 ppm	2.3 ppm	assumed that concentration in supernatant will not be higher than this item. However, CI concentration needs to be studied since accumulated water with high CI concentration		
Mg	<30 ppm	—	such as torus room water may be treated.		
SO4	<100 ppm	45 ppm	If aluminum sulfate is used, a large portion of sulfate ions remains in supernatant. However, existing water treatment facility has a record of receiving approx. 50ppm of sulfate ions on average and approx 100ppm at maximum.		
В	_	200 ppm	There is no record of being received. Upper limit of environmental release is 10mg/L. Thus, if boric acid water is used, it is necessary to sufficiently reduce concentration with boric acid collection equipment at the front.		
рН	7-8.5	7±0.5	Since pH of supernatant is adjusted to around pH7 in the process of coagulation-sedimentation treatment, this item is assumed to be satisfied.		
SS concentration	<20 ppm	10-20 ppm	Since it is assumed that SS concentration can be reduced to 10-20ppm due to coagulation- sedimentation treatment, reception requirements are assumed to be satisfied under SS concentration criteria. Since particles remaining in supernatant have high activity, removal of DF100 or more is required or particle components. Thus, it is assumed that residual particles need to be removed with UF (Ultra-Filtration) membrane.		
Gross α concentration	<7.4E+0 Bq/L 99% removal rate initial concentratio (after 0.45µm filteri		Set based on maximum α concentration of inlet water of existing water treatment facility. It is organized as a future issue to be studied. It is necessary to evaluate it based on assumed gross α concentration of inlet water as well as nuclide removal performance, addition amount and residual amount of powder adsorbents. Additionally, evaluation is necessary from the viewpoint of public exposure (DF100) and announcement density.		

It is assumed that supernatant after coagulation-sedimentation treatment can be delivered to existing water treatment facility, considering record of treatment at existing water treatment facility. However, it is considered that it is necessary to remove residual particles in supernatant with filtration, based on record of reception of existing water treatment facility with SS concentration and viewpoint of removal of activity adsorbed on adsorbents.



^{*}Set based mainly on FY2018 record of reception of PMB accumulated water

② Development of RO concentrated water treatment technology

Study of RO concentrated water treatment procedures

RO concentrated water treatment method was studied based on the development results.

Frequency of each treatment in the below table was tentatively set to simplify evaluation. It is necessary to set the actual treatment frequency taking overall operability of the system into account.

No.	Treatment procedures	Treatment frequency	Generated slurry/sludge	Generated amount	Sludge content	Remarks
1	Reception of RO concentrated water	_	RO concentrated water	14 m3/day	0%	Almost all solid components were removed with the particle removal equipment at the front, and it is assumed that there is hardly any solid components at the time of RO membrane treatment.
2	Nuclide adsorption treatment (addition of powder adsorbent)	1 batch / 1 day	Adsorption treatment liquid	14 m3/day	0.005% (50ppm)	Based on the test result of this subsidy project, addition amount of powder adsorbent is assumed at 50ppm (0.005%; in the case of titanic acid and titanium silicate).
3	Concentration treatment with cross	1 batch / 1	Permeable liquid	<14m3/day	Minute	Concentration factor is assumed to be 100 times. It is assumed that only a very small amount of particles pass through the cross flow filter.
-	flow filtering	ing day	Cross flow concentrate	0.14 m3/day	Approx. 1% (10000ppm)	It is assumed that sludge amount is concentrated to approx. 1% with cross flow filtering. (concentrated water is generated)
	Coagulation-	tion- ation 1 batch / 1 ent week	Supernatant	<1.0 m3/week	0.002-0.005% (20-50ppm)	Based on the coagulation-sedimentation test result, residual particle concentration is assumed at 20-50ppm. Residual particles are removed with UF membrane.
4	sedimentation treatment		Coagulation- sedimentation sludge	0.1 m3/week	<10wt.%	Coagulation-sedimentation treatment is performed for CF concentrate generated in pretreatment to sludge nuclides. Sludge content is assumed to be less than 10 wt.% and generated amount of sludge is assumed to be less than 10 vol.%.
			Separated water	<0.4 m3/month	Minute	When applying cartridge filter filtering, a very small amount of SS is assumed to be contained in separated water.
5	Sludge dehydration treatment *Item ③	1 batch / 1 month	Dry sludge	0.2 m3/month	Approx. 40wt.%	Sedimentation sludge is dehydrated to the target value of 60wt.% water content (40wt.% sludge amount) by passing water through the cartridge filter. However, water content may become larger depending on the used powder adsorbent, and sludge content will become approx. 20-30wt.%.
6	Long-term storage	_	Dry sludge	0.2 m3/month	Approx. 40wt.%	Apply long-term storage with sludge content greater than 40wt.%. However, stabilization treatment such as solidification needs to be separately studied.



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(2) Development of RO concentrated water treatment technology

Study of RO concentrated water treatment flow

Here is the RO concentrated water treatment flow studied based on the development results.



7. Implementation items ② Development of RO concentrated water treatment technology

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[Results to date]

- As candidate powder adsorbents for α nuclide removal, impregnated active carbon was selected to selectively remove α nuclides and titanic acid and titanium silicate were selected to remove gross activity including α nuclides.
- Aluminum sulfate was selected as the coagulant in order to perform coagulation sedimentation of powder adsorbents. Additionally, data on generated amount and properties of supernatant and sedimentation sludge generated as a result of coagulation-sedimentation treatment was acquired. A system applied with CF concentration treatment was also drafted, based on effectiveness of coagulation-sedimentation treatment toward high SS concentration liquid waste.
- Treatment procedures and treatment process for RO concentrated water were drafted based on test results.
 Additionally, the equipment-scale test confirmed feasibility of the developed treatment procedures.

[Future issues to be studied]

- In this PJ, removal performance was evaluated using cold tracers of Cs, Sr and Eu. Regarding other nuclides to be removed, it is necessary to evaluate whether the drafted treatment methods satisfy the prescribed removal performance (evaluation of removal performance of gross activity and gross α in water quality of actual liquid).
- CF concentration treatment was not subject to development in this PJ, but its system was studied through case studies and literature values. It is a future issue to conduct CF filter tests and evaluate whether concentrate factor satisfies the prescribed required performance.

[Summary]

Methods to sludge and separate nuclides in ROC concentrated water through treatment using adsorbents and coagulants were studied. Reagents were selected through element tests, required addition amount was evaluated, and RO concentrated water treatment procedures were drafted. Feasibility of this treatment method was confirmed, and conceptual study of the treatment process was conducted.



③ Development of secondary waste treatment technology

Study flow





③-1 Development of non-soluble nuclide removal technology

> Water quality conditions of input water of particle removal system

Properties of accumulated water transferred to and treated with particle removal equipment were assumed as shown in the following table.

No.	Item	Conditions	Study status
1	Temperature	Normal temperature	Assuming seasonal fluctuation of 10-40°C.
2	рН	5-9	Assuming neutral range. Basic water quality conditions of liquid phase system
3	lonic strength	Seawater component, boric acid, other	Ion strength is assumed to be low in principle. However, ion strength is assumed to become relatively high when there is intrusion of seawater components and when boric acid water is used.
4	Particle components	Fuel debris, reactor internals, concrete	At present, it is assumed that the three components on the left that have a large amount present in PCV are dominant. However, the respective content ratio is unknown. It is also possible that the components contain a very small amount of deposits (soil, sand, etc.) and precipitates.
5	SS concentration	<100ppm	Based on the processing test of simulated fuel debris conducted in the retrieval method PJ as well as evaluation result of target throughput, SS concentration of particles transitioning from PCV to liquid phase system was evaluated to be below 100ppm.
6	Particle size	0.1-100µm	Particles transitioning from PCV to liquid phase system were evaluated to have particle size smaller than 100µm, based on natural sedimentation speed and pump suction capacity.
7	Particle size distribution	Unknown	Since it is unknown at present, broad distribution of 100-0.1µm is assumed. Possibility of double-peaked distribution was suggested from the simulated fuel debris processing test result of the characterization PJ.
8	Particle form	Spherical, non-spherical	 Nearly spherical particles are generated in the case of heat input processing Flaky and block-like particles are generated in the case of machine processing Tests using spherical particles were conducted in subsidy projects up to FY2020. In this PJ, filter water flow test using non-spherical particles was conducted to evaluate the impact of particle forms.
9	Treatment flow rate	52 m³/day (subsidy project), 22 m³/day (engineering)	Work is carried out at a rate of 10h/day with 2.2m3/h of debris processing work water and 3m3/h of reactor injection water. System configuration studied in engineering only treats debris processing work water.

Table Assumed water quality conditions of input water of particle removal system



③-1 Development of non-soluble nuclide removal technology

Study of particle removal system configuration

Non-soluble α nuclide removal equipment (particle removal system)



Figure Conceptual diagram of particle removal equipment system configuration

Result of selection of candidate particle collector

Applicability of particle collectors of each system were evaluated based on literature review result and test result up to FY2022

Table Result of applicability assessment of candidate particle collectors

No.	System	Particle collector	Filter mesh size	Applicability evaluation	Remarks	
1	Rough removal	Auto strainer	50 µm	High	Long-term operation is possible by recovering differential pressure as appropriate with drain operation	
		Liquid cyclone	-	Medium	It is determined that it is less applicable than auto strainers in terms of removal precision and generated amount of liquid waste	
	Intermediate removal	Auto strainer	5, 10, 20 μm	Low	Removal precision is high, but it is determined that applicability is	
		Metal sintering filter	2, 5, 10, 20 µm	Low	low since increase in differential pressure due to clogging is	
2		Bag filter	3 µm	Medium	significant	
		MF membrane (ceramic filter)*	1.4 µm	Medium - high	Test conducted in this PJ. Result reported in this document.	
3	Final treatment	UF membrane (ceramic filter)*	20, 50, 100 nm	High	It is evaluated that applicability is high. It is necessary to select appropriate aperture.	

*MF membrane: Micro-Filtration, UF membrane: Ultra-Filtration



③-1 Development of non-soluble nuclide removal technology

Development issues of particle removal system

Selection of candidate equipment of intermediate removal system

• Filter performance was evaluated as shown in the following table based on the test result of each filter equipment (previous subsidy project).

System	Particle collector		Filtering precision	Removal efficiency	Replacement frequency (life)	Generated amount of secondary waste	Number of times of drain/backwash	Generated amount of liquid waste
			More than μm	More than %	times/year	kg/year	times/day	m3/year
	Auto strainer	Intermittent drain	50	99	2	11	2.5	167
Rough		Permanent drain	50	99	2	11	Permanent drain	1300
Terrioval	Liquid cyclone		40	80	-	-	Permanent drain	2000
	Auto strainer	Intermittent drain	20	96	2	11	502	33500
Intermediate		Permanent drain	20	96	2	11	118	9200
removal	Metal sintering filter		2	99	18300	146500	25.1	815
	Bag filter		3	99	5	/ 440	-	-
Final treatment	UF membrane	0.1µm	0.1	99	8	314	0.17	5.5
	UF membrane	0.05µm	0.05	99	3	152	0.11	48.2

Table Summary of result of filter performance assessment (result of subsidy project FY2019-2020)

Generated amount of liquid waste and generated amount of secondary waste are evaluated to be extremely large

- All equipment has extremely high particle removal rate, and was evaluated to appropriately remove particles in the collection range of each system.
- In the rough removal system and final treatment system, each filter equipment selected was evaluated to have high applicability to actual equipment as increase in differential pressure due to clogging is gradual.
- <u>All filter equipment selected as intermediate removal equipment shows significant increase in differential pressure due to clogging</u>, and generated amount of backwash liquid waste and generated amount of secondary waste are larger compared to other systems.

Since filter equipment applicable to intermediate removal system has not been selected, it is necessary to select candidate equipment.



③-1 Development of non-soluble nuclide removal technology

> Development issues of particle removal system

Impact assessment of particle form

Here are the particles used for the filter water flow test conducted in the previous subsidy project.



Figure SEM image of simulated particles used in the filter test of the previous subsidy project

- In the previous subsidy project, tests were conducted using simulated powder dominated by nearly spherical particles.
- However, if mechanical methods such as AWJ and boring processing are adopted, <u>generated chip powder is</u> <u>expected to be dominated by flaky and needle-like forms</u>.
- If particles take non-spherical form such as flaky and needle-like forms, there is concern of increase in differential pressure during water flow and impact on differential pressure recovery rate due to backwashing.



It is necessary to evaluate the impact of particle forms on each filter equipment.

In particular, it is necessary to <u>acquire operation data of each filter when passing non-spherical particles through water</u>, which have not been verified in the previous subsidy project.



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③-1 Development of non-soluble nuclide removal technology

- Details of element test (filter water flow test)
- Evaluation of applicability of MF membrane to intermediate removal system

Applicability of 1.4µm MF membrane to actual equipment shall be evaluated as intermediate removal filter which was not selected in the previous subsidy project.

1.4µm MF membrane water flow test

The following items shall be evaluated by performing water flow treatment for simulated liquid of assumed input water quality of intermediate removal system with 1.4µm membrane.

- Collection performance for particles in the range of several dozen to several µm which is the collection range of intermediate removal system
- Trend of increase in equipment differential pressure during water flow and time available for water flow
- Trend of recovery of differential pressure due to backwashing when differential pressure increases

Evaluation of dependence of particle form of each filter

Impact on each filter when particle form changes shall be evaluated, which was not evaluated in the previous subsidy project.

Evaluation test of dependence of particle form

Data, such as filter operation data, differential pressure behavior, particle collection rate and recovery rate of differential pressure due to backwashing, when passing spherical particles and non-spherical particles through water, shall be acquired.



*Filters to be tested were selected among filters that were evaluated to have high applicability in each system.

According to filter operation data acquired from the test, resistance of cake formed by spherical/non-spherical particles shall be evaluated and impact on filters attributable to particle form shall be evaluated.



③-1 Development of non-soluble nuclide removal technology

> Test system of MF membrane water flow test





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Housing

*7 filter elements were loaded in the test.

Test case of MF membrane water flow test

Test Case		Particle size [µm]	Flow rate [m ³ /h]	SS concentration [ppm]	Particle components mixing ratio [wt.%]			Water flow time
					WC	SUS316	SiO ₂	
	1-1	0.1-10		100	33	33	33	Until allowable differential pressure is reached
	1-2	0.1-10	10	100	58	30	12	
	1-3	0.1-10	10	500	33	33	33	
	1-4	1		100	33	33	33	

Table MF membrane water flow test case table

Figure Test system configuration diagram of MF membrane water flow test



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7. Implementation items ③-1 Development of non-soluble nuclide removal technology

MF membrane performance assessment parameter

Test parameters were set as shown in the following table based on assumed water quality condition.

No.	Item	Conditions	Remarks			
1	Particle collector	1.4µm MF membrane (ceramic filter)	Candidate equipment of intermediate removal system is selected			
2	SS concentration	100ppm, 500ppm	100ppm, which is the assumed condition for the liquid phase system, is set as the basic condition, and 500ppm is set as the high load condition			
3	Particle size	0.1-10µm, 1µm	$0.1\text{-}10\mu m,$ which is the range of particle size that could flow into the intermediate removal system, is set as the reference condition, and only $1\mu m$ particles are set as the high load condition			
4	Particle form Spherical particle		Spherical particles are used so that they can be studied in comparison with tests using spherical particles conducted up to FY2020			
5	Particle components (to be simulated)	Tungsten carbide (fuel debris), SUS316L (reactor internals), silica sand (concrete component)	Particles with true specific gravity close to their respective simulated components are selected			
6	Particle mixing ratio	 Equal mixing of three components on a weight basis Equal mixing of three components on a volume basis 	 ① is set so that it can be studied in comparison with tests conducted up to FY2020 ② is set as the condition with equal number of particles of the three components 			
7	Flow rate	10m ³ /h	10m ³ /h, which is the basic system flow rate of the liquid phase system, is set			
8	Water flow time	Water is passed through until backwash set pressure is reached. If long-term water flow is required, acceleration test is conducted by increasing SS concentration.				

Table MF membrane water flow test parameters

Table MF membrane water flow test backwash conditions

No.	Item	Set value	Remarks
1	Backwash flow rate	10 m3/h	
2	Backwashing pressure	0.10 MPa	Conditions are set upon confirming the appropriate
3	Backwash time	1 min/count	differential pressure recovery trend through tests.
4	Implemented number of times	Until recovering to initial differential pressure	



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③-1 Development of non-soluble nuclide removal technology

Fest result of MF membrane water flow test

Here is the observation on behavior of increase in equipment differential pressure during water flow, using representative examples.



- In test case 1-1 which is the basic condition, the time available for water flow until backwashing was evaluated to be 62min. It is
 evaluated to be highly applicable as increase in differential pressure is slow compared to candidate intermediate removal equipment
 which was tested in the previous subsidy project. The number of elements was set to seven elements in this test, but since water
 flow time until backwashing can be adjusted by adjusting the number of elements, it is desirable to design the number of elements
 to allow operationally appropriate backwash frequency and equipment scale.
- Test case 1-1 has broad particle size distribution, and test case 1-4 has the sharp peak of its particle size distribution at 1µm.
 Differences in differential pressure increase considered to be attributable to differences in particle size distribution were confirmed, and the time available for water flow in the condition where complete blockage is most likely to occur (test case 1-4) is considered to be 2/3 of that of cases with broad distribution.
- If backwashing operation is performed after differential pressure increases due to clogging, initial differential pressure largely recovers, and it has been confirmed that it can be reduced to near its initial value. However, since particles that cannot be discharged slightly remain, two or three additional backwashing is required in order to recover differential pressure to near its initial value. Necessity of complete recovery to initial differential pressure is low in the operation of actual equipment, and it is considered that operationally sufficient recovery rate can be secured with one backwashing.



3-1 Development of non-soluble nuclide removal technology

> Comparison of performance of candidate intermediate removal system filters

Candidate intermediate removal system equipment was compared and evaluated based on the element test result. (including results of subsidy projects prior to FY2020)

Generated Required Number of Generated amount of Filtering Removal Replacement number of times of amount of Applicability precision frequency efficiency secondary drain/backwash liquid waste Particle collector elements assessment waste result More than More than times/year Piece kg/year times/day m3/year % μm Intermittent 1 20 96 2 11 502 33500 Low drain Auto strainer Permanent 20 2 11 118 1 96 9200 Low-medium drain Metal sintering filter 146500 11 2 99 18300 25.1815 Low Bag filter 122 3 99 5 440 Medium **MF** membrane 70 1.4 99 1 77 0.05 30 Hiah

Table Comparison of performance of candidate intermediate removal system equipment

Result of evaluation according to the test result of this PJ was added *Evaluation conditions: treatment flow rate: 65m3/day, SS concentration: 100ppm, particle collection rate: 33% in each system *Replacement frequency of MF membrane was conservatively assumed and set with the service life of sealing materials such as gasket

- <u>It was evaluated that MF membrane has the best performance from the viewpoint of generated amount of secondary waste and generated amount of liquid waste (concentration factor).</u>
- Compared to other candidate equipment, MF membrane increases differential pressure due to particle collection more gradually and can collect more particles before backwash. It is considered to be a major factor that its effective filtering area is extremely large compared to other intermediate removal filters.
- Replacement frequency of ceramic filters such as MF membrane was set based on replacement cycle of sealing materials. It is assumed that deterioration of sealing materials such as gasket is faster than increase in initial differential pressure and deterioration of filter body due to repeating water flow and backwashing.

Based on this evaluation result, <u>1.4µm MF membrane was selected as candidate intermediate removal system equipment.</u>


③-1 Development of non-soluble nuclide removal technology

> Parameters of evaluation test of dependence of particle form

Parameters for this test were set as shown in the following table.

The test system used similar equipment as that of MF membrane water flow test.

No.	ltem	Conditions	Remarks					
1	Particle collector	50μm auto strainer 1.4μm MF membrane 0.05μm UF membrane	Particle collector evaluated to have high applicability in each system is selected *Increase in differential pressure was not observed with auto strainer					
2	SS concentration	100ppm, 500ppm	100ppm, which is the assumed condition for the liquid phase system, is set as the basic condition, and 500ppm is set as the high load condition					
3	Particle size	0.1-10µm, 1µm	0.1-10 μ m, which is the range of particle size that could flow into the intermediate removal system, is set as the reference condition, and only 1 μ m particles are set as the high load condition					
4	Particle form	Spherical particle, non-spherical particle	Spherical particles and non-spherical particles are selected Impact of particle form on filter performance is evaluated based on the test result of both forms.					
5	Particle size distribution	Double-peak distribution of 1µm, 10µm	Set with reference to cutting test of simulated fuel debris conducted in the characterization PJ. *See next page					
6	Particle components (to be simulated)	Silica sand (concrete component)	Silica sand is selected as the representative component. With other component particles, it is difficult to produce non-spherical fine particles.					
7	Flow rate	1m³/h, 10m³/h	10m ³ /h, which is the system flow rate of the liquid phase system, is set \rightarrow UF membrane is tested with 1m ³ /h and evaluated with proportional calculation.					
8	Water flow time	Water is passed through until backwash set pressure is reached.						

Table Test parameters of evaluation test of dependence of particle form

This test evaluates the impact of particle form on filter performance from the viewpoint of differential pressure behavior and particle collection rate.



3-1 Development of non-soluble nuclide removal technology

Simulated particles used in this test \geq

Here are the SEM image and particle size distribution of each particle used in this filter test



Adjustment was made so that there is no difference in particle size distribution of both types. (double-peak distribution) Since other parameters are also the same, it is evaluated that the difference observed in the test result is attributable to particle form.



particles.

Spherical particle

③-1 Development of non-soluble nuclide removal technology

Difference in behavior of differential pressure increase due to forms of particles passed through water

Non-spherical particles have slower increase in equipment differential pressure, but its difference is small.In addition, the difference tends to gradually increase.

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Figure Differential pressure behavior in filter test result

Comparing overall behavior of equipment differential pressure, spherical particles show larger differential pressure than non-spherical particles. In particular, difference starts to occur between the two types of particles approx. 30min. after starting water flow for the MF membrane and approx. 500min. after starting water flow for the UF membrane. The cause was observed as follows.

① Cake formation stage

Stage where particles are collected with filter filtering and cake is formed. Filter resistance is dominant, and impact of cake resistance is minor. Thus, difference in behavior of increase in differential pressure is unlikely to occur due to particle form.

2 Cake filtering stage

Stage where cake is formed and particles passed through water are cake-filtered. Impact of cake resistance appears in addition to filter resistance. Cake resistance differs depending on particle filling density of cake. Spherical particles have larger cake resistance since its filling density is large. Therefore, differential pressure is larger compared to the case when passing non-spherical particles through water. The difference becomes larger as cake grows thicker with longer water flow time.



