Subsidy Project of Decommissioning and Contaminated Water Management

Development of Safety Systems
(Liquid/Gas Phase Systems and Criticality Control Technology)

Accomplishment Report for FY2021

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

August 2022

International Research Institute for Nuclear Decommissioning
(IRID)

(1) Liquid/Gas Phase Systems

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1. Purposes and goals of subsidy projects

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

It is assumed that nuclear fuel has melted along with the reactor internals at Tokyo Electric Power Company (TEPCO) Holdings, Inc. Fukushima Daiichi Nuclear Power Station (NPS) and exists in the form of molten fuel debris in the Reactor Pressure Vessel (RPV) and the Primary Containment Vessel (PCV).

The fuel debris accumulated inside the RPV and PCV is estimated to be currently in a sub-critical state; however the plant itself is in an unstable condition unlike its initial design, since the Reactor Building (R/B), RPV, PCV, etc. have been damaged due to the accident. Therefore, it is necessary to retrieve the fuel debris in order to maintain the sub-critical state, and to prevent diffusion of radioactive materials.

Against this background, this project is intended to conduct studies based on the “Mid-and-Long-Term Road-map Towards Decommissioning of TEPCO’s Fukushima Daiichi NPS” (hereinafter “Mid-and-Long-Term Road-map”), aiming towards the implementation of large-scale fuel debris retrieval in coordination with the engineering and project management activities undertaken by TEPCO. The development results of this project will be used in TEPCO’s engineering activities.

The purpose of this project is to advance the level of Japanese science and technology by supporting the development of technologies that contribute to the safe decommissioning of the Fukushima Daiichi NPS and handling of contaminated water, based on the Mid-and-Long-Term Roadmap and the “FY2021 Decommissioning Research and Development Plan” (the 86th Secretariat Team Meeting for Countermeasures for Decommissioning and Contaminated Water Treatment).

Specifically, technologies for removal of soluble α-nuclides that are considered to be eluted in circulating cooling water from fuel debris, treatment of RO-concentrated water (*), treatment of secondary waste, and on-site operating methods for criticality approach monitoring and neutron absorber will be developed.

***Liquid waste with a high impurity concentration generated by Reverse Osmosis filter (RO filter) removal of impurities from cooling water**

[Overall development goal]

Development goals are to develop element technology necessary for systems and to conduct tests for technology for ensuring safety for further increasing the scale of retrieval of fuel debris and the reactor internals based on the R&D results obtained so far.

1. Purposes and goals of subsidy projects

The material in this report deals with the following items.
The objective of each item’s implementation is shown.

(1) Liquid/gas systems

① Development of soluble α-nuclide removal technology

- Element test assuming fuel debris retrieval work

  Selection of candidates for adsorbents that can remove α-nuclides (Pu, U, etc.) during fuel debris retrieval work, taking into account the gas phase environment inside the PCV (nitrogen atmosphere).

② Development of Reverse Osmosis (RO)-concentrated water treatment technology

- Selection of adsorbent and condensing agent

  Selection of powder adsorbents and flocculants needed to treat RO-concentrated water through element tests and devise a treatment method for RO-concentrated water.