7. Implementation items 3-1 Development of non-soluble nuclide removal technology

> Difference in backwash performance due to forms of particles passed through water

Differential pressure recovery behavior due to backwash operation when differential pressure increases by passing spherical/non-spherical particles through water was compared and evaluated.





Observation on backwash performance

- It is evaluated that <u>backwash performance is extremely high regardless of particle form</u>, as differential pressure was recovered close to initial differential pressure due to backwash for cases of passing both spherical and non-spherical particles through water.
- Comparative evaluation shows a trend of lower differential pressure after backwash when using non-spherical particles, but the difference between the two types of particles is minute and it is unclear whether the difference is attributable to particle form.

In general, backwash performance is known to improve when passing non-spherical particles that are close to fibrous form through water, rather than spherical particles, as particles can be easily discharged by backwashing. The effect is the same when particles of several µm order is dominant as in this test, but contribution is expected to decrease as the particle size decreases.

Regarding differential pressure recovery behavior during backwashing, difference attributable to particle form is minor, and almost all particles are expected to be discharged by backwashing.

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③-1 Development of non-soluble nuclide removal technology

Evaluation using Lewis' Equation of Filtration

Dependence of filtering performance on particle form was evaluated with average cake resistivity.

Average cake resistivity $\alpha av[m/kg]$

Physical quantity that indicates the resistance when fluid passes through cake that was formed on the filter surface due to filtering. In general, it is used to determine filtering performance of the filter to be used for liquid subject to filtering.

Reference judgment value [m/kg]

 $\alpha_{av} \le 10^{11}$: Easy $10^{12} \le \alpha_{av} \le 10^{13}$: Medium $10^{13} \le \alpha_{av}$: Difficult

*Judgment value is criteria based on general experience

Table Evaluation result of average cake resistivity / average filtering resistivity (comparative evaluation of representative cases)

Test case	2-1	2-2	3-1	3-2
Filter type	0.05µm UF membrane	0.05µm UF membrane	1.4µm MF membrane	1.4µm MF membrane
Particle form	Spherical	Non-spherical	Spherical	Non-spherical
Particle collection rate [%]	99.9	99.9	69.7	71.5
Average cake resistivity [m/kg]	7.9.E+12	6.9.E+12	1.4.E+12	1.2.E+12
Filter material resistance [1/m]	-3.0.E+11	-2.2.E+11	6.4.E+10	5.1.E+10
/				

*Phenomenologically, it cannot be a negative value, and so it is actually considered to be close to 0. ⇒ When liquid not containing particles passes through water, equipment differential pressure hardly increases.

- In this test, average cake resistivity was evaluated as 1.0E+12 order with UF membrane and 1.0E+11 order with MF membrane, regardless of spherical or non-spherical particles.
- As comparative evaluation indicates that <u>average cake resistivity tends to be larger with spherical particles but the difference</u> <u>is small</u>, it is considered that <u>cakes with similar properties were formed</u>.
- Since it is assumed that particles have agglomerated to some extent and formed agglomerates of several µm or greater, it is possible that dependence on particle form was small.
- When treating liquid containing large amounts of particles with larger particle size (several mm order), dependence on particle form is considered to be large. However, since cake resistance becomes smaller due to increase in porosity of cakes as particle size becomes larger, and most particles larger than several mm are assumed to settle in PCV, it is considered less necessary to evaluate the impact of particle form on filter performance when filtering large particles.

Based on this test result, it was judged that it is less necessary to design the particle removal system considering variations in particle form.



③-1 Development of non-soluble nuclide removal technology

> Evaluation of impact of each parameter on particle removal system

Table Evaluation of impact of each parameter on particle removal system

No.	ltem	Degree of impact on particle removal system	Test results	Details of study
1	Particle components	Auto strainer: high Ceramic filter: low	Highly feasible	Since auto strainer is a mechanism that separates particles also with centrifugal separation in addition to filtration, separation performance is high for particles with higher specific gravity such as fuel debris. However, centrifugal separation hardly has any removal effect for particles with low specific gravity such as concrete particles.
2	SS concentration	High	○Highly feasible	Since there is a significant impact on the speed of increase in differential pressure of each filter, it largely affects the concentration factor.
3	Particle size range	High	oHighly feasible	Particle size is considered to rate-control the percentage of the amount of particles collected in each
4	Particle size distribution	High	○Highly feasible	amount of particles in the particle collection range of each system.
5	Particle form	Low	○Highly feasible	Impact of particle form was evaluated to be small according to the filter test using spherical/non-spherical particles.
6	Particle strength	Low	×Low feasibility	Since particles generated in the fuel debris retrieval work are assumed to be basically solid materials with a certain degree of hardness, it was evaluated that the impact is small.
7	Precipitate	Medium	×Low feasibility	Generation of gel-like precipitates containing much moisture causes significant clogging. Impact is small if precipitates are highly crystalline.
8	lonic strength	Low	△Some may not be feasible	If ion strength is high, apparent grain size becomes larger due to saltation effect. With water quality considered in actual equipment, it is assumed that only almost negligible coagulation occurs.
9	System configuration	High	○Highly feasible	Concentration factor of the entire system can be improved by changing the type, aperture and filtering area of the filter to be installed, according to particle size distribution. Currently, a system that can handle a wide range of conditions is being studied.
10	Removal range (lower limit)	Medium	 Highly feasible 	Impact is small if the required value of filtering precision is up to around 0.1µm. Impact such as increase in generated amount of liquid waste and increase in equipment scale becomes apparent from around 0.01µm.
11	Concentration reduction request	Medium	\triangle Some may not be feasible	Impact is large since larger removal rate is required with larger reduction requirement of activity concentration.

Performance of particle removal system was evaluated using physical quantity with large impact (in red) as parameters.



3-1 Development of non-soluble nuclide removal technology

Evaluation of generated amount of liquid waste of particle removal system

The amount of filter liquid waste generated during the fuel debris retrieval period was estimated from the filter test result.

Equation for evaluation of generated amount of filter liquid waste

$$V_{bw}(c_{in}, a_n) = \sum_{n=1}^{3} \left(\frac{V_{in} \times v_n \times a_n \times c_{in}}{m_n} \right)$$

V_{bw} : Annual amount of generated liquid waste [m3/year]

- *V_{in}*: Annual treatment liquid amount [m3/year]
- c_{in} : SS concentration of input water [kg/m3]
- v_n : Amount of backwash water per treatment of each filter equipment [m3/count]
- a_n : Particle collection rate of each filter equipment [%] (particle size distribution)
- m_n : Particle collection amount up to backwashing of each filter equipment [kg/count]

lat	ble	Set con	ditions	of part	ticle col	llector	and	numl	ber of	elem	ents
											-

System	Particle collector	Loaded number of elements
Rough removal	50µm auto strainer	1 element
Intermediate removal	1.4µm MF membrane	70 elements ^{*1}
Final treatment	0.05µm UF membrane	70 elements ^{*1}

*1 Number of elements for which backwash frequency is less than once a day and the filter line velocity is sufficiently small

- · Above parameters are set using studied values of the liquid system and element test result as boundary conditions
- SS concentration c_{in} and particle collection rate a_n (particle size distribution) are set as variables and several patterns of water quality conditions of input water are assumed

Basic model of this evaluation (particle removal system)





Evaluation result of generated amount of liquid waste

The generated amount of liquid waste when liquid waste with the assumed water quality is treated was evaluated for each particle removal system configuration.

: 1.0 m3/year
: 100 m3/year
: 1000 m3/year

Table Evaluation result of generated amount of liquid waste of particle removal system [m3/year]

Particle size range [µm]		3-system configuration			2-system configuration			1-system configuration									
Rough removal	Intermediate removal	Final treatment	S	S conc	centratio	on [ppn	า]	:	SS con	concentration [ppm]			SS concentration [ppm]				
50~100	1~50	0.1~1	1	10	50	100	200	1	10	50	100	200	1	10	50	100	200
33%	33%	33%	3	27	135	271	542	3	33	165	330	660	2	25	123	246	492
90%	5%	5%	5	46	232	464	927	5	47	236	472	945	2	25	123	246	492
80%	10%	10%	4	43	215	430	859	4	45	224	447	894	2	25	123	246	492
5%	90%	5%	1	10	50	99	198	3	26	129	258	517	2	25	123	246	492
10%	80%	10%	1	13	65	130	259	3	27	136	271	542	2	25	123	246	492
5%	5%	90%	2	25	125	250	499	3	26	129	258	517	2	25	123	246	492
10%	10%	80%	3	25	127	253	507	3	27	136	271	542	2	25	123	246	492

*Effect of centrifugal separation of auto strainer is not considered in this evaluation. Qualitatively, centrifugal separation effect is larger for larger particles and particles with higher specific gravity.

Appropriate particle removal system configuration differs according to properties of input water. Appropriate system configuration was studied for each assumed condition.



3-1 Development of non-soluble nuclide removal technology

Basic system configuration of particle removal system



7. Implementation items ③-1 Development of non-soluble nuclide removal technology

Summary of performance assessment result of each particle collector

Assumed specifications of candidate particle collectors in each system were evaluated based on test results acquired to date.

System	Particle collector		Required number of units	Required number of elements	Rated flow rate	Filtering precision	Removal efficiency	Approximate dimension (per unit)	Required area dimension (per unit)	Replacement frequency (life)	Generated amount of secondary waste	Number of times of drain/backwash	Generated amount of liquid waste
			1 system per unit	Piece	m3/h	More than µm	More than %	φm×mH	mL×mW	times/year	kg/year	times/day	m3/year
Rough removal	Auto strainer	Intermittent drain	1	1	10-30	50	99	φ0.5m×1.0mH	1.4mL×1.4mW	2	11	2.5	167
		Permanent drain	1	1	10-30	50	99	φ0.5m×1.0mH	1.4mL×1.4mW	2	11	Permanent drain	1300
	Liquid cyclone		1	1	10	40	80	φ0.4m×1.1mH	1.6mL×1.6mW	-	-	Permanent drain	2000
	Auto strainer	Intermittent drain	1	1	10-30	20	96	φ0.5m×1.0mH	1.4mL×1.4mW	2	11	502	33500
ntermediate		Permanent drain	1	1	10-30	20	96	φ0.5m×1.0mH	1.4mL×1.4mW	2	11	118	9200
removal	Metal sinte	ering filter	3	11	6.5	2	99	φ0.21m×1.2mH	3.0mL×1.6mW	18300	146500	25.1	815
	Bag f	filter	4	122	10	3	99	φ0.7m×1.6mH	2.0mL×2.0mW	5	440	-	-
	MF men	nbrane	4	70	10	1.4	99	φ0.3m × 1.4mH	1.4mL × 1.4mW	1	77	0.05	30
Final reatment	UF membrane	0.1µm	2	33	6.5	0.1	99	φ0.21m×1.3mH	3.0mL×1.6mW	8	314	0.17	5.5
	UF membrane	0.05µm	3	70	10	0.05	99	φ0.3m×1.4mH	1.4mL×1.4mW	1	77	0.11	50

Table Summary of result of filter performance assessment (3-system water flow configuration)

Future development issues

*Evaluation conditions: treatment flow rate: 52m3/day, SS concentration: 100ppm, particle collection rate: 33% in each system

① Study of filter replacement method and evaluation of replacement cycle (life)

It is necessary to study remote replacement of filter elements and alternative replacement method when remote replacement is difficult. Additionally, filter life data for setting the replacement cycle needs to be evaluated in the future since there is shortage of such data. Assuming ceramic filters are rate-controlled by the replacement cycle based on service life of gasket materials (assuming 1-2 years), rather than replacement cycle attributable to deterioration of the filter body, it shall be considered to assume ceramic filters based on operation results of similar gasket materials of Fukushima Daiichi

2 Study of optimization of loaded number of elements and water flow velocity

Since filter and cake resistance is reduced as line velocity becomes smaller with more loaded number of elements or slower flow velocity, the amount of particles that can be collected up to backwash differential pressure becomes larger. However, because of the tradeoff between increase in equipment scale and prolonged treatment time, it is necessary to optimize treatment conditions such as number of elements and flow velocity taking various factors into account.



7. Implementation items 3-1 Development of non-soluble nuclide removal technology

[Results to date]

- ✓ 1.4µm MF membrane was selected as candidate equipment applicable to the intermediate removal system.
- Dependence of particle form on candidate equipment of each system was verified. As a result, it
 was confirmed that there is little need to incorporate particle form into the conceptual design of
 the particle removal system since impact of particle form is small.
- ✓ 3 patterns of particle removal system configuration, 1-system, 2-system and 3-system configuration, were studied with 3-system configuration as the basis.
- Estimated amount of liquid waste generated from the particle removal system was evaluated, and system configuration considered to be dominant for each input water quality was organized.

[Future issues to be studied]

- ✓ Fuel debris retrieval method shall be selected, and a more reasonable system shall be selected using the studied results when it becomes possible to estimate the properties (generated amount, components, particle size distribution, etc.) of generated chip powder
- Application of filter equipment is also studied for the soluble nuclide removal system and RO concentrated water treatment system, and it is assumed to perform concentration treatment for powder adsorbents with the cross flow method. It is necessary to study the applicability of cross flow filter when assuming such treatment.

[Summary]

Candidate filter equipment applicable to each system of particle removal system was selected, and performance assessment was conducted. Appropriate system was studied according to properties of input water quality with 3-system configuration as the basis.



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③-2 Development of sludge dehydration treatment technology

> Evaluation of volume reduction effect by dehydration

[Implementation items] Volume reduction of sedimentation sludge by dehydration was evaluated.

Behavior of volume reduction when removing moisture from assumed moisture content of sedimentation sludge of approx. 90vol.% was evaluated.



Figure Relation between moisture content and sludge volume

Figure Image of behavior of reduction of sludge volume

If initial moisture content is above 90vol.%, volume can be reduced to nearly 20vol.%, by dehydration treatment up to 60vol.% moisture content.

Since volume decrease hits the ceiling in the range below 60vol.% moisture content, effect of volume reduction due to dehydration is small.

Dehydration treatment technology shall be studied with target moisture content after dehydration treatment tentatively set* to 60vol.%.

*Sludge content 40vol.%



No.83

7. Implementation items

③-2 Development of sludge dehydration treatment technology

Selection of candidate technology

Conceptual flow of the sludge dehydration treatment system was assumed as shown in the following figure. Solid-liquid separation treatment



Figure Conceptual flow of solid-liquid separation system of powder adsorbent

No.	Candidate technology	Candidate applicable treatment	Approach
1	MF, UF membrane (DE) filtering		It is assumed to be applicable as technology that performs concentration treatment for liquid with SS concentration below 100ppm to several thousand ppm.
2	Cross flow (CF) filtering	Concentration treatment of solid components	It is assumed to be applicable as technology that concentrates liquid with SS concentration below 100ppm, to tens of thousands ppm.
3	Coagulation- sedimentation treatment		It is assumed to be applicable as technology that concentrates liquid with SS concentration of several hundred ppm to tens of thousands ppm, to tens of thousands ppm, and makes them into sedimentation sludge. However, residual amount of particles is relatively large, and particle collection rate is low.
4	RO concentration	Secondary nuclide	It is assumed to be applicable for the removal of particles and ionic nuclides that are difficult to be filtered and removed.
5	Evaporator	such as filter permeable water	It is assumed to be applicable for the removal of particles and ionic nuclides that are difficult to be filtered and removed.
6	Cartridge filter	Sludge dehydration	It is assumed that adsorption treatment liquid / concentrate / sedimentation sludge can be filtered with cartridge filter, dehydrated to solid concentration of around 40%, and converted into cake.
7	Depressurization / heatup dehydration	treatment	It is assumed to be applicable as technology that dehydrates concentrate / sedimentation sludge to solid concentration of 40% or higher.

Table Study of treatment method that can be applied with each solid-liquid separation technology

Applicability of each candidate technology to the sludge dehydration treatment system was studied.



③-2 Development of sludge dehydration treatment technology

Study on element test of sludge dehydration technology

Applicability of candidate technology selected based on literature review was verified through element tests.

No.	Required treatment	Candidate technology	Items to be confirmed in element test	Can be dehydrated to 60% of target value	Element test is required
1		MF, UF membrane (DE) filtering	It is necessary to evaluate concentration limit according to slurry property. It is assumed that it can be studied with reference to the filter test result of the previous year.	× Low possibility	× Not necessary
2	Concentration treatment of solid components	Cross flow (CF) filtering	It is necessary to evaluate concentration limit and throughput of treatment amount according to slurry property. It is assumed that it can be studied with reference to cases at overseas nuclear sites, but it may be difficult to evaluate throughput.	× Low possibility	△Necessary with certain conditions
3		Coagulation- sedimentation treatment	Implemented in item $\textcircled{2}$ of this subsidy project	× Low possibility	O (item 2))Necessary
4	Secondary nuclide	RO concentration	Not subject to element test as it was selected as nuclide removal technology	× Low possibility	_
5	such as filter permeable water	Evaporator	Not subject to element test as it was selected as nuclide removal technology	× Low possibility	_
6	Sludge dehydration	Cartridge filter	It is necessary to select appropriate filter elements (aperture, filtering area) and filtering conditions, according to sludge property (particle size distribution, moisture content, etc.).	OPossible	ONecessary
7	treatment	It is assumed that dehydration treatment is possible up to the target value, heatup dehydration / heatup dehydration is possible up to the target value, in the target value, without largely depending on sludge properties (particle size distribution, particle properties).		OPossible	× Not necessary
8	(Solidificatio	on treatment)	Solidification treatment is considered to be effective in terms of stabilization of sludge waste, but final disposal concept of Fukushima Daiichi waste is undetermined. Since it is desirable to conduct solidification treatment according to buried body requirements, it is not studied in this development project, and long-term storage shall be assumed after dehydration treatment.	△Possible with certain conditions	_

Operation data was acquired by conducting element test of cartridge filter.

Applicability of other candidate technologies was studied in desktop study.

IRID

3-2 Development of sludge dehydration treatment technology

Cartridge filter test

Here are the test procedures of the cartridge filter test conducted.

Test condition (test case 1-1)

Test solution: 1000 times diluted seawater, pH: 7.1, adsorbent: titanium silicate, 50ppm, coagulant: not added, 0ppm, Used amount of liquid: 13500mL, filtering pressure: 0.2MPa (pressurized filtering), filtering membrane: 1.2µm membrane filter $\times \phi 47$ mm



This test shall evaluate whether filtering velocity satisfies the target value, acquire data on properties of filtering cake, and evaluate applicability of cartridge filter as sludge dehydration technology.



7. Implementation items ③-2 Development of sludge dehydration treatment technology

Table Organization of slurry/sludge that may be treated with cartridge filter equipment

No.	Assumed slurry/sludge	Concentration of solid components	Particle components	Components	Remarks
1	Adsorption treatment water	Approx. 100ppm		_	Liquid waste generated after adsorption treatment due to powder adsorbent
2	CF concentrated water	Approx. 10000ppm		—	Concentrated water from concentration treatment of No.1 liquid waste with CF filtering
3	Coagulation- sedimentation sludge	<100000ppm(10%)	Powder adsorbent	Coogulant	Sedimentation sludge generated after adding coagulant to No.2 CF concentrated water and performing coagulation- sedimentation treatment
4 (addition)	Coagulation- sedimentation sludge (no preliminary concentration)	<100000ppm(10%)		component	Sedimentation sludge generated after adding coagulant to No.1 adsorption treatment water and performing coagulation-sedimentation treatment

RO concentrated water treatment process proposal

In addition to RO concentrated water sedimentation sludge (No.3) that was initially assumed to be treated, tests were also conducted for slurry/sludge assumed to be highly likely to be treated with the sludge dehydration treatment facility.

No.1 adsorption treatment water





③-2 Development of sludge dehydration treatment technology

Test parameters (setting of conditions of slurry/sludge)

Parameters concerning preparation of slurry/sludge are as follows.

Based on the RO concentrated water treatment test result, titanium silicate and impregnated active carbon were selected as the powder adsorbent and aluminum sulfate was selected as the coagulant.

No	Powder	adsorbent ^{*1}	C	oagulant	Tast solution conditions	s (to be simulated)	
NO.	Туре	Concentration [ppm]	Туре	Concentration [ppm]		(to be simulated)	
1-1		50		—		No.1 adsorption treatment water	
1-2		10000	Aluminum sulfate	_		No.2 CF concentrated water / DE filtering backwash water	
1-3	Titanium silicate	Approx. 100000		5000		No.3 coagulation-sedimentation sludge	
1-4		Approx. 100000	High base PAC ^{*2}	2000	1000 times diluted	No.3 coagulation-sedimentation sludge	
1-5		200		—		No.1 adsorption treatment water	
1-6	Impregnated active	10000	Aluminum sulfate	_		No.2 CF concentrated water / DE filtering backwash water	
1-7	carbon	Approx. 100000		1000		No.3 coagulation-sedimentation sludge	
1-8		Approx. 100000	High base PAC ^{*2}	1000		No.3 coagulation-sedimentation sludge	
1-9 ^{*3}	Titanium silicate	50	High base PAC	500	1000 times diluted	No.4 coagulation-sedimentation	
1-10 ^{*3}	Impregnated active carbon	200	High base PAC	500	seawater	(no preliminary concentration)	

Table Parameters concerning slurry/sludge

*1 High α nuclide removal performance was also confirmed with titanic acid, but it is not used in this test since the collectable sample amount is small. Test result of titanium silicate which has similar particle size and specific gravity is used for the evaluation.

*2 Only basic conditions are applied also for high base PAC

*3 Additional test: Simulation of treated sludge without concentration treatment and with coagulation-sedimentation treatment →It was confirmed that sludge becomes difficult to be filtered due to precipitation of gelatinous materials (explained later).



③-2 Development of sludge dehydration treatment technology

- Applicability assessment of cartridge filter
- Observation of filtering property of each slurry/sludge (average filtering velocity)

Here is the evaluation result of average filtering velocity obtained in each test. Filtering velocity that satisfies the decision

Filtering velocity decision threshold: Average filtering velocity V [ml/min]

Adsorption treatment water: V = 28[ml/min] CF concentrated water: V = 0.56[ml/min] Sedimentation sludge: V = 0.11[ml/min]

*Filtering vellocity that can treat 14m3/day of RO concentrated water

Overall trend of filtering velocity

-Adsorption treatment water \geqq CF concentrated water > sedimentation sludge -Active carbon > titanium silicate



Simulated materia

Adsorption

treatment water CF concentrated

water

Sedimentation

sludae

Sedimentation

sludge Sedimentation

> sludge Adsorption

treatment water CF concentrated

water

Sedimentation

sludge

Sedimentation

sludge Sedimentation

1-1

1-2

1-3

1-4

1-9

1-5

1-6

1-7

1-8

1-10

Adsorbent

Titanium

silicate

Impregnated

active carbon

50ppm

10000ppm

Approx.

Approx.

100000ppm

50ppm

200ppm

10000ppm

Approx.

Approx.

100000ppm

200ppm

10000ppm

100000ppm

Figure Evaluation result of average filtering velocity

- This test confirmed that filtering velocity exceeds the threshold in all tests.
- Even if liquid waste of high SS concentration after CF concentration treatment is treated, decrease in filtering velocity is small compared to liquid waste without concentration treatment (comparative evaluation of 1-1 and 1-2).

threshold is confirmed

- If coagulant is added, filtering velocity tends to become small compared to cases without coagulant addition. This is thought to be because cake thickness increases due to coagulant components. This trend was especially evident for cases treated under low SS concentration conditions, as in additional test 1-9 and 1-10. This is thought to be because filtering membrane is clogged by precipitates due to excessive addition of coagulant.
- Based on the above, the prospect was obtained that sludge dehydration treatment with the cartridge filter is feasible in all treatment patters set.





5000ppm

2000ppm

500ppm

1000ppm

1000ppm

500ppm

_

Aluminum sulfate

High base PAC

High base PAC

Aluminum sulfate

High base PAC

High base PAC

_

5000ppm

2000ppm

500ppm

_

_

Aluminum sulfate

High base PAC

High base PAC

7. Implementation items

3-2 Development of sludge dehydration treatment technology

- Applicability assessment of cartridge filter
- Observation of filtering property of each slurry/sludge (average filtering resistivity)

Here is the evaluation result of average filtering resistivity obtained in each test.



Adsorption treatment water = CF concentrated water > sedimentation sludge

Titanium silicate > active carbon

Figure Evaluation result of average filtering resistivity

Simulated material

Adsorption

treatment water CF concentrated

water

Sedimentation

sludge

Sedimentation

sludge Sedimentation

> sludae Adsorption

treatment water

CF concentrated

water

1-1

1-2

1-3

1-4

1-9

1-5

1-6

Adsorbent

Titanium

silicate

50ppm

10000ppm

Approx.

100000ppr

Approx.

100000ppm

50ppm

200ppm

10000ppm

- Average filtering resistivity calculated from this test result was below 1.0E+12m/kg in all test cases, which show that filtering performance of slurry/sludge is good (excluding 1-9 and 1-10).
- As an overall trend, it was confirmed that filtering resistivity becomes small when coagulant is added and sedimentation sludge is created. However, since cake thickness (volume) becomes large due to coagulant components, filtering velocity becomes slow as indicated in the previous page.
- Additionally, in conditions with no addition of coagulants (1-1, 1-5, etc.), active carbon has smaller filtering resistivity than titanium silicate. Since titanium silicate carries surface charge, water permeability is thought to have been blunted by electrostatic force that grips water molecules. Since addition of coagulants neutralize surface charge, filtering resistivity is thought to have decreased.
- When coagulation-sedimentation treatment is performed in conditions with low concentration of adsorbents (1-9, 1-10), it was confirmed that sludge becomes difficult to be filtered.





Figure Measurement result of moisture content of filtering residue (cake)

*Target value of moisture content after dehydration is set to 60vol.% from the viewpoint of volume reduction treatment

Titanium silicate

- When filtering is performed without adding coagulants, moisture content becomes high at approx. 80vol.%. When filtering is performed by adding coagulants and creating sludge, target value is satisfied at approx. 50vol.%.
- Since titanium silicate theoretically carries negative surface charge, moisture content in conditions with no addition of coagulants is considered to have become high due to the effect of holding water molecules by electrostatic action. Since crystal structure is three dimensional, it is considered unlikely that water will be held inside by swelling effect (it could contain hydration water).
- In conditions with addition of coagulants, moisture content is considered to have become relatively small, as surface charge is neutralized and the aforementioned electrostatic impact has been mitigated. Water content of 40-50 vol.% is considered to be pore water in the cake that is difficult to be dehydrated by filtering.





Impregnated active carbon

- Moisture content was 60-70vol.% in all treatment conditions, regardless of whether coagulant is added or not.
- It is difficult to perform dehydration treatment up to target value of 60vol.%, but it was confirmed that dehydration is possible to a close level.
- Since active carbon theoretically does not carry surface charge, water molecules are unlikely to be held by electrostatic action, and there is also no swelling effect. Thus, it is thought that <u>moisture content is mostly constant regardless of whether coagulant is added</u> <u>or not</u>.
- Additionally, the reason why moisture content was high compared to the condition where titanium silicate created sedimentation sludge (1-3, 1-4) is considered to be because <u>water may be held in micropores as numerous micropores exist on the surface of</u> <u>active carbon particles</u>.
- Since cake density becomes small due to small true density of active carbon particles, mass water moisture content tends to become large compared to that of titanium silicate.



③-2 Development of sludge dehydration treatment technology

> Applicability assessment of cartridge filter

• Observation on moisture content of filtering residue (cake)

Based on the test result, the following items are considered to be parameters that largely contributed to moisture content of filtering residue (cake) remaining after pressurized filtering with the cartridge filter.

- Porosity of cake (particle filling density)
- ·Electrostatic properties of particle surface
- Crystal structure of particles
- (existence of swelling, existence of micropores, etc.)

Cake moisture content increases due to surface properties and physical properties of the particles themselves. However, if coagulation-sedimentation treatment is performed under appropriate conditions, impact of surface properties of the particles can be mitigated, and dehydration is possible up to 60-70vol.%. Changes in cake moisture content due to filtering conditions such as filtering pressure, filtering time and filter aperture were minor.

This test result suggested the possibility that <u>moisture held in cake</u> pores is 40-50vol.% in terms of water content. Additionally, <u>moisture content of 20-30% is estimated to become</u> <u>even greater</u>, <u>depending on physical properties of powder</u> <u>adsorbents to be used</u>.



*Since it is difficult to remove pore water with the pressurized filtering method, it is necessary to apply heated dehydration treatment if dehydration treatment to a near-dry level is required.

However, in terms of hydrogen measures and volume reduction, dehydration treatment up to 60-70vol.% is considered to be enough.



③-2 Development of sludge dehydration treatment technology

 Observation on degraded filtering performance under sedimentation sludge (no prior concentration) conditions

In additional tests 1-9 and 1-10, CF concentration treatment was not performed after adsorption treatment using powder adsorbents but coagulation-sedimentation treatment was performed on liquid waste with SS concentration of several dozen to several hundred ppm to simulate the case of coagulation-sedimentation treatment. Later, cartridge filter filtering test was conducted using the generated sedimentation sludge. As a result, significant decrease in filtering velocity was confirmed, and it was observed that it was caused by clogging by gelatinous materials.

Gelatinous sludge confirmed in test case 1-10



Figure Cartridge filter test equipment (during pressure filtering test)

Since coagulant components were large, cakes with high viscosity and high moisture content were formed



(a) Before drying (b) After drying Figure Filtrate and filtering residue (cake)

Verage filtering testing the filtering testing the filtering testing t

No.93

Compared to other test cases, significant increase in average filtering resistivity was confirmed.

Filtering velocity also dropped significantly.

- <u>Gelatinous materials and powder adsorbent particles are mixed and deposited</u>, and it is confirmed that sludge volume has increased.
- As the test result (average filtering resistivity, etc.) indicates, <u>filtering velocity significantly</u> <u>dropped compared to other tests</u>.

It is thought that permeation of filtrate is inhibited by clogging of filter holes due to sludge with high moisture content (gelatinous materials).



No.94

3-2 Development of sludge dehydration treatment technology

> Observation on cause of degraded filtering performance under low adsorbent concentration conditions

Gelation of coagulant components

In test cases 1-9 and 1-10, cakes difficult to be filtered are thought to have formed due to decreased filtering velocity and increased average filtering resistivity. In test cases 1-9 and 1-10, it is possible that coagulants were excessively added as adsorbent concentration was small at several dozen to several hundred ppm.



Figure Image of consumption of supersaturated components of coagulant

(1) Components precipitated on the surface

- Components precipitated in narrow space or on chemically active areas on solid phase surface.
- · Reaction proceeds preferentially.
- Moisture content of precipitates is low.

(2) Components forming gel

- Components forming colloid and solid phase in supernatant.
- Excess components that can not precipitate on solid phase surface are consumed in this reaction.
- Moisture content of precipitates is high. Assumed to be amorphous with small crystallinity.

*Since AI reagents area used for the coagulant, it is considered to precipitate in the form of aluminum hydroxide.

As the amount of powder adsorbents becomes smaller, the percentage consumed for precipitation on the surface becomes smaller and percentage consumed for gel formation increases.

When adsorbent concentration is large (test case 1-3, etc.), because of the enormous surface area in liquid phase, almost all coagulant components are consumed for precipitation on the surface and there are only a few excess components for forming gel. On the other hand, when adsorbent concentration is small (test case 1-9, etc.), because there is not enough surface area in liquid phase, there are excess components of coagulant. Excess components are considered to become gelatinous as they precipitate containing a large amount of moisture in supernatant.

<u>Gelatinous materials cause significant blockage of filter holes</u>, which is considered to have caused predominant decrease in filtering performance.



③-2 Development of sludge dehydration treatment technology

Summary of element test result

- In treatment conditions No.1-3, it was evaluated that cartridge filter satisfies the required throughput.
- On the other hand, in treatment condition No.4, decreased throughput considered to have been caused by filter hole blockage due to gelatinous materials was confirmed, and it was suggested that dehydration treatment using cartridge filter may be difficult.
- Moisture content of sludge due to the cartridge filter somewhat affects front treatment, but it can be reduced to around 60-70%.

Treatment process evaluated to satisfy target throughput. Filtering performance of generated slurry/sludge is good.



③-2 Development of sludge dehydration treatment technology

[Results to date]

- ✓ Regarding sedimentation sludge generated by RO concentrated water treatment, volume reduction effect and generated amount of hydrogen were evaluated and target value after dehydration was set to 60vol.%.
- ✓ Technology using the cartridge filter was selected as the dehydration treatment method of slurry/sludge, and element test proposal was developed to evaluate applicability to actual equipment.
- Based on the element test result, adsorption treatment water, CF concentrated water and sedimentation sludge all showed good filtering performance with the cartridge filter and were evaluated to satisfy throughput.
- Possibility of low applicability was indicated for a system that performs coagulation-sedimentation treatment without concentration treatment after adsorption treatment, as sedimentation sludge becomes gelatinous form that is difficult to be filtered.
- ✓ Moisture content of cake after filtering was 60-70vol.%, depending on treatment conditions. This indicates the possibility of volume reduction of the generated amount of sludge to the set target level.

[Future issues to be studied]

- Equipment scale, throughput and operation pattern shall be studied for each treatment system proposed, thereby selecting a more appropriate system.
- ✓ Different dehydration technologies shall be studied, and development issues considered necessary for the study of system rationalization, such as discharge of cake by air backwashing, shall be organized.

[Summary]

Dehydration technology using the cartridge filter was selected as the sludge dehydration technology, and its applicability was evaluated with element tests. Based on element test results, multiple treatment processes where dehydration treatment using the cartridge filter is feasible were selected, and conceptual study of the sludge collection system was conducted.



8. Specific goals for achieving the purpose of the project

No.97

(1) Liquid/Gas Phase Systems	
(1) Development of soluble α nuclide removal	In order to verify the effectiveness and feasibility of soluble α nuclide removal
technology	equipment, test methods, test equipments and test plans assuming use of actual
• Study of soluble α nuclide removal test	liquid are materialized.
assuming use of actual liquid	(Target TRL at end: Level 3)
Element test assuming fuel debris retrieval work	Regarding soluble α nuclide removal equipment, candidate adsorbents are
WORK	selected upon acquiring a nuclide adsorption performance data under an
	adjustment policy is also set
	(Target TPL at end: Level 4)
2 Development of RO concentrated water	Regarding RO concentrated water treatment facility powder adsorbents and
treatment technology	coagulants are selected upon evaluating nuclide removal performance of
Selection of adsorbent and condensing	powder adsorbents and coagulation-sedimentation performance of coagulants
agent	(Target TRL at end: Level 4)
- Study of applicability to actual aquipment	Depending concentrated water treatment facility, concentual design of treatment
	Regarding concentrated water treatment facility, conceptual design of treatment
	nethous and equipment is implemented based on tests using the sedimentation
	(Torget TPL at and Level 4)
3 Development of secondary waste treatment	(Target TRL at end. Lever 4) Candidate stabilization treatment technology is selected for sludge generated
technology	from coagulation sedimentation tank. At the same time, pretreatment for liquid
Inspection of pretreatment technology	waste such as supernatant water is also selected
mapeellon of pretreatment teenhology	(Target TRL at end: Level 3)
Study of applicability to actual equipment	Regarding pretreatment technology, applicability to actual equipment is verified
	based on element test results. Conceptual design is also established for
	pretreatment equipment.
	(Target TRL at end: Level 4)

Goals indicated in the above table have been achieved.

*TRL: Technology Readiness Level





FY2021 Subsidy Project of Decommissioning and Contaminated Water Management Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)

Final Report

(2) Criticality Control Technology

June 2023

Technology Research Association International Research Institute for Nuclear Decommissioning (IRID)

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1. Purpose and goals of "Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)"

[Purpose of Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)]

At Tokyo Electric Power Company Holdings Inc. Fukushima Daiichi Nuclear Power Station (1F), it is considered that nuclear fuel melted with reactor internals and exists as fuel debris in the reactor pressure vessel (RPV) and primary containment vessel (PCV).

Fuel debris in RPV and PCV is currently considered to be in subcriticality, but since the plant itself is in an unstable state which is different from the initial design, such as the reactor building (R/B), RPV, PCV, etc. being damaged due to the accident, it is necessary to prevent the diffusion of radioactive materials and achieve a stable state by removing fuel debris and maintaining subcriticality of fuel debris.

Against the above background, this project will conduct reviews to realize large-scale fuel debris retrieval task under engineering and project management performed by Tokyo Electric Power Company Holdings Inc. (TEPCO) in accordance with the "Mid-and-Long-Term Roadmap towards the Decommissioning of TEPCO's Fukushima Daiichi Nuclear Power Station Units 1-4" (hereinafter Mid-and-Long-Term Roadmap). Development results of this project will be utilized in engineering by TEPCO.

The purpose of this project is to facilitate decommissioning and contaminated water measures of 1F and raise the level of science and technology of Japan, by implementing the project that supports the development of technologies that contribute to decommissioning and contaminated water measures of 1F in accordance with the Mid-and-Long-Term Roadmap and "FY2021 Decommissioning Research and Development Plan" (Decommissioning and Contaminated Water Team Meeting / Secretariat Liaison Meeting (86th)).

In specific, technology related to on-site operation methods shall be developed regarding soluble α nuclide removal technology for α nuclides considered to be eluted in circulation cooling water from fuel debris, RO concentrated water (*) treatment technology, secondary waste treatment technology, as well as criticality approach monitoring technology and neutron absorbent technology.





1. Purpose and goals of "Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)"

[Overall development goals]

Perform necessary element technology development and tests regarding system technology and technology for ensuring safety, required to further expand the scale of retrieving fuel debris and internal structures, in accordance with R&D results obtained up to date.

[Specific goals of development items]

① Development of on-site operating procedures

- Develop on-site operating procedures for criticality approach monitoring using neutron detectors and for non-soluble neutron absorbents, and provide information to be utilized by the fuel debris retrieval method team for design of the retrieval system and study of throughput.
- Evaluate subcriticality measurement performance of the three new types of neutron detector, and incorporate it in the on-site operation procedures.
- (2) Development of solidified absorbent technology
- Acquire data on impact on the drying process of fuel debris with solidified absorbent (water glass) attached, and utilize it for ① Development of on-site operating procedures as well as study on equipment operation such as drying time of fuel debris.



2. Overview of subsidy project

Development of on-site operation methods for criticality approach monitoring technology and neutron absorbent technology

Regarding criticality control technology, criticality prevention, criticality approach monitoring, criticality detection and impact mitigation have been developed in the Government-led R&D Program on Decommissioning and Contaminated Water Management up to the previous PJ. Among these, study on criticality prevention and criticality approach monitoring, which have issues remaining, was materialized based on past technical development results.





2. Overview of subsidy project Gradual criticality control

- Criticality control technology shall be gradually applied considering risks of criticality according to the retrieved amount and retrieval method of fuel debris.
- Since criticality risk relatively increases as the retrieval method has greater amount retrieved and larger change of state, it shall be made possible to select diversified criticality control technologies.

State of fuel debris

Small Small State of fuel debris Large							
Retrieval task		Internal	Fuel debris retrieval				
		investigation	Retrieval gradually expanding the scale			Full-scale retrieval	
Retrieved amount (se		Trace amount (several g)	Small amount (several kg)Small amount (several kg)		Full-scale (up to several hundred kg per day)		
Retrieval method		Grabbing, suction, etc.	Grabbing, suction, etc.	Core boring, etc.	Core boring, etc.	Core boring, chisel, etc.	
 Criticality prevention Criticality approach monitoring 	Work restriction	Method that does not makes changes to fuel debris	Method that does not makes changes to fuel debris	 Limit of processing amount per task Limit of retrieval position interval 	 Limit of processing amount per task 	 Limit of processing amount per task (expanded) 	
	Criticality approach monitoring	_	_	Neutron flux monitoring	Neutron flux monitoring	Neutron flux monitoring	
		_	_	—	-	Subcriticality measurement*1	Scope of
	Non-soluble neutron absorbent	-	-	-	-	Non-soluble neutron absorbent*2	this proje
	Soluble neutron absorbent	_	_	_	_	Sodium pentaborate water*3 (6000pp)	
③ Detection of criticality	Detection of criticality	PCV gas radiation monitor	PCV gas radiation monitor	PCV gas radiation monitor Neutron flux monitor	PCV gas radiation monitor Neutron flux monitor	PCV gas radiation monitor Neutron flux monitor	
④ Mitigation of impact	Termination of criticality	Injection of sodium pentaborate water	Injection of sodium pentaborate water	Injection of sodium pentaborate water	Injection of sodium pentaborate water	Injection of sodium pentaborate water	

*1, 2 and 3 are selected depending on criticality risk taking retrieval work details and field environmental conditions into account



3. Results of past projects and remaining issues

Development results of criticality approach monitoring technology and contents of this project

Final goals	Goals to be achieved before application to actual equipment	Achievement status	Remaining issues / contents of this project
Subcriticality Establishment of measurement technology	① Establishment of measurement methods	 Selection of method combining reactor noise method and neutron source multiplication method Study of detector sensitivity and arrangement to reduce measurement error 	
	② Development of system specifications	Development of system specifications of detector, measurement circuit, etc.	
	③ Specifications of neutron detector	Development of neutron detector specifications and selection of detector for prototype	
	④ System design and test manufacturing	 Test manufacturing of system consisting of neutron detector, measurement circuit and analysis PC, as test system 	
	(5) Confirmation of feasibility of subcriticality measurement	 Confirmation under uniform fuel debris simulation conditions (KUCA test #1/#2) Confirmation under large-scale fuel debris simulation conditions (KUCA test #3) Confirmation under non-uniform fuel debris / neutron absorbent simulation conditions (KUCA test #4) 	Applicability confirmation test of alternative neutron detectors (corona discharge type, SiC semiconductor type, multicell He-3 type) (KUCA test #5) (→7.1.2) KUCA: Kyoto University Critical Assembly
Incorporation into fuel debris retrieval system	 Confirmation of applicability of neutron detector in the field 	 Organization of specifications of neutron detector unit for transfer with robot arm Development of cable handling concept Test manufacturing of alternative neutron detectors (corona discharge type, SiC semiconductor type, multicell He-3 type) and confirmation of basic performance Design and test manufacturing of neutron detector unit Evaluation of radiation resistance of preamplifier 	
	② Solenoid noise reduction method	Evaluation of impact of simulated noise, organization of draft response measures	
	③ Study of criticality approach management procedures	Evaluation of time required for measurement Criticality approach monitoring procedures in each step of fuel debris retrieval method	Organization of measurement procedures before and after processing, judgment of measurement target, measurement time, work procedures of daily subcriticality maintenance management, repair and maintenance requirements, and request to processing side (\rightarrow 7.1.1)



3. Results of past projects and remaining issues

No.7

Development results of non-soluble neutron absorbent and contents of this project

Final goals	Goals to be achieved before application to actual equipment	Achievement status	Remaining issues / contents of this project
Establishment of criticality prevention technology	① Selection of candidate materials	 Selection of candidate non-soluble neutron absorbent upon confirming basic characteristics, irradiation characteristics and nuclear characteristics (selection among 4 types of candidate absorbent) 	
	② Confirmation of workability and specifications of conditions for maintaining subcriticality	 Estimation of necessary amount of injection with the assumed usage method Confirmation of workability of fuel debris crushed with chisel, confirmation of absorbent distribution, and evaluation of maintenance of subcriticality (solid absorbent) 	
	③ Evaluation of corrosion impact	 Based on long-term irradiation test, hydrogen generation G value (*) is below the design value and diluted seawater with leached absorbent components is pH6 or higher Evaluation of impact on rust inhibitor effect under irradiation 	
	③ Evaluation of secondary effect	 Increase in the number of storage canisters and amount of waste by approx. 10% at maximum with solid absorbents and approx. 40% at maximum with solidified absorbents 	Impact of water glass on fuel debris drying $(\rightarrow 7.2)$
Incorporation into fuel debris retrieval system	① Study of injection equipment and operation method for non-soluble neutron absorbent	 Determination of absorbent injection method Confirmation of impact of chisel processing Confirmation that underwater injection is possible 	Used amount of solidified absorbent Application conditions for processing scenes Recovery procedures from level 2 to level 1 Effectiveness of pre-injection $(\rightarrow 7.1.3)$
	② Design of absorbent injection equipment	 Development of absorbent injection equipment concept based on weight and dimensional constraints of absorbent injection equipment and absorbent transport route constraints 	Downsizing and weight reduction of equipment (\rightarrow 7.1.3)
	③ Verification of combinations of retrieval equipment	Organization of applicable absorbent for fuel debris processing method of each fuel debris position (*) Hudrogon generation C uplus: Amount of hudrogon	a concreted when obserbing 100sV of energy



4. Input and output information

This PJ was conducted in coordination with the Project of Development of Technology for Further Increasing the Scale of Retrieval of Fuel Debris and Internal Structures and Project of Development of Fuel Debris Collection, Transfer and Storage Technologies.