③ Development of secondary waste treatment technology

- Investigation of pretreatment technologies

1) Characterization of liquid waste generated from particle removal system

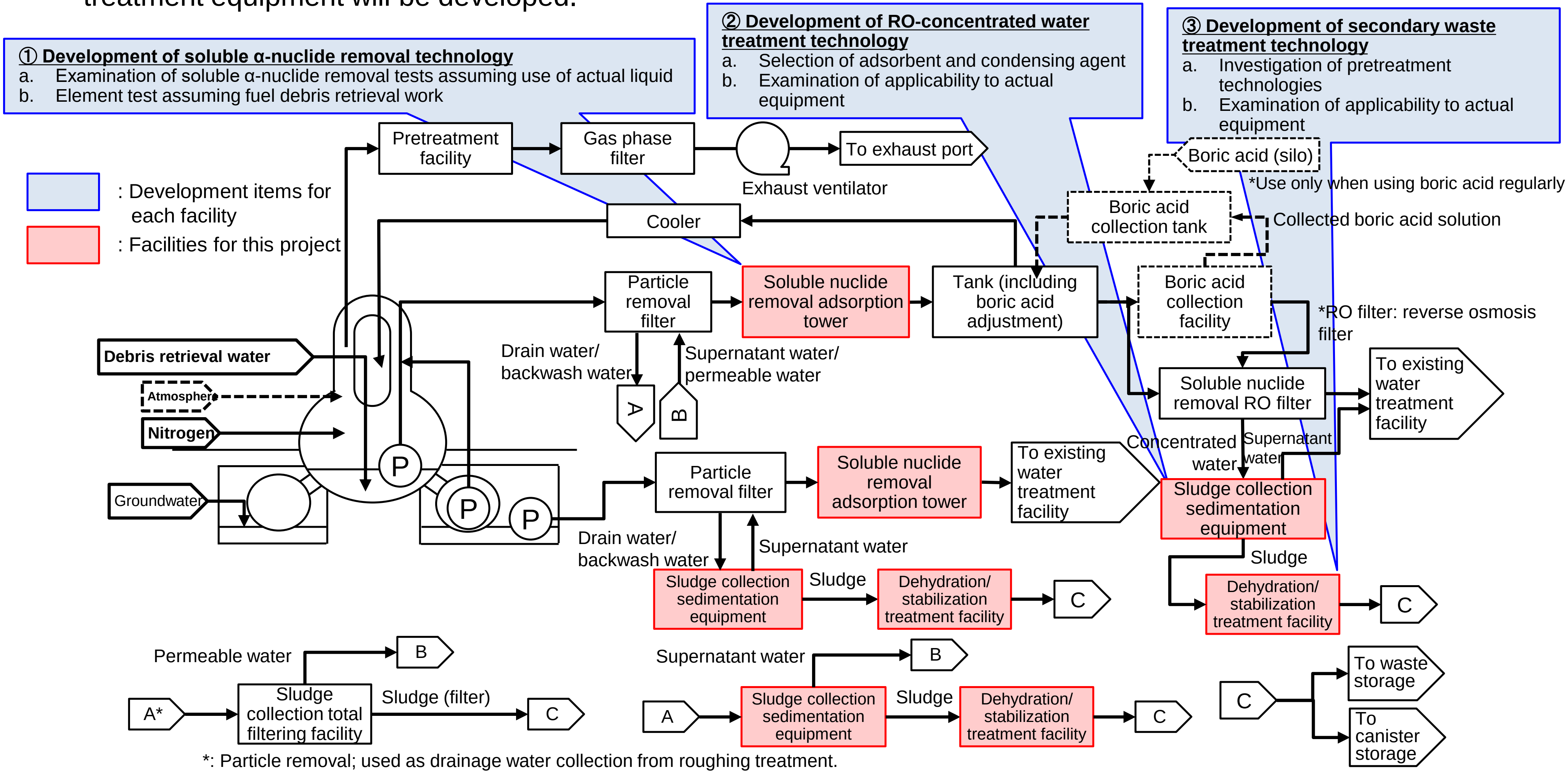
   Performance assessment tests on the filter components of the particle removal system are conducted. Then the properties of the filtered liquid waste discharged into the sludge collection system are consolidated.

2) Selection of dehydration technology for sedimentation sludge

   Applicable dehydration technologies for volume reduction treatment of sedimentation sludge from sludge collection equipment are selected from the literature research. Then a sludge treatment method is planned based on the results of the literature research and element tests.

2. Overview of subsidy projects

System configuration diagram of the liquid and gas systems under review for the subsidy project is shown.
In this year’s subsidy project, a soluble nuclide removal equipment and a sludge dehydration and stabilization treatment equipment will be developed.



① Development of soluble α-nuclide removal technology
a. Examination of soluble α-nuclide removal tests assuming use of actual liquid
b. Element test assuming fuel debris retrieval work

② Development of RO-concentrated water treatment technology
a. Selection of adsorbent and condensing agent
b. Examination of applicability to actual equipment

③ Development of secondary waste treatment technology
a. Investigation of pretreatment technologies
b. Examination of applicability to actual equipment

🟦 : Development items for each facility
🟥 : Facilities for this project

*Use only when using boric acid regularly

*RO filter: reverse osmosis filter

*: Particle removal; used as drainage water collection from roughing treatment.