ID	Demand-side project	Provider-side project	Description (overview)	Timing	Usage of information	
1	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Development of technology for further increasing the scale of retrieval of fuel debris and internal structures	Method of interference removal, method of access to fuel debris, method of processing and collecting fuel debris and reactor internals	June 2021	Study of on-site operating procedures for criticality approach monitoring and absorbent spraying	
2	Development of technology for further increasing the scale of retrieval of fuel debris and internal structures	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	On-site operating procedures for criticality approach monitoring with neutron detector, on-site operating procedures for non-soluble neutron absorbent	March 2023	Study of throughput of fuel debris retrieval method	
3	Development of fuel debris collection, transfer and storage technologies	Development of Safety System (Liquid/Gas Phase Systems, Criticality Control Technology)	Data on impact on the drying process of fuel debris with water glass (non-soluble neutron absorbent) attached	March 2023	Study of storage canister drying process	


5. Implementation schedule

Criticality Control Technology

	Implementing operator FY2021									FY	2022																
Large category	Small category	(Also list the subcontractor and testing site, if any)	Apr	Мау	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	Мау	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	
Major mile	estones		Pro	ject ste	ering	A meetir	ıg	Interin	n repor	t	Proje	ect stee	ering m	eeting	▲ Inter	im repo	ort	Projec	t steeri	ng_l	nterim	report	Pro	ject st		▲ Final r	l eport
(2) On-site operation methods for criticality approach monitoring technology and neutron absorbent technology ① Development of on-site operating procedures (1) Development of operating procedures of criticality approach monitoring	a. Organization of prerequisites b. Study of procedures c. Evaluation / revision d. Summary	 Hitachi GE Toshiba ESS Mitsubishi Heavy Industries 	0	rganiz	ation o	f prere	equisite	es Study	of proc	edures		•	Inter	im sun	nmary			Ev	aluatio	ng n / rev	ision			rr Su	mmary		
(ii) Subcriticality measurement test of neutron detector	a. Test plan b. Test preparation and arrangement c. Element test d. Summary	(1) Toshiba ESS Subcontractor: FEO/CETD Testing site: KUCA Nagoya University (2) Mitsubishi Heavy Industries Subcontractor: IPL Testing site: KUCA	Tes	prepar	ation a	and arr	CUCA to	ent est		Eļ	ement	test	•	Interim	sumn	t plan Irra	diation Ana	test lysis a	nd eva	luation			Eler	nent te	st Sum	mary	
(ii iDevelopment of operating procedures for non-soluble absorbents	a. Organization of prerequisites b. Study of procedures c. Evaluation / revision d. Summary	 Hitachi GE Toshiba ESS Mitsubishi Heavy Industries 	Orga	anizatio	on of p	rerequ	uisites	Study c	f proce	dures			Interin	m sumi	mary			Evi	aluation	n / revi	sion			/ Sur	nmary		
② Development of solidified absorbent technology	a. Test plan b. Test preparation and procurement of materials c. Element test d. Summary	(1) JAEA Subcontractor: Inspection Development Company Ltd. Testing site: JAEA Consignee: Shizuoka Institute of Science and Technology Testing site: Shizuoka Institute of Science and Technology		est pla	'n	Tes	t prepa	ration	and pro	Basi	c test		T Inte	est pre	parati	on and	procur	ement	Basi	ç test c	of para	meters		s	ummai	y	



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6. Implementation framework





- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring

[Issues]

Criticality approach monitoring in rough work steps of top/side access method is studied in a relevant project^(*) (Figure 1). In order to discuss operating procedures and throughput with the fuel debris retrieval method team, it is necessary to further refine the work steps and materialize criticality approach monitoring procedures.

[Implementation items]

- Criticality approach monitoring procedures using neutron detectors shall be organized in a step diagram for the fuel debris retrieval method and processing and collection equipment studied in the relevant project^(*).
- Neutron response to criticality approach during fuel debris processing shall be studied, and procedures for response to determine criticality approach shall be studied.
- Organization of measurement procedures before and after processing, judgment of measurement target, measurement time, work procedures of daily subcriticality maintenance management, repair and maintenance requirements, and request to processing side shall be studied.

[Goals]

 Develop on-site operating procedures for criticality approach monitoring using neutron detectors, and provide information to be utilized by the fuel debris retrieval method team for design of the retrieval system and study of throughput.



Figure 1 Criticality approach monitoring during fuel debris retrieval (example of side access PLAN-A)

(*) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)



- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - a. Organization of prerequisites

Functions that comprise criticality approach monitoring

Name	Subcriticality measurement detector	Criticality approach monitoring detector ^(Note 1)	Continuous monitoring detector ^(Note 1)			
Usage purpose	Grasp pre-work conditions	Determine start of processing	Detect unexpected changes			
Eurotion	Subcriticality measurement	Criticality approach monitoring before and after processing	Continuous monitoring of neutron flux during processing			
Function	Measurement of absolute value of multiplication factor	Measurement of relative cha	ange of multiplication factor			
Form	Arm-mounted type	Processing tool juxtaposition type	Platform-mounted type / processing tool juxtaposition type			
Weight	30kg-100kg	30-50kg	Less than 30kg			
Measurement time	Several hours to 1 week (depends on site environment)	Around 10min.	Continuous			
Measurement position	One point near the retrieval position(Note 2)	Move as necessary according to the processing position(Note 2)	Same as left / location with an overhead view of the processing position			

(Note 1) "Criticality approach monitoring" and "continuous monitoring" may be used concurrently under conditions such as weak gamma ray in the field and strong neutrons of fuel debris.

(Note 2) Measurement positions for fuel debris are not fixed and may be changed each time.







Platformmounted type



7.1.1 Operating procedures of criticality approach monitoring b. Study of procedures Overview of implementation items of 7.1.1.b - Top access method for transferring -----Side retrieval method ------- - - Top retrieval method - - the unitized structure -(3) Criticality monitoring 1 Criticality monitoring 2 Criticality monitoring ③ Criticality monitoring 10 items raised as method of top access method during top retrieval method of side retrieval method of side retrieval issues were studied. method for transferring PLAN-A method PLAN-B method (→No.14) in discussing the unitized structure (→No.15) (→No.16-18) operating (→No.16-18) procedures and throughput with the (4) Monitoring range of three types of criticality approach monitoring (\rightarrow No.19) fuel debris retrieval method team. (5) Organization of measurement procedures during fuel debris processing and retrieval work (\rightarrow No.20) (6) Organization of roles of criticality approach monitoring (\rightarrow No.21) (7) Basic approach of criticality monitoring based on organized environmental conditions of partial submersion processing and submersion processing (→No.22) (8) Organization of criticality risk in submersion processing (\rightarrow No.23-26) (9) Monitoring procedures for representative processing examples (laser processing) (\rightarrow No.27) (10) Maintenance of monitoring system (→No.28)



No.13

7. Implementation items

- 7.1 Development of on-site operating procedures

- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - b. Study of procedures ① Criticality monitoring method during top retrieval

[Study results]

Method of criticality approach monitoring in fuel debris retrieval steps was studied using the top retrieval method as a representative example.

Criticality approach monitoring consists of three types of neutron measurement, but it is unnecessary to have all three types of monitoring in all work steps.

Necessary monitoring functions shall be used differently according to the assumed criticality risk. (3 types or 2 types)



Upper grid plate

Remaining fuel in the core

No.14



Arm-mounted type detector (subcriticality measurement)

Processing tool

detector

- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
- b. Study of procedures⁽²⁾ Criticality monitoring method of side retrieval PLAN-A method

[Study results]

Criticality approach monitoring method was studied for the side retrieval PLAN-A method.

Of the three criticality monitoring functions, criticality measurement shall be performed with the armmounted type detector and criticality approach monitoring and continuous monitoring during processing shall be performed with the processing tool juxtaposition type detector, thereby reducing the replacement frequency of processing tools and detectors and minimizing the impact on fuel debris retrieval work throughput.

Subcriticality measurement using armmounted type detector Processing tool before fuel debris juxtaposition type processing detector Confirmation of conditions using arm-mounted type detector before fuel debris Arm-mounted type detector processina Arm-mounted type detector Criticality approach monitoring before processing and continuous 1.1.1.1.1.1.1 monitoring during processing, using processing tool N MEMORY CONTRACTOR juxtaposition type detector Continuous monitoring during fuel debris using tool juxtaposition type detector during processing nterference objectives processing Processing tool Processing too Processing tool juxtaposition juxtaposition Processing tool type detector type detector

Figure Criticality monitoring during fuel debris retrieval (excerpt)

Figure Criticality monitoring during interference removal (excerpt)

Criticality monitoring method of side retrieval PLAN-A method

7.1 Development of on-site operating procedures

7.1.1 Operating procedures of criticality approach monitoring b. Study of procedures③ Criticality monitoring method of side retrieval PLAN-B method / top access method for transferring the unitized structure

• Overview of side retrieval PLAN-B method / top access method for transferring the unitized structure



IRID

7.1 Development of on-site operating procedures

7.1.1 Operating procedures of criticality approach monitoring b. Study of procedures③ Criticality monitoring method of side retrieval PLAN-B method / top access method for transferring the unitized structure

 Regarding the work steps of the side retrieval PLAN-B method and top access method for transferring the unitized structure, criticality approach monitoring procedures when applying the processing and collection method using the following equipment were studied, and issues and risks were identified.



Loading and moving of neutron detector unit (simulated) with the heavy weight object transfer equipment (Side retrieval PLAN-B method)



Loading and moving of crushing and suction equipment with the heavy weight object transfer equipment (Side retrieval PLAN-B method)



Drawing of interference removal work at core bottom (Top access method for transferring the unitized structure)

No.17

Drawing of fuel debris retrieval at pedestal bottom after carrying out bottom RPV mirror and CRD housing (Top access method for transferring the unitized structure)

7.1 Development of on-site operating procedures

7.1.1 Operating procedures of criticality approach monitoring b. Study of procedures③ Criticality monitoring method of side retrieval PLAN-B method / top access method for transferring the unitized structure

[Studied results]

 Issues and risks regarding work steps of side retrieval PLAN-B method and top access method for transferring the unitized structure were identified in the table on the right.

No.	Issues
1	Can existence and amount of fuel debris be confirmed from camera footage, neutron flux distribution and characteristic gamma ray distribution?
2	Can the neutron detector be installed at an appropriate position and distance with the heavy weight object transfer equipment which is used to transfer neutron detectors?
3	Can subcriticality be measured for fuel debris that is above criticality mass and weighs several dozen kg?
4	Can the neutron detector be installed if there is an inclined surface on fuel debris?
5	Can the neutron detector be installed at an appropriate position and distance by transferring the neutron detector on the platform?
6	Can reactivity input be caused by crack propagation and water intrusion by directly installing the equipment on fuel debris?
7	How would it be managed if uncollectible lumps and puddles in depression are formed when lowering the water level and processing and collecting sediments?
8	The cutting surface is predicted to become lower than the pedestal bottom, when removing the fuel debris mixed with bottom concrete. Thus, measures against accumulation of cooling water discharged from the worker access tunnel are considered to be necessary.
9	Can the neutron detector be transferred between the shroud perimeter and RPV and installed at an appropriate position and distance?
10	Reactivity may be input due to discharge of cooling water being prevented by measures to prevent outflow of filling solidification agent (transient state until solidification agent injection).
No.	Risk
1	Reactivity input due to crack propagation and water intrusion caused by equipment drop and impact on fuel debris.
2	Crack propagation due to falling of fuel debris, and excessive criticality mass, during processing.
3	Reactivity input due to crack propagation and water intrusion caused by detachment, falling and impact of fuel debris from solidified parts.

IRID

- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - b. Study of procedures (4) Monitoring range of three types of criticality approach monitoring
- Respective monitoring range of the three types of criticality approach monitoring functions was organized. (figure shows the example of top retrieval)

Function: **Subcriticality measurement** Measurement point: 1 point near the retrieval start point

Usage purpose: Grasp pre-work conditions

Function: **Criticality approach monitoring before and after processing** Measurement point: Move as necessary according to the processing position Usage purpose: Determine start of processing

Function: **Continuous monitoring of neutron flux during processing** Measurement point: Location with an overhead view at a distance from the processing position(*1) Usage purpose: Detect unexpected changes

Continuous monitoring Monitoring area



(*1) Image of continuous monitoring when moving as necessary according to the processing position has the same monitoring area as criticality approach monitoring in the figure in the center.



- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring

b. Study of procedures (5) Organization of measurement procedures during fuel debris processing and retrieval work

Measurement procedures during fuel debris processing and retrieval work were organized.

- Measure neutron multiplication factor (keff) with the reactor noise method before starting fuel debris retrieval work 1

This is the basis for the neutron source multiplication method.

- -Measure neutron counting rate before and after fuel debris processing 2
- Perform continuous monitoring of neutron counting rate during fuel debris retrieval processing
- Evaluate neutron multiplication factor (keff) with the neutron source multiplication method if the neutron counting rate changes
- Stop work when the neutron multiplication factor reaches the judgment criteria
- ·Use an absorbent
- Measure neutron multiplication factor (keff) with the reactor noise method after using the absorbent (1) This is a re-measurement of the basis for the neutron source multiplication method.

 k_{eff} : Neutron multiplication factor k_{ref} : Neutron multiplication factor at the time of measurement with the reactor noise method ϕ_{ref} : Neutron flux at the time of measurement with the reactor noise method

 $\boldsymbol{\phi}$: Pre-retrieval neutron flue







- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - b. Study of procedures 6 Organization of roles of criticality approach monitoring
- Roles of the three types of monitoring functions when approaching criticality were organized.
- ·Measure neutron multiplication factor (keff) with the reactor noise method before starting fuel debris retrieval work (1)
- This is the basis for the neutron source multiplication method.
- •Measure neutron counting rate before and after fuel debris processing 2

measurement

- Perform continuous monitoring of neutron counting rate during fuel debris retrieval processing
- · Evaluate neutron multiplication factor (keff) with the neutron source multiplication method if the neutron counting rate changes
- · Stop work when the neutron multiplication factor reaches the judgment criteria
- Use an absorbent
- ·Measure neutron multiplication factor (keff) with the reactor noise method after using the absorbent (1)
- Determine work restart restart work
- The aforementioned monitoring functions shall be used differently according to the situation in the field, as indicated in sheets No.14-15.





k=1.00 criticality multiplication factor k-alarm Measurement (2) Measurement (2) Retrieval Retrieval Measurement (1) Neutron source multiplication method Neutror Reactor noise method Retrieval work (time lapse)

Criticality approach monitoring before and after processing



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- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring

b. Study of procedures (7) Basic approach of criticality monitoring based on organized environmental conditions of partial

scattering.

submersion processing and submersion processing

- Environmental conditions of partial submersion processing and submersion processing, which are prerequisites of criticality control, were organized.
- Aerial criticality risk is small and does not require the same kind of control as being underwater, but localized attention such as puddles is necessary.

Partial submersion processing **Submersion** processing Water table has not formed above fuel debris. Water table has formed There is possibility of puddle below fuel debris. above fuel debris. Cooling water is free-flowing. Water drops are falling. underwater from start to Processing is performed while spraying water collection. around the processing area to prevent dust

to prevent dust scattering Water drops are falling. Possibility of puddle below debris

Figure 1 Environmental conditions of partial submersion processing

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Processing is performed

- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring

b. Study of procedures[®] Organization of criticality risk in submersion processing

(Example of processing work) Laser gouging

[Processing test result] Case of underwater to water flow

- Processing speed 330g/min (maximum)
- Particle size distribution of chip powder (Table 1) Particles greater than 1mm account for a large portion at 70-80%.

[Criticality risk assessment]

- Criticality evaluation result (Figure 1) indicates that critical mass is around 40kg(*3), and it requires around 2 hours of processing time to reach said amount.
- Most chip powder settles out quickly (speed 40-80[cm/s]) even if they swirls up underwater, and critical mass will not be reached.
- There are hardly any criticality risks during processing, but attention is needed so that chip powder does not collectively accumulate.

(*1) Development of Fundamental Technology of Retrieval of Fuel Debris and Internal Structures FY2016 Final Report IRID

(*2) Development of Fuel Debris Criticality Control Technology FY2014 Research Report $\ensuremath{\mathsf{IRID}}$

(*3) Easing of restrictions is expected if knowledge of fuel debris properties is gained in the future.





State of laser gouging (figure on left) and simulated test body after processing (figure on right)

Table 1 Particle size distribution of sediment $(\%)^{(*1)}$



- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - b. Study of procedures[®] Organization of criticality risk in submersion processing

Fuel debris processing method and overview of criticality evaluation^(*1)

Fuel debris processing method	Assumed criticality scenario	Illustration of events	Overview of criticality evaluation			
Cutter, wire saw, abrasive water jet (AWJ)	Multiple cracks form on fuel debris and water enters		When cracks change the volume by about 1vol% from the state of lump-like fuel debris containing 20wt% structural material, reactivity of about $0.1\%\Delta k^{(*2)}$ is applied.			
Chisel	Crushed and fragmented fuel debris falls into water		When falling and crushing change the volume by about 10vol% from the state of lump-like fuel debris containing 20wt% structural material, reactivity of about $1\%\Delta k^{(*2)}$ is applied.			
Core boring	Water enters drilled holes		When holes 7cm in diameter and 1m in depth are drilled from the state of lump-like fuel debris containing 20wt% structural material, reactivity of about $0.5\%\Delta k$ is applied. Since processing speed of core boring is around 1cm/min., reactivity of about $0.005\%\Delta k/min.^{(*2)}$ is applied.			
Laser	Granular fuel debris cut underwater swirls up	Laser	Since processing speed is slow and swirled up fuel debris settles out before reaching critical mass of 40kg ^(*2) , criticality does not occur. (from previous sheet)			

(*1) FY2015 supplementary budget, Subsidy Project of Decommissioning and Contaminated Water Management, Development of Fuel Debris Criticality Control Technology, Research Report, March 2018, IRID

(*2) Easing of restrictions is expected if knowledge of fuel debris properties is gained in the future.



- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring b. Study of procedures® Organization of criticality risk in submersion processing
- Criticality risk of the processing method studied in the relevant project^(*1) was evaluated based on analysis.

Usage different from the originally intended usage method



There is no criticality

risk in the work itself that picks up small pieces of fuel debris up to 200mm (size that can go into storage canister) underwater. However, attention is needed in work that stirs and breaks up small pieces of fuel debris with a robot arm.



Attention is needed when dropping small pieces of fuel debris and collapsing the deposited small pieces of fuel debris. Processing that could exceed the size of minimum critical mass at optimum deceleration state

Attention is needed

when fuel debris fine powder is swirled up in a wide range underwater. (Guideline: More than 46cm in diameter, more than 71kg in weight)^(*2)

Attention is needed when a large amount of fuel debris fine powder deposits underwater. (Guideline: More than 35cm in radius, more than 499kg in weight)^(*2)



(*1) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)



When crushing fuel debris with a chisel, there is criticality risk when the processing range exceeds $16 \times 16 \times 16 \text{cm}^{(*2)}$.

(*2) Easing of restrictions is expected if knowledge of fuel debris properties is gained in the future.



7.1 Development of on-site operating procedures

7.1.1 Operating procedures of criticality approach monitoring

b. Study of procedures

(8) Necessity of criticality monitoring in submersion processing

- Necessity of criticality monitoring in each processing method was organized by partial submersion processing and submersion processing. (However, criticality risks are not determined only by the processing method but largely depends on fuel debris properties. Thus, need for monitoring does not necessarily mean criticality risks are high.)
- Even if criticality risks are not expected for the processing method . itself, attention is needed for secondary effects such as unexpected usage and deposits in a location different from the processing point.
- In the case of "Attention", it shall be studied to perform criticality approach monitoring or continuous monitoring in case of emergency, but processing range limitations and subcriticality measurement may be omitted by adopting appropriate processing procedures.
- Since chisel processing instantaneously changes the form of fuel debris, it was determined that criticality monitoring is necessary. The basis is processing range limitations and subcriticality measurement + criticality approach monitoring + continuous monitoring, but subcriticality measurement may be omitted if the condition of fuel debris is known.



Table Necessity of criticality monitoring in each processing method

Category	Processing method	Characteristics / overview ^(*)	Necessity of criticality monitoring			
			Partial submersion processing	Submersion processing		
Pick up	Grabbing	Grab small pieces of fuel debris	No	Attention		
	Bucket	Scoop up small pieces of fuel debris	No	Attention		
Suction	Suction	Suction and collection of fuel debris fine powder	No	Attention		
Cutting	Laser gouging	Cutting of fuel debris surface with laser	No	Attention		
erforation	Core boring	 Punching process with hole saw Separate core cutting is necessary Water for discharging chips and cooling blades is necessary 	Be careful if there are puddles	Attention		
	Ultrasonic core drill	Can process ceramics and metals at the same time		Attention		
Cutting	Circular saw	Can process ceramics but tool damage is severe. Blade tip is jagged.		Attention		
	Cutting wheel	Can process ceramics and metals at the same time		Attention		
	Hydraulic cutter	Hydraulic-driven scissors		Attention		
	AWJ	Cutting with high-pressure water mixed with abrasive Dispersion of cutting width to the surrounding environment		Attention		
	Laser cutting	Thermal cutting by laser heat		Attention		
rusning	Hammer (chisel)	crusning by pressing the chisel against the target and striking blows with up-and-down movement of piston		Yes		

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Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)

- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - b. Study of procedures
 - Monitoring procedures for representative processing examples (laser processing)
- Laser cutting, which is easy to understand, was used as a representative example to study the measurement procedures during fuel debris processing. (*1)
- A condition of criticality approach due to a straight cutting line forming on the fuel debris and water entering from there was assumed.
- Neutron multiplication factor of fuel debris before processing is near keff=0.95, and it is assumed that it barely does not exceed the judgment criteria.
- Measure neutron counting rate before processing.
- When processing starts, reactivity of Δk=0.016^(*2) is added in 10min. of processing, and neutrons discharged from fuel debris increase by 1.5 times. (Figure)
- Measure neutron counting rate after processing. It is observed that the neutron counting rate increased by 1.5 times, and it is estimated with the neutron source multiplication method that the neutron source multiplication factor changed from 0.95 to 0.967.
- It is judged that the neutron multiplication factor exceeded the judgment criteria, and retrieval work is stopped. Since increase of neutron counting rate stops almost at the same time as when processing ends (Figure), operators can make decisions with plenty of time.

(*1) There is no risk of criticality with one cutting line, but extreme conditions were set as an easy-to-understand example.

(*2) Easing of restrictions is expected if knowledge of fuel debris properties is gained in the future.



[Evaluation conditions]

Fuel debris composition: Concentration 4wt% uranium^(*2)

No structural material, FP, Gd

Water is in the pores of the fuel debris, and keff=0.95 of the initial condition before processing.

Processing to insert a cutting line 2cm wide, up to 60cm deep and up to 60cm long is assumed. $(^{\prime}1)$

Processing speed of laser cutting is 1[mm/sec], and 10min. of processing is performed. Effective multiplication factor was calculated with the MVP code, and neutron response was calculated with single-point reactor kinetics analysis.

Neutron source strength 1[n/sec] Neutron detector detection efficiency 100 [%]



Figure Simulation of neutron response assuming laser cutting



- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring
 - b. Study of procedures (1) Maintenance of monitoring system
- Approach on daily inspection, periodic inspection and corrective maintenance was organized for maintenance of the criticality approach monitoring system (Table).
- In daily inspection, the detector unit is moved to the maintenance area (assuming SFP, etc.^(*)) in the building^(Note 1) to perform remote operation^(Note 2). (Figure)
- In some periodic inspection and corrective maintenance when defects occur, the unit is decontaminated and transferred outside the building and worked on by workers.



Table Maintenance details of criticality approach monitoring system

Figure Maintenance area inside building

	Purpose	Frequency	Inspection description	Inspection items	Method
(Note 1) Moving the fuel debris	Preventive maintenance	Daily inspection	System integrity, noise impact	Confirmation of operation with neutron source, background measurement	Move the detector unit to the remote maintenance area, and perform indirect maintenance of fuel debris processing and collection equipment in remote operation, using the neutron source equipment installed in said area. (required time is around 1 hour) Replacement is necessary as neutron source itself also attenuates.
the detector unit to the maintenance area and putting it on standby is a routine operation.		Periodic inspection 1-2 times per year	Integrity of detector / connector / cable, detector sensitivity	Discrete plateau, insulation resistance, capacitance, cable characteristic, source calibration	If remote maintenance in the maintenance area is difficult, remove the detector unit from the fuel debris processing and collection equipment, take it out of PCV, and perform direct maintenance after decontaminating it. (required time is around several days)
(Note 2) Method of remote operation of neutron detector calibration using neutron source needs to be developed in the engineering stage.	Corrective maintenance	Irregular	Disassembly and inspection, repair, replacement	Gas leak, decreased insulation, poor contact, disconnection, circuit diagnosis, etc.	If the system experiences defects and the detector cannot be restored inside PCV, remove the detector unit from the fuel debris processing and collection equipment, take it out of PCV, and perform inspection with direct maintenance after decontaminating it. (more than 1 week if disassembly and assembly are required)



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🖊 (*) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)

- 7.1 Development of on-site operating procedures
- 7.1.1 Operating procedures of criticality approach monitoring d. Summary

[Result]

 Regarding criticality approach monitoring during fuel debris retrieval, image of on-site operation of items, such as monitoring range of neutron detector, measurement procedures, criticality risk during processing and system maintenance, was materialized, and information was provided for the fuel debris retrieval method team to utilize it to study the retrieval system design and throughput.



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector

[Issues]

Three types of neutron detector (corona discharge type, SiC semiconductor type, multicell He-3 type), which are candidates from the viewpoint of subcriticality measurement, are developed in a relevant project^(*) (Figure 1). In order to confirm that these detectors can be applied in the field, it is necessary to verify that subcriticality can be measured and evaluate the performance of criticality approach monitoring.

[Implementation items]

Structure a subcriticality system simulating fuel debris with Kyoto University Critical Assembly (KUCA), and perform subcriticality measurement test using the developed neutron detector. (Figure 2)

Additionally, evaluate neutron

measurement performance under gamma ray environment through tests.

[Goals]

- Evaluation of subcriticality measurement performance of neutron detector
- Incorporation into operating procedures of neutron detector assuming the field



Figure 2 Kyoto University Critical Assembly

(*) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)

discharge type detector



No.31

7. Implementation items

- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - a. Test plan

Test 6	Neutron detector	Purpose	Result
KUCA test (1st time, 2nd time)	B-10 proportional counter	Confirm that subcriticality can be measured in a small system using uranium fuel	 Measurement error (2%-10%) of different subcriticality, from near criticality of neutron multiplication factor =0.95 to deep subcriticality of 0.7 Impact on measurement error due to uncertainty (neutron energy spectrum) of mixing state of fuel debris and water is small Measurable distance between fuel debris and detector is within 20cm underwater and within 35cm in air
KUCA test (3rd time)	B-10 proportional counter	Confirm that subcriticality can be measured in a large (uniform) system using uranium fuel	Confirm that local subcriticality near the detector can be monitored (monitoring of the entire system is difficult)
KUCA test (4th time)	B-10 proportional counter	Confirm impact of large and uniform system and arrangement of neutron absorbents	Subcriticality was measured with a system simulating uneven fuel debris, and positional dependence was confirmed. Test was conducted in a condition simulating surface application of non-soluble neutron absorbent, and it was confirmed that subcriticality measurement can be performed. $(-15 \sim +14\%)$
KUCA test (5th time)	Corona discharge type SiC semiconductor type Multicell He-3 type	Confirm that alternative candidates of the neutron detector can measure criticality	Implemented June 28 - July 9, 2021 Test target this time

Table Summary of KUCA tests conducted in relevant projects up to previous PJ and test details of this project



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - a. Test plan

No.	Detector name	Supplier	Category	Remarks				
1	Fission ionization chamber	Photonis	Fission ionization chamber	Neutron detector considered to be adopted in PCV				
2	Improved small-size B-10 proportional counter	IRID / Hitachi-GE		(studied in relevant project (*))				
³ Te	B-10 proportional counter Est implemented up to previous PJ			Neutron detectors being studied in criticality				
4 Test tar	He-3 proportional counter (Multicell-type He-3 proportional counter) get this time	CETD	Gas detector	PJ(confirmed to be usable for subcriticality measurement with the Feynman-α method)				
5	B-10 corona discharge type detector	RosRAO						
6	He-3 corona discharge type detector	RosRAO		Neutron detector considered to be adopted in PCV internal investigation				
7	SiC semiconductor detector	IPL	Semiconduc	(neutron detector with radiation resistance, studied in relevant project*)				
8	CMOS detector	IRID / Hitachi-GE	tor detector					

Table Candidate neutron detectors for subcriticality measurement



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - b. Test preparation and arrangement

Appearance of neutron detectors that underwent performance assessment in this KUCA test



Corona discharge type neutron detector (B-10/He-3)



Multicell-type He-3 proportional counter



 $50 \times 50 \times 540$ mm

SiC semiconductor neutron detector





- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector c. Element test

KUCA test system in this project

- Test reactor is composed of fuel bodies combining enriched uranium and polyethylene (solid moderator)
- The neutron detector to be tested is arranged adjacent to the fuel bodies, and analysis is conducted with the Feynman-α method by collecting time series data of neutron counts



Layout of KUCA test core and detector (keff=0.95)



State of actual test

IRID



measurement of neutron detector

c. Element test

Evaluation procedures of subcriticality measurement (Feynman- α method)



Frequency distribution of time list data

Time interval (sec)



Plot variance-to-mean ratio (Y value)

$$Y(\Delta T) = Y \infty \left(1 - \frac{1 - \exp(-\alpha \Delta T)}{\alpha \Delta T}\right)$$

Derive α (prompt neutron attenuation constant) by fitting (red line) with the theoretical equation.



 α : Prompt neutron attenuation constant

(1/sec) β : Delayed neutron percentage ℓ : Prompt neutron life (sec) k_{eff} : Neutron multiplication factor Δ T: Gate time width of neutron count (sec)

Y: Variance-to-mean ratio of neutron count

Derive keff with the measured $\alpha,$ and I and β calculated in advance.

keff obtained above and keff (reference solution) calculated with the analysis code were compared.



No.36

7. Implementation items

- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - c. Element test

This KUCA test result (corona discharge type, multicell type)

- It was confirmed that the corona discharge type and multicell He-3 type detectors can measure subcriticality with the same level of precision as the conventional B-10 proportional counter (Figure, Table).
- With the multicell He-3 type detector, event where operation became unstable due to changes in count indication value was confirmed.



Figure Difference between measured value and reference value (corona discharge type, multicell type)

Since keff calculation of the KUCA test system using the MVP code tends to overestimate by around 0.5%dk at critical conditions, this is calibrated



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - c. Element test

This KUCA test result (SiC semiconductor type)

Data obtained with the SiC semiconductor detector is processed with the Feynman-α method, and subcriticality was evaluated. As the following Figure and Table show, it was confirmed that subcriticality can be measured with a difference of around 1%dk. If subcriticality is deep, difference from the reference value increased to around 4.8%dk, but it was confirmed that subcriticality can be measured with the same level of precision as the conventional B-10 proportional counter.



Table Measurement result of each test system

Test core	Reference value	Measured value	Difference (%dk)	Remarks
Case1-1 (keff=0.95)	0.948	0.944	-0.4	
Case1-2 (keff=0.97)	0.962	0.958	-0.4	
	0.979	0.968	-1.1	Measurement
Case1-3	0.979	0.958	-2.1	changing pulse
(ken=0.98)	0.979	0.981	0.2	discrimination parameters
Case1-4 (keff=0.70)	0.681	0.729	4.8	

Reference value: Core analysis value by MVP

Figure Difference between measured value and reference value (SiC semiconductor neutron detector)



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - c. Element test

Overview of gamma-ray resistance test

[Test purpose]

- Check the upper limit of neutron-discriminable gamma dose rate of the candidate neutron detector for subcriticality measurement, to see up to how much strength of gamma dose rate neutrons can be discriminated.
- Check if measurement is possible with the Feynman-α method in high gamma ray environment.

[Test location]

Nagoya University Co-60 irradiation room

[Test target]

- B-10 corona discharge type detector
- He-3 corona discharge type detector
- Multicell-type He-3 proportional counter
- B-10 proportional counter

Other neutron detectors have been separately evaluated with vendor tests, etc.





State of actual test

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1



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - c. Element test

Example of gamma-ray resistance test result (Wave height discrimination characteristics of B-10 corona discharge type detector)



- 600mV is set as the discrete level that can separate gamma ray and neutrons, based on the measurement result in the figure on the left.
- Decrease in neutron detection sensitivity due to gamma ray was 47% at 10Gy/h.
- It was checked whether Feynman-α measurement is possible with this detector sensitivity. (next page)

Pulse wave height discrimination level of measurement circuit



Number in parenthesis is the total count; no lead shielding

©International Research Institute for Nuclear Decommissioning (Note 1) Unmeasurable unless operation is stabilized at high voltage

- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector

c. Element test

Gamma-ray resistance performance verification test of SiC semiconductor detector (1/2)

Test details	Verification items and judgment criteria		Result					
(1) Confirmation of γ ray removal Conducted at NPL (National Physical Laboratory) which owns equipment that can irradiate ⁶⁰ Co of 1.5×10^{13} Bq. (see below figure) Data of irradiation up to 100Gy/h was collected, and pulse form of noise attributable to neutrons and γ ray was	Pulse discrimination that can discriminate γ ray and neutrons can be set	It was confir width and ris neutrons is p As a result, t as neutron is problems wit	width and rise time that pulse discrimination of γ ray and neutrons is possible. As a result, the probability of erroneously recognizing γ ray as neutron is around 2%, which indicate that there are no problems with using the detector in the field.					
evaluated.		gamma O	98.2%	1.8%				
SiC detector behind shield		neutron	6.2%	93.8%				
			gamma	neutron Predicted Class				



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector

c. Element test

Gamma-ray resistance performance verification test of SiC semiconductor detector (2/2)

Gamma-ray	resistance pe	ance								
Test details	Verification items and				Result					
	judgment	criteria								
(2) Impact of cumulative dose of $\boldsymbol{\gamma}$ ray	Confirm that the operates without	e detecto It any pro	r oblem	It was cont irradiation	firmed that th of total cum	ne detector pro ulative dose of	perly operates after 63kGv. (waveform			
Conducted at DCF (Dalton Cumbria Facility of Univ. of Manchester) which owns equipment that can irradiate ⁶⁰ Co up to 6540Gy/h.	after irradiation.			was confirmed with α source, count rate was confirmed) Trend of significant increase of leakage current was not confirmed even after irradiation, and it was determined that the detector could operate without problem even considering extrapolating such increase to 1MGy.						
Test board including the sensor chip and preamplifier was irradiated (installed				test result at DCF						
inside chamber in below picture)		Test No	Time	Dose (Gy/hr)	Duration (m)	Test Dose (Gy)	Accumulated Dose (Gy)			
Cumulative dose in the usable area of the		1	12:35	60	30	30	30			
facility is confirmed.		2	13:07	114	30	57	87			
		3	13:20	27	30	13.5	100.5			
		4	13:55	275.4	30	137.7	238.2			
		5	15:16	275.4	30	137.7	375.9			
		6a-f	15:51	275.4	60	275.4	651.3			
		7	17:00	275.4	990	4544.1	5195.4			
		- 8	9:30	275.4	30	137.7	5333.1			
		11a-c	12:05	275.4	30	137.7	5470.8			
		12	12:50	571.8	20	190.6	5661.4			
		13	13:27	1386	20	462	6123.4			
		14	14:35	2796	20	928	7051.4			
		15	15:28	6540	20	2180	9231.4			
		16	17:00	3270	990	53955	63226.4			
		17	11:25	120	30	60	63286.4			
		18	12:55	120	30	60	63346.4			
				Dose: Dose Duration: In Test Dose: Accumulate	e rate at irrac radiation tim Cumulative ed Dose: Tot	liation, e, dose of each te al cumulative do	st			

©International Research Institute for Nuclear Decommissioning Note) This test is a verification test conducted with PCV detailed inspection detector using the same SiC semiconductor detector.

- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector

d. Summary

Performance assessment (1/3)

O: Applicable, Δ : Conditionally applicable, \times : Not applicable

Category	ltem	Requirement specification	Fission ionization chamber	Improved B-10	B-10 proportional counter	Multicell He-3 proportional counter	B-10 corona	He-3 corona	SiC detector	CMOS detector
onment	Atmosphere environment	Underwater or 100% humidity	O IP68 ⁻¹	O IP68⁺¹	O Secured by water-sealing case	O Secured by water-sealing case	O IP68 ^{*1}	O IP68 ^{*1}	O IP68⁺¹	O IP68 ^{*1}
Applicability to field envirc	Gamma ray dose rate (average value)	10 Gy/h	O 1 × 10 ⁴ Gy/h ^{*2}	O 650 Gy/h	O 30 Gy/h ^{*3}	× ^{*3} (30 Gy/h ^{*4})	O 10 Gy/h*³ (700 Gy/h*⁵)	∆ 0.5 Gy/h ^{*3} Respond with shielding (2.5cm thickness)	O 100 Gy/h	O 1000 Gy/h
	Gamma ray dose rate (maximum value)	100 Gy/h	O Same as above	O Same as above	۵ Respond with shielding (1cm thickness)	× *3	ے Respond with shielding (2cm thickness)	A Respond with shielding (4.5cm thickness)	O Same as above	O Same as above
	Cumulative dose (usage period)	More than 0.6MGy (8 months)	O 1000 MGy	O >1MGy (including preamplifier) ^{*6}	O 1MGy⁵	∆ ^{∗8} 0.5 MGy	O 0.64 MGy ^{*9}	No data Estimated as the same level as B-10 corona	O 1 MGy ^{*10}	× 0.013 MGy
	Atmosphere temperature	50 °C	О 300 °С	O 100 ℃	O 50 °C	О 50 °С	O 50 ℃	O 50 °C	O 50 ℃	∆ 40 °C

*) See separate page for notes



- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - d. Summary

Performance assessment (2/3)

O: Applicable, Δ : Conditionally applicable, \times : Not applicable

Category	ltem	Requirement specification	Fission ionization chamber	Improved B-10	B-10 proportional counter	Multicell He-3 proportional counter	B-10 corona	He-3 corona	SiC detector	CMOS detector
Applicability to subcriticality measurement	Thermal neutron sensitivity*1	10 cps/nv	× (2.5 units) ^{*12} (5265cm3) ^{*11}	O (50 units) (192cm3) ^{*11}	O (6 units) (730cm3) ^{∗11}	O (0.5 units) (642cm3)* ¹¹	O (6 units) (693cm3)*11	O (1 unit) (120cm3)*11	O (-) ^{*13}	× (1250 units) ^{*11} (15708cm3)
	(Thermal neutron sensitivity of single alone)		4 cps/nv* ¹⁴ @100Gy/h	0.2 cps/nv @100Gy/h	1.7 cps/nv @0Gy/h 0.8 cps/nv @28.8Gy/h ^{*3}	21 cps/nv @0Gy/h ^{*4} 0.2 cps/nv @0Gy/h ^{*3}	1.9 cps/nv @0 Gy/h 0.6 cps/nv @8 Gy/h *3	11 cps/nv @0 Gy/h 7.2 cps/nv @0.6 Gy/h* ³	0.05cps/nv/c hip @70Gy/h ^{*13}	0.008 cps/nv @100Gy/h
	(Size of detector alone)		Ф80mm × 419mm ^{*14}	Ф7mm × 100mm	Ф25.4mm × 245mm	Ф76mm × 283mm	Ф25.4mm × 260mm	Ф25.4mm × 260mm	_*13	Ф20mm × 40mm
	Pulse width (detector)	Below 2000 nsec	0	O 1000 nsec (dependent on preamplifier method)	O Below 400 nsec	O Below 800 nsec	O 2000 nsec	O 1800 nsec	O ^{*15} 4000 nsec	-
	ldentifiable adjacent pulse interval (circuit system)	Below 100 nsec	0	O 1000 nsec (dependent on preamplifier method)	O 100 nsec	O 100 nsec	O 100 nsec	O 100 nsec	O 100 nsec	-
	Variation in pulse output delay time (circuit system)	Below 10 ns	0	To be confirmed	O 10 nsec	O 10 nsec	O 10 nsec	O 10 nsec	O 10 nsec	-

*) See separate page for notes


- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - d. Summary

Performance assessment (3/3)

O: Applicable, Δ : Conditionally applicable, \times : Not applicable

Category	ltem	Requirement specification	Fission ionization chamber	Improved B-10	B-10 proportional counter	Multicell He-3 proportional counter	B-10 corona	He-3 corona	SiC detector	CMOS detector
Applicability to remote handling	Unit size	Less than Φ700 mm×L1000 mm	O 5265 ^{*16} cm3	O 192 ^{*16} cm3	O 730 ^{*16} cm3	O 642 ^{*16} cm3	O 693 ^{*16} cm3	O 120 ^{*16} cm3	O*17	O 15708 ^{*16} cm3
	Unit weight	50-100 kg	O Shielding unrequired	O Shielding unrequired	∆ 1cm shielding	_	∆ 2cm shielding	× 4.5cm shielding	O Shielding unrequired ^{*18}	-
General evaluation		Not applicable (due to low thermal neutron sensitivity)	Applicable for subcriticality measurement in the field at 1F	Applicable for subcriticality measurement in the field at 1F	Evaluated as not applicable in the current situation (due to unstable operation)	Applicable for subcriticality measuremen t in the field at 1F	Not applicable (due to shielding thickness being too large)	Applicable for subcriticality measurement in the field at 1F	Not applicable (due to low thermal neutron sensitivity and low cumulative dose)	

*) See separate page for notes

It is evaluated as "Applicable" with O, Δ and if there is no $\,\times\,$





No.46

7. Implementation items

- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector

d. Summary

List of notes for the performance assessment

*1 Indicator for waterproof performance in JIS/IES standards. IP68 indicates that it can continue to be used underwater.

- *2 MIRION CFUG08
- *3 Measured value of gamma-ray resistance test

*4 Vendor evaluation

*5 Not verified for gamma ray of fuel debris in Digital Signal Processing mode in the vendor evaluation

*6 Irradiation test was conducted with dose at the preamplifier position at 2.3kGy/h using 60Co source. The results showed that it can be measurable at cumulative dose of 1MGy.

See FY2018 supplementary budget "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" Final Briefing Material No.840. *7 Evaluation value of only the sensor, since the preamplifier is installed inside PCV with shielding. It is a value based on model specifications of the same type, and irradiation has not been performed on the subject prototype.

*8 Evaluation value of only the sensor, since the preamplifier is installed inside PCV with shielding. It is a value based on model specifications of the same type, and irradiation has not been performed on the subject prototype.

*9 Evaluation value of only the sensor, since the preamplifier is installed outside PCV without shielding. It is a value based on the irradiation test result of a prototype of the same type in the relevant project "Project on Development of Fundamental Technology of Retrieval of Fuel Debris and Internal Structures (Development of Small-size Neutron Detector)", and irradiation has not been performed on the subject prototype.

*10 Vendor evaluation (evaluation from the result of factory inspection). Irradiation of 60Co up to 6540Gy/h was performed with irradiation equipment on the test board including the sensor chip and preamplifier (no additional shielding).

Usability from change in behavior after irradiation of total cumulative dose of 63kGy to 1MGy was confirmed.

*11 In order to compare achievable sensitivity when assembling the detector unit, volume for the number of detectors that yield 10cps/nv is indicated.

*12 Size-up may be subject to nuclear material regulatory restrictions

*13 Multiple sensor chips make up the detector, but sensitivity of each chip changes depending on composition. Typical values of the detector used in the KUCA test are indicated here. The total unit volume including shielding and moderator to obtain 10cps/nv is expected to be around 4000cm3.

*14 MIRION CFUG08

*15 Since it is configuration where multiple detectors independently conduct measurement and superimposed pulses can be separated by pulse waveform analysis, pulse width size does not have an adverse effect such as count omission.

*16 Shielding thickness, and moderator polyethylene size in the case of partial submersion processing are added, but requirement specifications are evaluated to be satisfied even when taking such into account

*17 In order to achieve 10cps/nv, total unit volume including shielding and moderator is expected to be around 4000cm3.

*18 Shielding is applied to the preamplifier as necessary, but the unit weight is expected to be less than 20kg even when taking such into account.



No.47

- 7.1 Development of on-site operating procedures
- 7.1.2 Confirmation of applicability to subcriticality measurement of neutron detector
 - d. Summary

[Result]

- Subcriticality measurement test was performed with KUCA on the three types of new neutron detectors (corona discharge type, SiC semiconductor type, multicell He-3 type), and it was confirmed that subcriticality can be measured with the same level of precision as the conventional B-10 proportional counter.
- Neutron measurement test was performed with the corona discharge type and multicell He-3 type in gamma ray environment, and the upper limit of neutron-discriminable gamma dose rate was confirmed. Additionally, it was confirmed that measurement is possible with the Feynman-α method in high gamma ray environment.
- Candidate neutron detectors to be used for subcriticality measurement in the field of fuel debris retrieval at 1F were tested and evaluated from the viewpoints of applicability to on-site environment, applicability to subcriticality measurement and applicability to remote handling, and the following was selected.
 - B-10 proportional counter (conventional / improved small-size)
 - B-10 corona discharge type
 - SiC semiconductor type



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents

[Issues]

Method of injecting non-soluble neutron absorbents to fuel debris is studied in a relevant project^(*). (Figure 1) In order to discuss operating procedures and throughput with the fuel debris retrieval method team, it is necessary to materialize the procedures for neutron absorbent spraying and injection equipment operation.