Figure. Liquid and gas systems during fuel debris retrieval (based on subsidy projects)

Note: This conceptual diagram is an example of facility configuration (in the case of no leakage from PCV)

3. Previous fiscal year project results and remaining issues

① Development of soluble α-nuclide removal technology

🟨 : For examination in this project

No.	Items	Efforts and results of the previous project*	Remaining issues
1	Nuclides targeted for removal	The effects of powder generation for 15 nuclides of 5 elements (U, Np, Pu, Am, and Cm) were verified by tests. It was verified that the necessity of adsorption and removal of U is low because its solubility near neutral pH 5 to 9 under atmospheric conditions and the concentration in the PCV stagnant water are about two orders of magnitude lower than the announcement density.	Dissolution and adsorption behavior of α-nuclides for U, Np, Pu, Am, and Cm, respectively, assuming a nitrogen environment in the PCV
2	Quality of water to be treated	The evaluation was conducted assuming elution of seawater components, elution of concrete components, and injection of sodium pentaborate, a criticality inhibitor.	Effects of oil, coating, rust inhibitor, and non-soluble criticality inhibitor components
3	Treatment flow rate	If an adsorption/removal system is installed in the facility that circulates and cools the PCV stagnant water, a treatment flow rate of 10 m^3/h can be set to maintain a lower concentration of α-nuclides. If an adsorption/removal system is installed outside of the facility that circulates and cools the PCV stagnant water, only the volume of water increase from fuel debris retrieval work is treated so the water balance can be maintained if the treatment flow rate is 3.5 m^3/h or higher.	Setting of treatment flow rate considering facility operating schedule
4	Concentration of α-nuclides in assumed water quality	The dissolution behavior of each α-nuclide was determined based on the water quality that is assumed under conditions of elution of seawater components and the injection of sodium pentaborate, a criticality inhibitor. Regarding the effect of concrete elution, it was verified that evaluation in a nitrogen atmosphere similar to the PCV environment is necessary because in an atmospheric environment α-nuclides co-precipitate with the formation of calcium carbonate.	Dissolution and adsorption behavior of α-nuclides at high pH assuming elution of concrete components
5	Requirements for concentration reduction	The target DF for reduction of public exposure effects is 100, and the concentration reduction target for transferring to water treatment facility is the announcement density. (No progress)	Necessary DF setting accounting for the concentration of particulate α-nuclides Setting of concentration reduction targets for individual nuclides

* Subsidy Project of Decommissioning and Contaminated Water Management in the FY2018 Supplementary budgets
"Development of Technology for Retrieval of Fuel Debris and Reactor Internals"
(2) (ii) Technical development of treatment of fuel debris and deposit
① Removal technology for soluble nuclides in circulating cooling water
② Treatment technology for deposits collected from inside PCV
Hereinafter referred to as "the previous project."

3. Previous fiscal year project results and remaining issues

① Development of soluble α-nuclide removal technology

No.	Items	Efforts and results of the previous project	Remaining issues
6	Adsorbent application	As a result of the evaluation of α-nuclide adsorption performance by immersion tests, activated carbon, zirconium phosphate, and titanic acid were selected as candidate adsorbents for application. At this stage, since the adsorbent with a large equilibrium adsorption capacity was selected through immersion tests, it is necessary to verify its performance when applied to water flow treatment. In addition, since it is not possible to conduct high quantity α-nuclide circulation tests in a laboratory, the evaluation method is an issue.	Method for evaluating adsorption/removal performance when applied to water flow treatment
7	Adsorption tower configuration	Each tower has a series of activated carbon for removing colloidal α-nuclides, zirconium phosphate for adsorbing Pu, Am, Cm, Np, titanic acid for adsorbing Np, etc., with activated carbon at the top. Assuming merry-go-round operation, each tower is counted as two towers. The tower configuration should be reviewed taking into account the impact assessment of each nuclide simultaneously present in solution, as well as the removal performance and replacement frequency during the water flow treatment.	Interactive effects of multiple α-nuclides Effects of adsorption inhibitor components Review of tower configuration based on removal performance and replacement frequency during water flow treatment
8	Replacement timing /Replacement frequency	All of the adsorbents selected as candidates for application have low cesium (CS) adsorption performance, so the risk of replacement due to an increase in the tower surface dose is low. Although it is typical to conduct a circulation test to determine the replacement frequency due to the deterioration of adsorption performance, high α-nuclide quantity makes such a test impossible in a laboratory setting. Thus, the evaluation method remains an issue.	Evaluation method of adsorbent replacement frequency
9	Replacement method	Whether replacing the entire adsorption tower or only the adsorbent, in either case it is necessary to design equipment according to the treatment policy of the used adsorbent (long-term storage for the time being).	Operation at the time of replacement Handling of used adsorbents
10	Tower size	At a treatment flow rate of 10 m^3/h, the size is about the same as the adsorption tower of the multi-nuclide removal system (outside diameter: approx. 1 m, height: approx. 2.5 m).	Adsorption tower design with input of shielding policy, replacement method, etc.