[Implementation items]

- Procedures to spray non-soluble neutron absorbents to fuel debris shall be organized in a step diagram for the fuel debris retrieval method and processing and collection equipment studied in the relevant project (*).
- Conditions of application to fuel debris processing scene, use of different absorbents, effectiveness of pre-injection, and downsizing and weight reduction of injection equipment shall be studied.
- Operability and feasibility of overall procedures shall be evaluated along with the operating procedures of criticality approach monitoring (7.1.1).

[Goals]

 Develop on-site operating procedures for non-soluble neutron absorbents, and provide information to be utilized by the fuel debris retrieval method team for design of the retrieval system and study of throughput.



Glass material Gd2O3 containing B•Gd particle

Water glass / Gd2O3 granulated powder

Example of injection equipment for solids (powder)



Figure 1 Drawing of injection of absorbents to fuel debris

(*) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - a. Organization of prerequisites

Method of use of different absorbents

- Various forms of fuel debris is assumed, such as rod-like and granular. Policies to use the optimum type of neutron absorbent for the respective fuel debris form have been established.
- Non-soluble neutron absorbents are divided between a solid type and a type that solidifies from liquid to solid over time (water glass type). Since the water glass type has fluidity and viscosity immediately after injection, it is effective when residual fuel (stump fuel) is forested vertically (Figure 5), when gaps in fuel debris are small (Figure 3, 4) and when fuel debris surface is largely uneven (Figure 6).



Figure 1 Use of solid type of absorber on pebble-like fuel debris



Figure 2 Use of solid type of absorber on rock-like fuel debris



Figure 3 Use of liquid to solidified type of absorber on pebble-like fuel debris



Figure 4 Use of liquid to solidified type of absorber on rock-like fuel debris



Figure 5 Use of liquid to solidified type of absorber on rod-like fuel debris



Figure 6 Use of liquid to solidified type of absorber on fuel debris with large unevenness





- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - a. Organization of prerequisites

Effect of mixing of absorbent and fuel debris

- Effect of absorbent when mixing fuel debris and absorbent by processing was analyzed and evaluated.
- Conditions were set for the retrieval area so that fuel debris (UO2) turns into grain due to processing and criticality occurs due to water entering the gaps.
- •Neutron multiplication factor becomes maximum when volume share of fuel debris is 20vol% and water is 20vol%. (Figure 1)
- •When mixing absorbent (Gd2O3; grain), subcriticality occurs at only 3vol% (Figure 2).
- ·It was indicated that particle size of absorbent is more effect at 1mm than 1cm.



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- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - a. Organization of prerequisites

Effect of arranging absorbent on fuel debris

- •Assuming ring-like absorbent in 20cm width, analytical evaluation was conducted to see if it had an effect to expand the retrieval area.
- When absorbents are arranged on the fuel debris surface in a collectable form, reactivity reduction effect is around 2%Δk at most, and the effect is small although the retrieval area can be expanded by 2cm (from 16cm to 18cm per side).



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - a. Organization of prerequisites

Entry of absorbent in gaps in fuel debris

- Solidified type (water glass / Gd2O3 granulated powder) of absorbent was dropped to the test body (granular lava; several cm in diameter) simulating fuel debris, and it was confirmed that absorbent penetrates deep into and adheres to gaps in fuel debris.
- Since particle size of solid type (granular) of absorbent is smaller than 1mm and its specific gravity is sufficiently larger than water at 3-4, said absorbent is considered to penetrate gaps larger than 1mm.

Viscosity [mPa ∙ s]	Injection amount [g]	Number of tests	Increased weight [g]	Increased weight (average) [g]
	1000	1	109.2	107.2
2000	1000	2	105.4	107.3
2000		1	19.5	04 F
	100	2	23.4	21.5

Measurement result of weight of neutron absorbent attached to granular lava

Neutron absorbent flowing underwater (injection amount 1000[g])

Granular lava





Figure 1 State of test of attachment to granular lava

From FY2015 supplementary budget "Subsidy Project of Decommissioning and Contaminated Water Management" (Development of Fuel Debris Criticality Control Technology)



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - a. Organization of prerequisites



(b) Appearance before and after test (top)

(*1) Easing of restrictions is expected if knowledge of fuel debris properties is gained in the future.

Mixing of fuel debris and absorbent by processing

Underwater chisel processing test was conducted with solid type absorbent sprayed on the test body (MCCI simulation) simulating fuel debris.
Absorbent on the surface layer was removed after crushing, and it was confirmed that absorbent had mixed into the area of crushed fuel debris fragments.



Figure 1 Mixed amount and required amount of absorbent

From FY2017 supplementary budget "Subsidy Project of Decommissioning and Contaminated Water Management" Sophistication of Method and System of Retrieval of Fuel Debris and Internal Structures (Technical development related to establishing the criticality control method)



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - b. Study of procedures

Overview of implementation items of 7.1.3.b

6 items raised as issues were studied, in discussing operating procedures and throughput with the fuel debris retrieval method team.

- - - Top retrieval method - - - - - - Side retrieval method (PLAN-A/B) - - - - - -

(1) Judgment of use of absorbent, procedures for recovery to Level 1 (\rightarrow No.55)

② Specifications of supply equipment of absorbent (\rightarrow No.56~57)

(3) Operation process of absorbent (\rightarrow No.58)

(4) Carry-in/out process of absorbent (\rightarrow No.59)

(5) Curing time of solidified absorbent (\rightarrow No.60)

- Top access method for transferring the unitized structure - -

⑥ Effectiveness
 evaluation of pre application of
 absorbents (→No.61)

No.54

IRID

- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - b. Study of procedures ① Judgment of use of absorbent, procedures for recovery to Level 1



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents

b. Study of procedures ② Specifications of supply equipment of absorbent (simplification of supply mechanism)

- Based on the required amount of absorbent (Table) revised in the relevant project^(*1), specifications of supply equipment of absorbent were revised to downsize the volume by about half. (Figure 1, 2)
- Supply equipment is different from the safety system, but since it is intended to maintain fuel debris at subcriticality, high reliability equivalent to that of fuel debris retrieval equipment is required.

Non-soluble neutron Absorbent weight^{(*2)(*3)} Absorbent absorbent (kg/day) capacity(*2)(*3) (liter/day) Gd₂O₃ particle 6.4 1.5 Water glass / Gd₂O₃ 6.3 3.0 granulated powder material

(*2) Fuel debris retrieval target per day is assumed at 300 kg.

It is assumed that 50% of the injected absorbent effectively functions.

(*3) Easing of restrictions is expected if knowledge of fuel debris properties is gained in the future (No.87).



Figure 1 Supply equipment for solid type (Gd₂O₃ particle)

(*1) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)



Table Required amount of absorbent

No.57

- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - b. Study of procedures ② Specifications of supply equipment of absorbent (confirmation of injection range)
- It was studied whether absorbent (glass material containing B•Gd) can be stably injected even if the injection mechanism is simplified by eliminating the motor and screw and directly attaching a valve to the hopper for the solid type absorbent injection equipment.
- Blockage was caused by hydrous absorbent with type with the smallest equipment outlet diameter, but it was confirmed that stable supply is possible with other types. Additionally, it was confirmed that the diffusion range tends to expand with more distance from the target location of input such as water depth becoming deeper and distance from equipment outlet to water surface becoming farther.
- It is considered better to have equipment outlet and injection location as close as possible to prevent increase in the amount of waste due to excessive injection during actual operation, so it is considered necessary to study detailed design including the fact that the equipment operates in an environment where the equipment itself is somewhat submerged.



Figure 1 Test equipment and used injection equipment type

Figure 2 Distribution of absorbent after injection (example)



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - b. Study of procedures③ Operation process of absorbent
- The process assumed for on-site operation of absorbent was studied (Table 1, 2).
- In the case of solidified type absorbent (water glass), there are more operation processes than the solid type since raw materials need to be mixed and injection equipment needs to be cleaned with water after use.

Table 1 Operation process of solid type $(Gd_2O_3 \text{ particle, glass material containing } B \cdot Gd)$

No.	Operation process	Details
1	Storage of absorbent	Glass material containing B \cdot Gd or Gd2O3 particle is stored in a state that can be immediately used as absorbent.
2	Weighing of absorbent	Measure the amount of absorbent required for work.
3	Loading of absorbent to injection equipment	Load absorbent to the hopper of the injection equipment.
4	Transfer of injection equipment to the field	Connect the injection equipment loaded with absorbent to the manipulator and move it directly above the fuel debris retrieval site.
5	Injection of absorbent to fuel debris	Inject absorbent from directly above the fuel debris retrieval site to spray it onto the fuel debris surface.
6	Return of injection equipment	After finishing injection, return the injection equipment from the fuel debris retrieval site to the prescribed location.

Table 2 Operation process of solidified type (water glass / Gd₂O₃ granulated powder)

No.	Operation process	Details
1	Storage of raw materials	Raw materials of water glass neutron absorbent are ① water glass, ② gadolinia granulated powder, ③ cement, ④ monosodium phosphate, and ⑤ water. Temporarily store them at the place of production. ④ and ⑤ can be mixed and stored as aqueous solution.
2	Weighing of raw materials	Respectively measure raw materials required for the amount of application of absorbent. Another method is to inject the prescribed amount of raw materials stored in a tank into the smelter using a dedicated squeeze feeder.
3	Mixing of raw materials ^(*)	Mix the raw materials with the smelter and produce water glass neutron absorbent.
4	Loading of absorbent to injection equipment	Load the water glass neutron absorbent produced with the smelter to the hopper of the injection equipment.
5	Transfer of injection equipment to the field	Connect the injection equipment loaded with water glass neutron absorbent to the manipulator and move it directly above the fuel debris retrieval site.
6	Injection of absorbent to fuel debris	Inject water glass absorbent from directly above the fuel debris retrieval site to apply it onto the fuel debris surface.
7	Return of injection equipment	After finishing application, return the injection equipment from the fuel debris retrieval site to the prescribed location.
8	Cleaning of injection equipment ^(*)	Clean the used smelter and injection equipment with water to wash away the adhered water glass absorbent. Collect and store cleaning water.

(*) Process in red is specific to the solidified type and is not required for the solid type.



7.1 Development of on-site operating procedures

7.1.3 Operating procedures for neutron absorbents

b. Study of procedures (4) Carry-in/out process of absorbent

Example of solid type (Gd particle) of top retrieval method (mobile cell method)

No.59

- Transport procedures from storage to use and collection were studied for the solid type absorbent (Figure 1, 2).
- In order to save the limited space of R/B, absorbent is stored and prepared in a different building used for maintenance. Transport equipment
 (mobile cell) developed in the relevant project^(*) is used to transfer the contaminated absorbent injection equipment between buildings. The mobile
 cell is intended to transfer unit cans storing fuel debris.
- The time required for transfer from preparing solid type absorbent to injecting it to fuel debris and collecting the injection equipment was estimated at around 1.5 hours for outbound trip + 3 hours for return trip (due to cleaning and air replacement in the transfer area) = 4.5 hours in total.
- If the injection equipment is prepared in the maintenance area in advance, injection can be performed within 1 hour.



(*) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020)

- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents
 - b. Study of procedures (5) Curing time of solidified absorbent
- Issues of solidified type absorbent (water glass / Gd2_O3 granulated powder) became evident with respect to transport time.
- It is necessary to adjust water glass absorbents so that they do not solidify before being injected (up to 2 hours). It is also necessary to make adjustments so that the absorbents do not solidify before cleaning residuals remaining inside the container after injection. (around 3 hours when cleaning with the dryer separator pit (DSP), around 6 hours when cleaning at the maintenance building)
- Range of viscosity that allows injection and cleaning is 0-5000[mPa·s].
- Test was conducted changing concentration (0.95-1.87wt%) of curing retardant (Table), and time until 5000[mPa·s] is reached was measured.
- Test results showed the prospect of delaying curing time by more than 5 hours (Figure). It is slightly short of 6 hours, but there is a change of achieving it if the transport process is studied in detail.

Table Raw materials of water glass / Gd₂O₃ granulated powder

Raw materials	Category
Main material	Unit 1 sodium silicate (Baumé specific gravity 55)
Curing agent	Ordinary Portland cement
Curing retardant	Monosodium phosphate
Water	Ion exchange water
Neutron absorbent	Gadolinium oxide



Figure Test result of elapsed time and viscosity after mixing of water glass / Gd₂O₃ granulated powder



- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents

b. Study of procedures (6) Effectiveness evaluation of pre-application of absorbents

- Applicability of non-soluble absorbent was studied as a method to respond to criticality risks outside the fuel debris processing area. Specific criticality risks including falling of equipment and fuel debris
- Pre-injection is proposed since local subcriticality cannot be measured
- Effectiveness in preventing criticality risks due to falling of fuel inside the large transport cask was evaluated as an example





→Reference urawing of storage inside transport cask





No.62

7. Implementation items

- 7.1 Development of on-site operating procedures
- 7.1.3 Operating procedures for neutron absorbents d. Summary

[Result]

 Regarding criticality prevention during fuel debris retrieval, image of onsite operation of items, such as judgment of use of non-soluble absorbent, procedures for recovery after use, supply equipment, transport procedures and pre-application, was materialized, and information was provided for the fuel debris retrieval method team to utilize it to study the retrieval system design and throughput.



7.2 Development of solidified absorbent technology (water glass)

[Issues]

Of non-soluble neutron absorbents developed in the relevant project^(*1), water glass type absorbent (hereinafter water glass material) is a viscous solidified material that <u>adheres to fuel debris surface by covering it</u> (Figure 1).

When water glass material is covering the fuel debris surface (Figure 2), there is concern of <u>inhibiting drying of</u> <u>fuel debris</u>.

[Implementation items]

- <u>Changes in moisture content of the test body covered by water glass</u> <u>material</u> was measured, with reference to the conditions of fuel debris drying process obtained in the relevant project^(*2).
 - Al₂O₃ porous ceramics which had the same drying behavior as UO_2 was used for the test body.
 - The test of FY2021 was conducted under conditions where the surface in contact with outside air is completely covered.
 - Target value of dry state is set to 0.5 vol% moisture content (equivalent to 0.1wt% fuel debris)

[Goals]

Acquisition of data on impact on the drying process of fuel debris with water glass material covered



Figure 2 Estimation of state of fuel debris in the storage canister, and simulated sample

Main results:

- Quantitative data on impact on the drying process of fuel debris with water glass (insoluble neutron absorber) attached is acquired.
- Adjustment with each PJ (related to retrieval work and drying treatment) at present can support the study on applicability in the field, which is necessary to grasp the impact of the retrieval work process and set drying conditions.
- By setting the target of the test body to "<u>0.3wt% moisture content (zeolite equivalent)</u>", the evaluation is equivalent to the fuel debris drying test conducted in the storage canister PJ and can feed back the results of this research.

(*1) Relevant project "Development of technology for further increasing the scale of retrieval of fuel debris and internal structures" (FY2019, FY2020) (*2) Relevant projects "Technology for identifying and treating properties of fuel debris" (FY2014), "Fuel debris characterization" (FY2015, FY2016)



No.63

Figure 1 State of injection of water

7.2 Development of solidified absorbent technology (water glass)a. Test plan Overview of solidified absorbent

- No.1 sodium silicate (water glass), etc. mixed with Gd₂O₃ granulated powder which is a neutron absorbing material (Table 1).
- Viscous liquid that solidifies over time.
- Applicable for uneven surface and slanting surface which differ from flat surface. Can be adhered to fuel debris (Figure 1).
- Neutron absorbent is used differently according to the various forms of fuel debris assumed such as rod-like and granular (Figure 2).

Table 1 Components of water glass type

Component	Chemical formula	Debris	of debris form	
No.1 sodium silicate	Na ₂ O · nSiO ₂ · xH ₂ O	Absorbent (solid)		í Alaha
Cement	SiO_2 , CaO, AI_2O_3 , Fe_2O_3 , CaSO ₄			
Monosodium phosphate	NaH ₂ PO ₄ ·2H ₂ O	Example of applying solid absorbent for pebble-like debris	Example of applying solidified absorbent for pebble-like debris	Example of applying solidified absorbent for rod-like debris
Water	H ₂ O	and the second s		135
Gadolinium oxide	Gd_2O_3	FTY		
		Example of applying solid absorbent for rock-like debris Figure 2	mple of applying dified absorbent for -like debris Examples of different u	e of applying solidified absorbent for ith large unevenness use of neutron absorbents
Figure 1 Example of v	vater glass absorbent covering	there is concern	of inhibiting drying	of fuel debris.



Water glass material can be used regardless

7.2 Development of solidified absorbent technology (water glass) b. Test preparation Overview of FY2021 test on impact assessment of fuel debris drying characteristics of solidified

Overview of FY2021 test on impact assessment of fuel debris drying characteristics of solidified neutron absorbent



Implementation items of FY2021

- Sample imaged with fuel debris covered with water glass material was prepared, and moisture and drying test system suitable for such evaluation was rebuilt.
- Test with the condition with water glass material entirely covering the sample (most conservative condition) was conducted to evaluate whether it is affected by water glass material.
- Property analysis of the water glass material itself was conducted for a quantitative evaluation of the impact of water glass material.



7.2 Development of solidified absorbent technology (water glass)

Reference: Past knowledge (investigation of drying behavior of fuel debris) (*1)

- In the past, a basic research on evaluation of drying behavior of fuel debris was conducted in cooperation with the collection and storage PJ.
- In the case of fuel debris, since many parameters (form, material, drying temperature, etc.) are predicted to affect drying behavior, focus was placed on the drying characteristic curve which enables uniquely centralized expression and comparison.
- Since it is applied to fuel debris that has various specific gravity and is not comparable by moisture content to study the optimum illustration method., a method to evaluate the relationship between drying speed and residual water was newly developed
- The mechanism leading to drying differs between water glass material and porous material, but impact on drying treatment can be inferred by clarifying the degree of impact on drying speed.



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(*1) Relevant projects "Technology for identifying and treating properties of fuel debris" (FY2014), "Fuel debris characterization" (FY2015, FY2016)



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7. Implementation items

7.2 Development of solidified absorbent technology (water glass) b. Test preparation Impact assessment of fuel debris drying characteristics of solidified neutron absorbent Results and challenges of FY2021 test
Aging curve of moisture content Porous material + water glass material

Results and challenges

- It was confirmed that saturated porous Al₂O₃ (simulated fuel debris) will dry even when water glass material is applied.
- It was also confirmed that water glass material itself will dry to the point moisture is not released.
- Drying behavior of water glass behavior is different to that of porous Al_2O_3 alone, which suggests the impact of bound water, etc. in water glass material.
- It was confirmed that the injection amount (water content) of water glass material also affects drying time.

The following was set as issues to be solved in FY2022

- Specific impact of water glass material on drying time of fuel debris cannot be observed just with pellet scale study
- Impact of scale-up shall be inspected to make the data easier to be incorporated in the study on drying process
- Since material balance of injection amount of water and weight after drying does not match, presence of water needs to be identified in terms of hydrogen generation
- Composition of water glass material after drying shall be elucidated for impact assessment of bound water, etc.
- Data on quantitative evaluation of drying time shall be acquired to investigate impact of injection amount (percentage) of water glass material



0.1wt% of fuel debris equivalent ≒ 0.3wt% of zeolite equivalent ≒ 0.5vol% Target moisture content: Less than 0.5vol%



In the test scale of FY2021, the drying test was subject to small-scale (10g order) test bodies. Thus, scale-up is essential to gain quantitative data.

Overview of scale-up test

Knowledge on scale-up factors shall be obtained by conducting bench scale test using the same evaluation method as that of laboratory scale.

(Pilot scale test which assumes actual operation is unsuitable for identifying factors that lead to changes in drying time)

• Flow of development of fuel debris treatment technology (drying)



7. Implementation items 7.2 Development of solidified absorbent technology (water glass)b. Test preparation

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(*1) Relevant projects "Technology for identifying and treating properties of fuel debris" (FY2014), "Fuel debris characterization" (FY2015, FY2016)



FY2021: Drying test in the most conservative conditions (laboratory scale)

FY2022: Drying test in the bench scale covering previous test conditions

Test ① Simulated fuel debris drying test

Investigation of drying behavior with the drying characteristic curve of zeolite applied to water glass material and measurement of amount of change of water content



- Test ② Crystal structure analysis of water glass material
- > Investigation on moisture properties with composition analysis of water glass material alone

7.2 Development of solidified absorbent technology (water glass)c. Element test
 Test ① Simulated fuel debris drying testTest result of zeolite + water glass material (5mm equivalent)

Zeolite alone



Appearance before and after drying test (Zeolite, water glass material 5mm equivalent)



Zeolite + water glass material (5mm equivalent)

7.2 Development of solidified absorbent technology (water glass)c. Element test Test ① Simulated fuel debris drying testEvaluation of impact on drying time



- It was confirmed that total amount of moisture contained in zeolite and moisture contained in water glass material affect the drying end time^(Note).
 - There was no inhibition of drying treatment caused by zeolite pores and blockage of gaps by water glass material.
 - As water content of the test body increased due to injection of water glass material, drying end time tended to increase compared to the condition without water glass material.
 (around 100min, when approx, 50% of volume of hydrous zoolite is replaced with water glass material)

(around 100min. when approx. 50% of volume of hydrous zeolite is replaced with water glass material)

 Surface area largely contributes to drying speed of the test body, which suggests the possibility of dependence on container dimensions.



7.2 Development of solidified absorbent technology (water glass)c. Element test
 Test 2 Crystal structure analysis of water glass material

Structural analysis using X-ray diffraction analysis (XRD) and nuclear magnetic resonance analysis (NMR) Evaluation of the drying characteristic curve indicates that the test body reaches a sufficiently dry state, but has confirmed residual weight of moisture.

Impact of residual weight of moisture shall be observed by clarifying the crystal structure of water glass material after drying treatment.

- For structural analysis of amorphous regions like water glass material, it is effective to use XRD analysis.
 - In the structural analysis of FY2022, measurement was conducted with analysis equipment suitable for analysis of amorphous regions.
- NMR, which is expected to evaluate electrochemical characteristics of analyzed sample surface, shall be used to observe the differences of crystal structure (network formation status of Si-Si bonding) that cannot be analyzed with XRD.
- Such structural analyses shall qualify and quantify amorphous regions in water glass material and help understand the state of bond water of water glass material.

(re-commissioned to Shizuoka Institute of Science and Technology)

⇒ Derived ratio of free water to bond water in water glass material and drying behavior of bond water shall be incorporated in the bench scale drying test results.





7.2 Development of solidified absorbent technology (water glass)c. Element test Test (2) XRD analysis result of crystal structure analysis of water glass material



Quartz: Compound similar to silica gel (SiO₂· nH₂O)

Tobermorite: Compound similar to tobermorite ($5CaO \cdot 6SiO_2 \cdot 5H_2O$)

- It was revealed to contain structures similar to silica gel and tobermorite, which are known as amorphous components existing in cement and concrete materials.
 - The test result shows that the main components of water glass material do not form strong crystalline bodies in underwater curing.
 - > The test result shows that there is very little crystalline water composition of water glass water material in underwater curing.



7.2 Development of solidified absorbent technology (water glass)c. Element test

Test ② Comparison of gas phase curing and underwater curing in NMR analysis of solids of crystal structure analysis of water glass material

No.74



NMR analysis result of water glass material after drying treatment

- NMR analysis revealed that peak intensity of high-dimensional structure of SiO₄ tetrahedral structure tends to increase.
- Amorphous nature of solidified materials including neutron absorbents has a structure that does not hold water strongly as it does not have a pore structure.
 - Water existing in water glass material is crystalline water, and results suggest that this crystalline water affects residual water content.
 - Crystalline water is known not to contribute to hydrogen generation under irradiation^{1), 2)}.
 - > It agrees with the results of γ ray irradiation test of water glass material conducted in the past³⁾.
- Existence of Gd₂O₃ does not affect the crystal growth process, and it was suggested that it is held in the crystal structure (maintains neutron absorption effect).



1) Masaomi Takahashi and Tomonari Fujita, "Effect of condition of the water in cement paste for hydrogen gas generated by gamma-ray irradiation", CRIEPI Report, L11020, 2013

2) Taisuke Tsukamoto, Hideaki Uchiyama, et al., "Development of Fuel Debris Canister (9) Measurement of the Quantity of Hydrogen by the Radiolysis of the Concrete Component Water under the γ-Ray Irradiation", Atomic Energy Society of Japan Fall Meeting, 2D18, 2017

3] Ryo Ishibashi, Toshiyuki Fujita, et al., "Criticality control technique development for Fukushima Daiichi fuel debris (25) Evaluation of Gamma-ray Irradiation Resistance of Insoluble Neutron Absorber", Atomic Energy Society of Japan Fall Meeting, 2H20, 2016

7.2 Development of solidified absorbent technology (water glass)

d. Summary

[Result]

- Test ① Simulated fuel debris drying test (scale-up test)
- It was confirmed that total amount of moisture contained in zeolite and moisture contained in water glass material affect the drying end time.
 - > There was no inhibition of drying treatment caused by zeolite pores and blockage of gaps by water glass material.
 - As water content of the test body increased due to injection of water glass material, drying end time tended to increase compared to the condition without water glass material.

Test 2 Crystal structure analysis of water glass material

- XRD analysis revealed that water glass material contains structures similar to silica gel and tobermorite, which are known as amorphous components existing in cement and concrete materials.
 - It was revealed that the main components of water glass material do not form strong crystalline bodies in underwater curing.
- NMR analysis revealed that amorphous nature of solidified materials including neutron absorbents has a structure that does
 not hold water strongly as it does not have a pore structure.
 - Components that contribute to crystal water of water glass material used in an actual environment tends to be trace amounts, and it is inferred that amount of water evaporation generated by heating of fuel debris due to such crystal water is also negligible.
- It was revealed that water glass material after drying has a composition with very little crystal water and also has a structure that does not hold water strongly.
 - It was confirmed that impact of radiolysis of water glass material on hydrogen generation is limited.

While water glass material does not inhibit drying of fuel debris, results showed that water glass material extends drying time. Thus, injection amount of water glass material shall be minimized.

It was also revealed that water glass material after drying treatment has a composition that does not generate hydrogen.



7.3 Overall summary of criticality control

- Based on the aforementioned studies of 7.1-7.2, requirements and monitoring procedures were organized as the overall summary of criticality control.
- Procedures according to the fuel debris retrieval method need to be selected for the criticality control method, but basic requirements that should be followed in all cases were organized to unify the approach.
- Developed examples of a representative method are presented to confirm whether the requirements are appropriate.
- Specific procedures and approach on judgment criteria were organized as supplemental explanation of criticality monitoring procedures incorporated in criticality control procedures.

Organized items

ltem		Requirement	
1) Purpose of criticality control	- Burness and goals of criticality control	Purpose of criticality control	
i) Purpose of childanty control	· Fulpose and goals of criticality control	Risk identification	
	 Elimination of criticality approach factors 	Maintenance of subcriticality	
		Prevention of reaching criticality	
2) Criticality prevention	Termination before reaching criticality even when criticality is	Criticality approach monitoring (predictive monitoring)	
	approached	Condition monitoring during work (constant monitoring)	
	Early detection of abnormality in deep subcritical state assumed	Long-term monitoring	
	Terminals the event hafens have mine a sident level with some	Detection of criticality	
3) Detection and termination of	I erminate the event before becoming accident level with early detection and termination, even when criticality occurs	Termination of criticality	
criticality	detection and termination, even when criticality occurs.	Measures after termination of criticality	
4) Implementation conditions of retrieval task	_	_	
5) Containment of retrieved material	-	-	



7.3 Overall summary of criticality control

Configuration

- Requirements common to all methods are organized (red box), and specific development of a representative method is shown on the right (examples shown in below table).
- Development to each method is indicated by each assumed risk

The lower (lateral) retrieval method is organized into three categories: no boric acid water, normal use of boric acid water, and interference removal.

·Criticality control procedures are attached as supplemental explanation

i) Subcriticality measurement, ii) Predictive monitoring, iii) Constant monitoring, iv) Detection and termination of criticality

	Requiremen	ts of criticality control	Specific development: Lower lateral retrieval work (without normal use of boric acid water)			
ltem	Requirement	Requirement (details)	Over-processing of fuel debris	Accumulation of chip powder	(omitted)	
2) Criticality prevention	2-1) Implement subcriticality maintenance measures (criticality prevention measures) for identified risks.	 2-1-1) (Criticality control personnel) Implement measures, ① Maintenance of subcriticality and ② Prevention of reaching criticality, for criticality prevention. 2-1-2) (Criticality control personnel) For ① Maintenance of subcriticality, implement measures such as preventing events that cause criticality approach, suppressing excessive criticality approach (suppressing addition reactivity) and maintaining subcriticality with neutron absorbents. 2-1-3) (Criticality control personnel) For ② Prevention of reaching criticality, implement measures to monitor criticality approach and stop work before reaching criticality. However, if criticality risk of retrieved objects is determined to be small in ①, or when enhancing the method to maintain subcriticality with neutron absorbents, it is acceptable to relax criticality monitoring. 	Retrieval work is the direct initiating event. Thus, implement measures for ① Maintenance of subcriticality and ② Prevention of reaching criticality in 2-2) and 2-3), respectively, considering the occurrence frequency is large. Selection of criticality control method for each target is shown in supplemental flow 1.	Retrieval work is the initiating event, but a long period of time is required for chip powder to accumulate from the generated amount to critical mass, and multiple condition superpositions are required such as concentrated accumulation in specific locations. Thus, implement implement measures for only ① Maintenance of subcriticality in 2-2), considering the occurrence frequency is small.	(omitted)	



7.3 Overall summary of criticality control

Purpose of criticality control

- "Accident" is defined as an event that causes an external impact exceeding the normal level
- The purpose of criticality control is to "prevent criticality, and prevent criticality from spreading to a criticality accident in case it occurs."

Identification of criticality risk

• Each method is required to analyze and identify risks and select the management method according to risk management (characteristics and magnitude or risk).

Item	Requirement	Requirement (details)
) Purpose of riticality control	1-1) The purpose of criticality control is to prevent criticality associated with fuel debris retrieval work, mitigate the impact in case criticality occurs, and prevent criticality accidents from occurring (prevent expanding to criticality accident). Earthquakes and other natural disasters are not subject to management under this requirement.	1-1-1) Definition of criticality accident Criticality accident is defined as an event that causes an impact exceeding the normal level due to additional release of radioactive materials due to criticality. In "normal level", exposure dose of the general public at site boundar is 1mSv/year and exposure dose of workers is 50mSv/year (Note 1). 1-1-2) Definition of criticality Criticality is when effective multiplication factor (keff) =1.0. However, considering uncertainty of criticality events, keff=0.95 is set as the
	1-2) Analyze risks associated with fuel debris retrieval, and implement measures to prevent criticality according	criticality control standard in studies of various measures (Note 2). 1-2-1) (Criticality control personnel) Identify criticality risks in subject work and methods.
	to the identified risk and measures to prevent spread to an accident in case criticality occurs.	1-2-2) (Criticality control personnel) Implement independent criticality prevention measures for the identified risks and measures to mitigate impact (prevent spread to criticality accident) when criticality occurs.

to fuel debris retrieval.

Note 2) In criticality evaluation, keff=0.95 is often set as the judgment criteria including various uncertainties.



7.3 Overall summary of criticality control

Level of defense in depth and approach of response measures

 In the approach on safety management, external impact is divided into levels (Note 1), in line with "accident" being defined as <u>an event that causes an external impact exceeding the normal level</u>. The policy is to establish means to determine the level and establish measures to prevent transition to a higher level and recovery measures in case of such transition.

Note 1) Since external impact may change due to various nuclear output even with the same criticality, accident standard is set as "event that causes an external impact exceeding the normal level" (that is to say, it does not mean that keff of the system is 1.0). However, consideration shall be given so that worker exposure is not exceeded.

Since it has been confirmed in past studies that external impact does not exceed the normal level by promptly detecting and terminating criticality, the goal of future equipment on-site procedures is to keep the level below the normal level even during criticality.

Level 1: Normal state

Level 2: Abnormal state (physical criticality has occurred but external impact is below the normal level)

Level 3: Criticality accident state (external impact has occurred)

 Above approach does not allow for criticality. Multiple measures are implemented for criticality prevention, as mentioned later.

C	0.0	1.0	2.0 Time (b)	3.0	4.0			
1.0E-14 1.0E-15				, 				
1.05-13		the normal level. (*1)						
1.00-12		supercriticality, but it is within						
1.0E-11		ILIS IEIA	lively large					
1.0E-10		It is rola	tivoly largo		1			
D 1.0E-09	1	attacheo	d to 1 CRD.					
1.0E-08	1	falling o	f 450kg of fu	uel debris				
0 1.0E-07	-	when criticality occurs due to						
1.0E-05	<u>+</u>							
E 1.0E-05	1							
S 1.0E-04	1							
1.0E-03	1	55.10 ····						
1.0E-02	Concerno.							
1.0E-01	No	rmal standard						
1.0E+00								
1.0E+01	Ace	cident standard						
1.0E+02								

(*1) Easing of impact is expected if knowledge of fuel debris properties is gained in the future.

Example of criticality exposure assessment (from FY2016 subsidy project results)



7. Implementation items 7.3 Overall summary of criticality control

No.80

Criticality control method during large-scale retrieval (no boric acid water) Case of normal use of boric acid water is shown in the reference material

Level 1 (Normal state) Prevention of abnormality = criticality prevention	Level 2 (Abnormal state) Prevention of spread of abnormality = prevention criticality accident	Level 3 (Accident) Mitigation of impact of accident	Functional requirement
keff 1.	0 •		
Site boundary dose	1m	Sv 5m	Sv
(Maintenance of subcriticality) (Prevention of reaching criticality)	Xe = 1 Bq/cm ³ (*)	e of criticality" in the current	Subcriticality is
Retrieval work restriction Failure (criticality approach occurs)	implementation plan		maintained within the control standards
Criticality measurement Predictive monitoring before processin Criticality approach monitoring (neutron cetector, gas sampling system)	g, constant monitoring during Failure (criticality occurs)	processing	Prevention of criticality by detecting criticality approach more than expected and stopping retrieval work
Success (recovery to normal state by removing areas	,		Prevention of
neutron absorbent) (According to the situation) Transition to normal use of boric acid water (**)	Detection of criticality (gas sampling system, neutron detector)	Failure (normal level exceeded)	external impact greater than the normal level, by detecting criticality and promptly
work in boric acid water	Emergency boric acid	1	stopping operation
Success (recovery to normal state by removing critical areas)	injection	Additional injection of boric acid water Decreased water level due to feedwater shutdown	Prevention of overexposure of general public
termination of criticality)	······································	Suppression of release of radioactive gas	and workers
7.3 Overall summary of criticality control

•Requirements corresponding to the level of defense in depth

Level	Required function	Method
Level 1 (Normal state) Prevention of abnormality = Criticality prevention	[Maintenance of subcriticality] →No.84 Subcriticality is maintained within the control standards	$\begin{array}{l} [\textbf{Subcriticality measurement}] \rightarrow \textit{No.84} \\ \text{Start of work upon quantitatively grasping margin until criticality} \\ [\textbf{Retrieval work restriction}] \\ \text{Suppression of excessive reactivity addition} \\ [\textbf{Maintenance of subcriticality with neutron absorbent}] (option) \rightarrow \textit{No.85} \\ \text{Normal use of boric acid water, non-soluble absorbent} \end{array}$
	[Prevention of reaching criticality] Prevention of criticality by detecting criticality approach and stopping retrieval work	[Criticality approach monitoring] (predictive monitoring / constant monitoring) → <i>Reference</i> Monitoring of conditions during retrieval work to stop work before reaching criticality
Level 2 (Abnormal state) Prevention of spread of abnormality = Criticality accident prevention	[Detection and termination of criticality] Prevention of external impact greater than the normal level, by detecting and promptly terminating criticality	[Detection and termination of criticality] Detection of criticality, termination with boric acid water injection
Level 3 (accident) Mitigation of impact of accident	Prevention of overexposure of general public and workers	[Securing of method to terminate criticality and method to mitigate external impact] Securing of supplementary method of termination and method to mitigate external impact, in case criticality is not terminated with boric acid water injection

Italic numbers indicate the location of relevant explanation



7.3 Overall summary of criticality control

Maintenance of subcriticality

Maintenance of subcriticality

Estimate current subcriticality, and start work upon confirming the margin until criticality

Elimination of criticality approach factors

Fuel debris processing volume limitation (requirement for criticality monitoring every 4,000cm3^(*1))

Requirements of criticality control		uirements of criticality control	Specific development: Lower lateral retrieval work (without normal use of boric acid water)	
ltem	em Requirement (details)		Over-processing of fuel debris	
2) Criticality preventior	2-2) Maintenance of subcriticality Eliminate factors of criticality approach and maintain subcriticality during work.	 2-2-1) Maintenance of subcriticality Start work upon confirming that subcriticality is maintained even when considering criticality is approached due to the implemented work. 2-2-2) Eliminate factors of criticality approach. Also select work methods that do not cause excessive reactivity addition. 2-2-2-1) Judgement criteria of excessive reactivity 	Before starting work, measure subcriticality using subcriticality measurement neutron detector loaded to the retrieval arm, and start work upon confirming that criticality does not occur even when considering reactivity addition associated with processing work. →Subcriticality measurement procedures Perform criticality monitoring (predictive monitoring) at every fixed processing volume (4,000cm3) ^(~1) . Take uncertainty of processing volume into consideration for the judgment criteria of predictive monitoring, and confirm validity of the margin at the phase before large-scale retrieval.	
		are maintenance of subcriticality (criticality is not reached with one addition), and amount of addition reactivity that does not cause immediate criticality in case criticality occurs.	monitoring) is set at less than 0.5%Δk/k.	

Other requirements are organized in similar ways. (predictive monitoring, constant monitoring, long-term monitoring, means of criticality detection and termination, response after termination, retrieval work conditions)

(*1) Easing of impact is expected if knowledge of fuel debris properties is gained in the future.



7.3 Överall summary of criticality control

•Make specific developments for each method, based on basic requirements.

Here are developments to normal use of neutron absorbent (boric acid water) and removal of interference objectives (non-fuel debris)

ltem	Requirement	Lower lateral retrieval work (without normal use of boric acid water)	Lower lateral retrieval work (normal use of boric acid water)	Interference removal inside pedestal
1) Purpose of criticality control	Omitted	Common		
	2-1) Implement subcriticality maintenance measures (criticality prevention measures) for identified risks.	Major risks: Excessive fuel debris processing / accumulation of chip powder / falling of heavy weight object / increase in water level	Criticality risk is reduced by boric acid water, and decrease in boron concentration becomes the major risk.	Since processing amount of fuel debris is small, risk is low, and falling of heavy weight object becomes the major risk.
2) Criticality	2-2) Maintenance of subcriticality Eliminate factors of criticality approach and maintain subcriticality during work.	Confirmation of margin with subcriticality measurement Limitation of fuel debris processing	Maintenance of subcriticality with boric acid water of concentration (6,000ppm) ^(*1)	Not applied since changes due to processing are small
prevention	2-3) Prevention of reaching criticality 2-3-1) Criticality approach monitoring (predictive monitoring)	Perform predictive morning at every fixed volume processing	Boron concentration monitoring and maintenance	Not applied since criticality risk is small
	2-3-2) Condition monitoring during work (constant monitoring)	Perform constant monitoring during processing	Perform continuous monitoring (constant monitoring) of neutron flux as backup of boron concentration monitoring.	Perform constant monitoring in view of unexpected situations.
	2-3-3) Long-term monitoring of the	Confirm that more changes than expected have not occurred before and after processing	-	-
		1/m, trend analysis of gas monitor indication value	Same as left (implemented with less frequency)	Same as left (implemented with less frequency)
3) Detection and termination of criticality	3-1) Detect criticality and promptly terminate criticality to prevent spread to a criticality accident.	Detection of criticality using the PCV gas monitor, termination of criticality due to emergency boric acid water injection	Same as left (detection of criticality, emergency injection of boric acid water)	Same as left (detection of criticality, emergency injection of boric acid water)
4) Implementation	4-1) Retrieval work must not be performed without above means functioning.	Same as left	Same as left	Same as left
conditions of retrieval task	4-2) Confirm that detectors used for monitoring have been appropriately calibrated and are operable.	Same as left	Same as left	Same as left
5) Containment of retrieved material	When storing retrieved objects in the transfer container (unit can, waste container), no special controls (measures such as monitoring and absorbent addition) are implemented.	The prerequisite is that appropriate criticality prevention is implemented for canisters with shape management.	Same as left	Same as left
	(*1) Easir	ng of impact is expected if knowledge of fuel de	bris properties is gained in the future.	

ltem

7.3 Overall summary of criticality control

Requirement

Subcriticality measurement procedures

Approach

1) Purpose / function	 Quantitatively grasp margin until criticality Set initial values of predictive monitoring (refine monitoring) 	 Start retrieval work upon confirming enough margin Expand the operational range by refining predictive monitoring (keff monitoring)
2) Measurement target	 Fuel debris submerged on a potentially critical scale (deposit inside pedestal, deposit of RPV lower head) 	 Can be handled with predictive monitoring and/or constant monitoring, if fuel debris processing scale is small
3) Measurement method	 Acquire neutron pulse time history data with the neutron detector near fuel debris, and apply the reactor noise method (Feynman-α method) 	•The Feynman- α method is a representative method for subcriticality measurement.
4) Measurement position	 Measure at least one point on a lump of fuel debris Re-measure if the retrieval position moves to a different lump Re-measure for every 10-20cm of digging 	 A lump of fuel debris has one subcriticality regardless of measurement position. Consider the possibility of composition and properties changing in the vertical direction such as MCCI fuel debris Neutrons are not necessarily emitted equally in all directions. Considering the detector could go behind the robot arm, simulate neutron measurement as necessary to determine detector placement.
5) Measurement time	 Assumed as 1 hour to several days depending on fuel debris form (size of system) 	 Estimate required measurement time based on neutron flux in the field and detector sensitivity
6) Uncertainty expected in the measurement result	 Expect uncertainty of subcriticality measurement (prompt neutron attenuation coefficient α) and uncertainty of conversion to keff (uncertainty of <i>ℓ</i>*) 	•Evaluate uncertainties with KUCA test results
7) Method of setting judgment criteria	 Confirm that criticality will not occur even when adding addition reactivity of several times of processing to the measured keff 	 Confirm that criticality does not occur with the first processing (for second time and onward, monitor criticality approach with predictive monitoring)
8) Measures when measurement cannot be performed	 Perform re-measurement increasing the measurement time Perform monitoring with 1/m instead of keff for predictive monitoring. In some cases, limit fuel debris processing volume to a small amount, and gradually expand the scale while performing predictive monitoring. 	 There are cases when measurement cannot be performed due to the size of fuel debris system and depth of subcriticality. A method is required to differentiate the causes of not being able to perform measurement between deep subcriticality (outside measurement range) and other reasons.
9) Alternative means	•Method of estimating subcriticality based on the percentage of Xe/Kr measured value on the PCV gas monitor	 It has been applied to Unit 1. It is desirable to perform measurement with the same method also at Units 2 and 3. Organize the position and necessity regarding the relationship with subcriticality estimation with neutron detector.
IKID	Predictive monitoring procedures are shown in the reference materials	

7.3 Overall summary of criticality control

Procedures on injection of non-soluble neutron absorbent

No.85

(Example of using near-critical fuel debris)

Item	Requirement	Approach
1) Purpose / function	 When it is confirmed by subcriticality measurement or predictive monitoring that fuel debris has approached criticality, deepen subcriticality to recover to a normal state. 	 Prevention of abnormality of criticality control method (no boric acid water) during large-scale retrieval is addressed.
2) Method	 Load non-soluble absorbent to the injection equipment, transport it above the target fuel debris with robot arm, and spray absorbent to the surface of fuel debris from above. 	 It order to use it locally, the prerequisite is that the scope of target fuel debris is known.
3) Injection frequency	• When necessary. (irregular)	 It is not used on a daily basis but used only when criticality approach is confirmed.
4) Injection position	 Local fuel debris area which has been confirmed to be approaching criticality 	 The target range is equivalent to daily fuel debris retrieval target of 300kg.
5) Injection time	Around 1 hour	 It is assumed that fuel debris retrieval is performed after injecting absorbent once a day. There is no urgency since it is Level 1 operation.
6) Method of setting injection amount	 Inject absorbent in the field upon setting the absorbent type (solid or solidified), injection amount and range. Confirm that the predetermined amount has been injected in the predetermined range. Perform subcriticality measurement after injection. 	 Issues are the approach of determining absorbent type, injection amount and range, and the confirmation method. (→effectiveness evaluation) Confirm the effect of injection with subcriticality measurement.
7) Response to shortage of injection amount	 If the predetermined amount and range are not achieved, repeatedly inject absorbent until they are satisfied. If effects are not confirmed by subcriticality measurement, change the absorbent type. 	 If solid type absorbents do not enter gaps in fuel debris and effects are not confirmed, effects may be obtained by injecting solidified type absorbents.
8) Effectiveness evaluation	 Perform criticality evaluation by simulating the observed fuel debris form, and confirm effectiveness of the absorbent. 	 It is recommended to simulate representative patterns of fuel debris forms assumed in advance. This analysis will be used to determine the injection amount in 6).
9) Response to equipment failure	 Position standby of non-soluble absorbent injection equipment as a condition of retrieval work. 	 Injection equipment failure itself is not a cause of criticality event. However, since it cannot be recovered to the normal state, fuel debris retrieval cannot be performed.
10) Other	 Establish procedures and judgment criteria for restarting fuel debris retrieval work after injecting absorbent. 	 Perform subcriticality measurement to confirm that keff is lower than the judgment criteria. The judgment criteria is a value smaller than 0.95, considering increase due to work.
IKID		

7.3 Overall summary of criticality control Summary

 Basic requirements were organized for criticality control during large-scale fuel debris retrieval.

Purpose of criticality control

Maintenance of subcriticality: Subcriticality measurement, retrieval work restriction,

normal use of neutron absorbent

- Prevention of reaching criticality: Criticality approach monitoring (predictive monitoring / constant monitoring), long-term monitoring
- Detection and termination of criticality: Detection of criticality, criticality termination due to emergency injection of boric acid water

• Criticality monitoring procedures were organized.

Subcriticality measurement, predictive monitoring, constant monitoring, detection and termination of criticality

Injection procedures for non-soluble neutron absorbent were organized.



7.4 Organization of issues for the future

Based on knowledge and issues obtained so far, issues to be addressed by full-scale retrieval are shown below while taking into account materialization of retrieval methods, internal information and new information of fuel debris forms.

(Table 1) Issues of methods

• It is necessary to take note that importance of issues to be solved changes according to the adopted method

ltem	Issues
Measures against falling of heavy weight object	 Identification of objects that could fall or collapse by confirming the situation inside. Considerations in the retrieval plan. Analysis of fuel debris properties (evaluation of criticality risk by grasping fragility "destructive energy") Evaluation of external impact when criticality occurs in a representative event
Measures against chip powder	 Accumulation potential evaluation by chip powder flow analysis based on generated amount of chip powder of each processing method, water level during large-scale retrieval, and coolant circulation amount (As necessary) Study on criticality detection method in vent pipe, S/C and torus room, and boric acid water injection method Evaluation of external impact of chip powder accumulation criticality
Throughput improvement	 Re-evaluation of criticality risk based on acquired information, study on simplification of management methods Study on possibility of acceleration of fuel debris retrieval with normal use of boric acid water
Other	 Confirmation of opinions of experts regarding fuel debris retrieval without boric acid water Criticality evaluation used as basis for limitations on methods, such as processing limits, water level limits, minimum critical mass and handled amount, is performed under conservative conditions taking uncertainties of fuel debris into account. If information such as fuel debris enrichment, heterogeneity and U content is obtained through future internal investigations and gradual retrieval, limitations of methods are expected to be mitigated by revising the excessively conservative conditions and re-studying criticality evaluation.



7.4 Organization of issues for the future

(Table 2) Issues of criticality monitoring

Item	Subcriticality measurement	Predictive monitoring	Constant monitoring	Detection and termination of criticality
Purpose / function	-	-	-	-
Method	 Preliminary verification of subcriticality measurement method (addition of verification data, blind test) Confirmation by pilot application in gradual retrieval Confirmation by estimation with the PCV gas monitor and by cross-checking with sampling analysis Judgment of whether the Feynman-α method should be applied (different use from 1/m) 	_	_	 Identification of the location when a criticality event other than the processing area is detected with the noble gas monitor, and proposed methods of countermeasures (measures of recovery to Level 1)
Measurement frequency	_	 Method of confirmation of processing volume in the field Confirmation of uncertainty of processing volume in the phase before large-scale retrieval 	_	_
Measurement position	 Development of monitoring plan with neutron flux distribution 	 Development of monitoring plan with neutron flux distribution Setting of measurement position considering impact of higher mode changes in neutron flux distribution 	_	_
Measurement time	 Setting of required count to obtain significant measurement results 	_	 Setting of average moving time according to equipment noise impact and neutron flux in the field 	_
Method of setting judgment criteria	Confirmation of validity of margin until criticality	_	Confirmation of preconditions (uncertainty of processing amount, processing volume, etc.) of effectiveness evaluation (confirmation in the phase before large-scale retrieval)	 Criticality detectability of vent pipe, S/C (study in cooperation with the system side)
Uncertainty expected in the measurement result	 Reduction of uncertainties attributable to ℓ (prompt neutron life) Examples: Criticality monitoring by α eigenvalue Impact after absorbent injection (analysis evaluation of KUCA test data) 	 Impact after absorbent injection (analysis evaluation of KUCA test data) 	_	_

7. Implementation items7.4 Organization of issues for the future (Table 2) Issues of criticality monitoring (continued)

ltem ()	Subcriticality measurement	Predictive monitoring Constant monitoring		Detection and termination of criticality
Response to deviation from judgment criteria	-	· Organization of procedures in the field		 Incorporation of shutdown capacity requirements into the boric acid water system design
Response to equipment failure	 Method of remote calibration using neutron source 	 Study of confirmation method of operation in the field and method of sensitivity calibration. Method of remote calibration using neutron source 	 Study of confirmation method operation in the field and ethod of sensitivity alibration. Study of confirmation method of operation in the field and method of sensitivity calibration. Method of remote calibration using neutron source 	
Other	 Measures when measurement cannot be performed Method of confirming validity of measurement with the Y- value curve Method of differentiating the causes of not being able to perform measurement between subcriticality depth (outside measurement range) and other reasons (not measurable). Incorporate in a framework so that the following procedures can be implemented without having criticality engineers. 	 Incorporate in a framework so that the following procedures can be implemented without having criticality engineers. 	 Incorporate in a framework so that the following procedures can be implemented without having criticality engineers. 	 Effectiveness evaluation Explanation of validity of PORCAS-F code^(*) Measures when criticality is not terminated To be studied with system and safety personnel including response with temporary equipment Incorporate in a framework so that the following procedures can be implemented without having criticality engineers.

(*) In order to confirm validity of PORCAS-F code, comparisons have been made with criticality transient experiment simulating solution system (TRACY experiment) and criticality transient experiment simulating fuel rod system (SPERT experiment). However, it is different from conditions with granular and lump-like forms, such as fuel debris, being cooled by natural convection. No cases of experiments simulating such criticality behavior have been found, and it is considered difficult to implement such experiment. A method of confirming validity without experiments is the method of conducting sensitivity analysis based on PIRT analysis and evaluating uncertainties expected in analysis results.



7. Implementation items7.4 Organization of issues for the future

(Table 3) Issues of neutron absorbent

ltem	Boric acid water (normal use, emergency injection)	Non-soluble neutron absorbent
Purpose / function	 For normal use and emergency. Development of usage method. 	 Development of respective usage methods as multiple usage is possible. (hereinafter is an example of the "case of using near-critical fuel debris")
Method	 Materialization of required equipment for normal and emergency injection Impact assessment of other equipment when using boric acid water, study of impact mitigation measures 	 Confirmation of remote operation combined with robot arm and prototype of injection equipment Confirmation by pilot application in gradual retrieval
Injection frequency	 (Normal use) Setting of operation concentration taking into account changes in concentration during operation (Emergency injection) Setting of representative concentration required for termination of criticality event 	 In order to set the absorbent injection frequency less than once a day, confirm the processing range of fuel debris retrieval per day and determine the capacity of injection equipment for sufficient amount of injection.
Injection position	 Study of criticality prevention measures at a position where coolant is not applied 	 Approach of determining absorbent type and injection amount for the fuel debris form in the processing range pre-planned per day Confirm that absorbent can be injected with the same positioning accuracy as fuel debris processing equipment. Establishment of method of confirming the injected range by visual inspection
Injection time	 (Emergency injection) Study of equipment to set and realize injection time that keeps criticality impact within the permissible range 	 Confirm the time until injection equipment is transported to the field by remote operation using robot arm. Additionally, confirm the time from collecting equipment after injection to completing preparations to load absorbents for next use.
Method of setting injection amount	 (Normal use) Setting of method and interval of boron concentration monitoring Backup with neutron flux constant monitoring 	 Prepare the approach of determining absorbent type, injection amount and range for the fuel debris form to be processed.
Response to shortage of injection amount	Method of maintaining concentration (study of response to decreased concentration)	None in particular (If the predetermined amount and range are not achieved, repeatedly inject absorbent until they are satisfied.)
Effectiveness evaluation	 Analyze criticality evaluation accordingly incorporating fuel debris sampling, confirm effectiveness of absorbent, and revise required 	s composition and form gained through internal investigation and concentration and injection amount
Response to equipment failure	 Study of alternative means of concentration monitoring, and boric acid supplementation method 	 Method of confirmation of operation in the field Method of collecting failed injection equipment and replacing with spare equipment
Other		Provision of information to waste PJ regarding waste after use

8. Specific goals for achieving the purpose of the project

(2) On-site operation methods fo absorbent technology	r criticality approach monitoring technology and neutron
① Development of on-site operating procedures	On-site operating procedures for criticality approach monitoring with neutron detectors are developed. Subcriticality measurement performance of neutron detectors is evaluated and incorporated in the on-site operation procedures. On-site operating procedures for non-soluble neutron absorbents are developed. (Target TRL at end: Level 4)
② Development of solidified absorbent technology	Data on impact on the drying process of fuel debris with solidified absorbent (water glass) attached is acquired. (Target TRL at end: Level 4)

Above goals have been achieved.



Reference

Reference 1 Criticality control method during largescale retrieval Reference 2 Basic requirements of criticality control Reference 3 Explanation of terms



Reference 1 Criticality control method during large-scale retrieval No.93 (normal use of boric acid water) Inject boric acid water in advance and perform retrieval work in boric acid water Level 1 (Normal state) Level 2 (Abnormal state) Level 3 (Accident) Prevention of spread of Prevention of abnormality = criticality prevention **Mitigation of impact Functional** abnormality = prevention criticality of accident requirement accident keff 1.0 Site boundary dose 1mSv 5mSv (Maintenance of subcriticality) (Prevention of reaching criticality) Subcriticality is **Drastically mitigated or eliminated** maintained Retrieval work restriction within the Failure control Maintenance of subcriticality with boric (criticality approach occurs) standards acid water, and monitoring of boron concentration Prevention of Only constant monitoring during processing criticality by detecting criticality Criticality approach monitoring (neutron detector) approach more than expected and stopping retrieval Failure (criticality occurs) work Stopping of retrieval work Success (recovery to normal state by removing areas approaching Prevention of **Detection of criticality** criticality, by deepening subcriticality with increased boron external impact (gas sampling system, concentration) greater than the neutron detector) normal level, by detecting criticality Failure (normal and promptly level exceeded) Emergency boric acid stopping operation injection Success (recovery to normal state by removing critical areas) V Additional injection of boric acid Prevention of water Decreased water level due to overexposure of Teedwater shutdown general public Success (recovery to normal state by removing areas where criticality occurred, after termination of criticality) Suppression of release of radioactive gas and workers

IKID

Reference 1 Criticality control method during large-scale retrieval (falling of heavy weight object)

Level 1 (Normal state) Prevention of abnormality = criticality prevention	Level 2 (Abnormal state) Prevention of spread of abnormality = prevention criticality accident	Level 3 (Accident) Mitigation of impact of accident	Functional requirement
keff 1. Site boundary dose	0 1m	Sv 5m	Sv
Maintenance of subcriticality) (Prevention of reaching criticality) Prevention of falling objects Maintenance of subcriticality with boric acid water • Monitoring of boron concentration	Failure (critic occurs)	ality	Subcriticality is maintained within the control standards
(Option)			
Success (recovery to normal state by	↓ Detection of criticality (Gas sampling system) ↓ Emergency boric acid injection	Failure (normal level exceeded)	Prevention of external impact greater than the normal level, by detecting criticality and promptly stopping operation
removing critical areas) Success (recovery to normal state by criticality occurred, after termination of	removing areas where f criticality)	Additional injection of boric acid water Decreased water level due to feedwater shutdown Suppression of release of radioactive gas	Prevention of overexposure of general public and workers



Reference 1 Criticality control method during large-scale retrieval (accumulation of chip powder)

Level 1 (Normal state) Prevention of abnormality = criticality prevention		Level 2 (Abnormal state) Prevention of spread of abnormality = prevention criticality accident	Level 3 (Accident) Mitigation of impact of accident	Functional requirement
keff	1	.0	- -	
(Maintenance of subcriticality) Collection of chip powder Prevention of outflow of cooling water by controlling water level Maintenance of subcriticality with boric acid water, and monitoring of boron concentration	(Prevention of reaching criticality)	nt monitoring)	<u>nsv</u> 5ms	Subcriticality is maintained within the control standards
(Option)	Criticality approach monitoring (neutron detector / PCV gas monitor) V Stopping of retrieval work	Failure (criticality occurs)		Prevention of criticality by detecting criticality approach more than expected and stopping retrieval
Success (recover approaching critic increased boron o Success (recover	y to normal state by removing areas cality, by deepening subcriticality with concentration) ry to normal state by removing critical areas)	Detection of criticality (Gas sampling system)	Failure (normal level exceeded)	work Prevention of external impact greater than the normal level, by detecting criticality and promptly stopping operation
Success (recovery to normal	state by removing areas where criticality occurred	d, after termination of criticality)	Additional injection of boric acid water Decreased water level due to feedwater shutdown Suppression of release of radioactive gas	Prevention of overexposure of general public and workers

Reference 1 Criticality control method during large-scale retrieval

Supplemental explanation of defense in depth

Response when criticality is not terminated regardless of boric acid water injection
 In past criticality control studies, criticality prevention as well as termination and
 mitigation of external impact of criticality were mainly studied, but it is desirable to
 prepare additional means for termination and mitigation of external impact when
 assuming criticality is not terminated regardless of boric acid water injection (specific
 studies are future issue).

In past discussions, there were comments saying to include mobility response instead of only addressing all additional means with equipment. Thus, it is desirable to analyze the possibility of criticality not being terminated upon studying equipment that achieves up to Level 2, and study measures including response using temporary equipment (future issue).

Measures after Level 3

Additional measures when criticality is not terminated regardless of boric acid water injection are tentatively indicated as Level 3 measures, but essentially, specific measures should be discussed upon studying definition of levels and events to be handled as a whole (including criticality) (after Level 4, it will be primarily addressed by operators).



Reference 2 Basic requirements of criticality control

Prevention of reaching criticality

-Below is an excerpt of predictive monitoring procedures.

Confirmation of whether criticality is approached and determining whether processing can be started, based on neutron flux measurement before processing.

	Requirements of critic	cality control	Specific development: Lower lateral retrieval work (without normal use of boric acid water)
ltem	Requirement	Requirement (details)	Over-processing of fuel debris
2) Criticality prevention	2-3) Prevention of reaching criticality Even if criticality is approached, monitor this and terminate the event before reaching criticality.	No details	Even if criticality is approached, implement measures to monitor this and terminate the event before reaching criticality. Details stipulated in 2-3-1)-2-3-3) ^(*) .
	2-3-1) Criticality approach monitoring (predictive monitoring)	2-3-1-1) Have means to monitor criticality approach during work	Use predictive monitoring neutron detector to be loaded to the retrieval arm.
		2-3-1-2) Measurement position of neutron flux is in a position that can appropriately monitor criticality approach.	Perform measurement on an "island" of fuel connected to the processing position.
		2-3-1-3) Measure neutron flux after fuel debris processing, and start work upon determining that criticality will not be reached in the next processing.	Measure neutron flux near the fuel debris processing position after fuel debris processing. Derive post-processing keff based on initial keff (subcriticality measurement result), and perform the next processing after confirming that it is below the judgment criteria. If keff cannot be measured, make the decision with inverse multiplication factor (1/m).
		2-3-1-4) Criticality approach monitoring frequency Measure intervals of addition reactivity stipulated in 2-2-2.	Perform criticality approach monitoring at every processing volume of 4,000cm3.
		2-3-1-5) Approach on judgment criteria of criticality approach Criticality control standards reduced by at least one count of addition reactivity, margin, measurement uncertain and evaluation uncertainty.	Judgment criteria is the value reducing criticality judgment criteria of keff (0.95) by 5 times the addition reactivity and measurement uncertainty as uncertainty of processing volume. Even in the case of 1/m, it is set from criticality control standards.
		2-3-1-6) If criticality approach judgment criteria is violated, perform cause analysis without starting the next work. Implement pre-developed procedures for work restart. Support procedures for work restart.	If criticality approach judgment criteria is violated, do not start the next work. Confirm whether criticality is approached by measuring subcriticality. Confirm the margin until criticality. Based on the result, study continuation of retrieval with smaller processing volume limit or switch to retrieval with normal use of boric acid water.

No.98

Predictive monitoring procedures

	tem	Requirement	Approach
1	1) Purpose / iunction	 Detect criticality approach, and stop processing work to prevent criticality. 1) Detect excessive criticality approach (keff=0.95, etc.) and stop work 2) Detect signs of criticality approach early on by grasping greater changes than expected 	 •2) is a preliminary analysis that captures greater changes than expected, in order to ensure 1).
	2) Method	 Measure neutron flux with the neutron detector near the processing position before fuel debris processing, extrapolate the amount of change from the previous measurement, and evaluate the possibility of reaching criticality in the next processing to determine whether processing can be started. 	 Criticality approach evaluation is based on the neutron source multiplication method generally used for criticality approach monitoring. Perform monitoring with 1/m if initial keff is not provided.
; 1	3) Measurement frequency	 Perform monitoring for each processing equivalent to the volume equivalent to addition reactivity 0.5%Δk/k. (for example, for every fuel debris processing volume of 4,000cm3) Set the processing volume taking into account uncertainties associated with processing (set uncertainties to be expected based on various information obtained in prior small-scale retrieval). 	 Addition reactivity 0.5%Δk/k is ①sufficiently small so that criticality is not reached in one processing, and ②set to prevent prompt criticality even when fuel debris processing is performed and reactivity is added from a critical state.
4	4) Neutron measurement position	 Lateral direction: Perform measurement within 50cm in the lateral direction (in the case of partial submersion method) from the processing position with the "lump" of same fuel debris as the fuel debris processing position. If lateral direction distance from the measurement position increases due to progression of processing, re- measure the reference count rate and move the measurement point. 	 Since the consecutive lumps of fuel debris have the same subcriticality and the same relative change in neutron flux, measurement points can be fixed regardless of the processing position. Distance of 50cm between the processing position and measurement point is a tentative value based on KUCA and other experiences. Neutrons are not necessarily emitted equally in all directions. Considering the detector could go behind the robot arm, simulate neutron measurement as necessary to determine detector placement.
	5) Neutron measurement time	•Measurement time is set so that uncertainties to be expected in judgment criteria are satisfied (assumed at around approx. 10min.)	•Statistical uncertainties of neutron flux count is set from the allowable uncertainty of the ratio to reference count rate with standard deviation σ = $$ (count value).
	6) Method of setting judgment criteria	When using keff) Set by reducing addition reactivity (0.5%Δk/k × margin) and uncertainty of the next processing from the criticality judgment criteria (0.95) When using 1/m) Set by reducing uncertainty from 1/m=0.5	See left
	7) Method of setting uncertainty	Statistical uncertainty of count value, reproducibility of measurement point (misalignment), impact of noise, fluctuations and attenuation of detector sensitivity, etc.	•Confirm the impact of noise in pre-mockup tests of retrieval equipment.
	8) Response to deviation from judgment criteria	 Stop processing work due to possibility of criticality approach. Re-measurement and subcriticality measurement extending the measurement time 	•Based on the result, study measures such as downsizing of processing size (shortening of monitoring interval) and application of neutron absorbent
	9) Monitoring method in normal state (deep subcriticality)	•Cause analysis when exceeding the normally expected magnitude of change •Convene criticality control experts and start analysis, while continuing retrieval work.	 Evaluate assumed changes according to initial keff. Work can be continued since criticality risks do not occur unless judgment criteria of 6) is not violated
	10) Response to neutron detector failure	 Position proper operation of neutron detector as a condition of retrieval work. 	•Neutron detector failure itself is not a cause of criticality event, but fuel debris retrieval cannot be performed since monitoring is not possible in the situation.
	11) Predictive monitoring when neutron absorbent is applied	 Criticality approach prevention (maintenance of subcritical state) when using neutron absorbent can be replaced with absorbent concentration management. 	 It is better to have regular neutron flux level monitoring as backup of concentration monitoring.

Reference 3 Explanation of terms

Term	Explanation
Reactor noise method	When measuring neutrons from nuclear fission reaction with a detector, neutron count rate within a certain time period is observed to fluctuate around the average value. Such temporal fluctuation in neutron count is referred to as reactor noise. The reactor noise method is a method that analyzes such noise to obtain core characteristics of the system.
Feynman−α method	It is a type of reactor noise method. It is a method that investigates the average and dispersion ration of the number of detected neutrons in a given time width T, and obtains prompt neutron attenuation constant α by plotting against time width T. It was proposed by R.P.Feynman.
Non-soluble neutron absorbent	Neutron absorbents have the property of absorbing neutrons to make it less likely to become critical. Non-soluble neutron absorbents have "non-soluble" or "insoluble" properties, in contrast to soluble neutron absorbents such as boric acid water.
Neutron detector - Corona discharge type	It is a detector with B10 which reacts with neutrons applied inside the cylindrical tube, and is characterized by applying voltage between the center line and cylindrical tube and operating in a high voltage region where corona discharge occurs. It is a technology by Russian company FEO (former RosRAO), and a prototype was made in a relevant subsidy project.
- SiC semiconductor type	It is a thin semiconductor detector doped with B which reacts with neutrons. Since it has low gamma-ray sensitivity and can reduce the shielding body, compact and lightweight high-sensitivity detector can be configured. It is a technology developed by British company IPL, and a prototype was made in a relevant subsidy project.
- Multicell He-3 type	It has a structure with multiple anode and cathode cores arranged inside the cylindrical tube filled with He-3 gas, and is designed aiming for high neutron detection sensitivity. A prototype was made in a relevant subsidy project by domestic company CETD.





END



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