3. Previous fiscal year project results and remaining issues

② Development of RO-concentrated water treatment technology

③ Development of secondary waste treatment technology

🟨 For examination in this project

No.	Items	Efforts and results of the previous project	Remaining issues
1	Water quality for treatment	The assumed water quality of the drain water and backwash water discharged from the particle removal system, which is the input water to the sludge collection system, was regulated based on the results of element tests and filter tests of the fuel debris retrieval method project. However, since the **intermediate filter of the particle removal system has not yet been selected**, it is necessary to select candidate components and evaluate the properties of the generated liquid waste. In addition, in the previous fiscal year's element tests, evaluation was only possible when spherical particles were passed through, and it is necessary to **evaluate the filter behavior and the properties of the generated liquid waste when non-spherical particles are passed through**.	Candidate components for the intermediate filter will be selected by literature research. Applicability will be verified through element tests and the properties of the generated liquid waste should be determined.
			Element tests will be conducted to assess the impacts on the particle removal system when treating non-spherical particles. If it is assessed that there is a predominating impact on the behavior of component differential pressure increase, recovery rate, or the amount of liquid waste generated, this will be reflected in the assumed input water quality of the sludge collection system.
2	Treatment flow rate	The treatment flow rate requirement of the sludge collection system was assessed based on the operating schedule of the fuel debris retrieval method, the filter test results, and the liquid system flow rates.	If the assumed input water quality of the liquid system changes as a result of future studies, this will be reflected in the treatment flow rate requirement of the sludge collection system.
3	Treatment method	Coagulation-precipitation treatment was selected as the treatment method for filtered liquid waste and RO-concentrated water, and usable flocculants were selected through element tests. The use of sedimentation tanks in the coagulation-precipitation treatment procedures were also examined. It is necessary to **examine the applicability to actual equipment for the flocculation sedimentation treatment of RO-concentrated water** in the case of sedimentation tank usage. In addition, it is necessary to investigate treatment procedures to simplify operations and reduce the amount of sludge generated.	Adsorption using powdered adsorbent and flocculation-precipitation using flocculant as treatment methods for RO-concentrated water will be selected. Usable powdered adsorbents and flocculants will be selected.
4	Sedimentation sludge properties and the amount generated	The amount of sedimentation sludge generated was estimated based on flocculation sedimentation test. The amount of sedimentation **sludge generated from RO-concentrated water was rated high** due to the high treatment flow rate and the high moisture content of sedimentation sludge, and there were concerns about the complexity of the discharge process and the load increase at the discharge point. Therefore, it is necessary to **reduce the amount of the sedimentation sludge**.	In order to reduce the amount of sedimentation sludge generated, each step of the RO-concentrated water treatment procedures will be reviewed. Through element tests, the effects of reducing the quantity of flocculant and powder adsorbent, and optimization of stirring conditions, etc. will be examined.
			The application of dehydration treatment to sedimentation sludge will be analyzed, applicable dehydration technology will be selected via literature research, and the applicability to actual equipment will be assessed through element tests.

3. Previous fiscal year project results and remaining issues

② Development of RO-concentrated water treatment technology

③ Development of secondary waste treatment technology

No.	Items	Efforts and results of the previous project	Remaining issues
5	Supernatant liquid properties	The quantity of suspended solids (SS) and other components remaining in the supernatant after solid-liquid separation were sorted.	The required acceptance conditions of the soluble nuclide removal equipment and the existing water treatment facility will be sorted out, and the necessity of neutralization treatment and removal of neutralized salt and trace amounts of residual SS components will be examined.
6	Operating methods	The process of coagulation-precipitation treatment of filtered liquid waste and RO-concentrated water using a sedimentation tank was examined based on the operating schedule of fuel debris retrieval method and element test results. An operating cycle in which one batch process is completed per day was examined.	The results of this year’s element tests will be applied to the coagulation-precipitation treatment process studied in the previous project.
7	Equipment specifications	Equipment tests verified the operability of the sedimentation tank and each component. Since large particles were caught when opening and closing the gate valve, it is necessary to reconsider the specifications of the gate valve.	In element tests using RO-concentrated water treatment test equipment to be conducted from this fiscal year onwards, each mechanism of the sedimentation tank will be re-evaluated. In addition, valves that can be applied to sedimentation tanks will be examined.
8	Methods for discharge of sedimentation sludge	The discharge method of sedimentation sludge using a sludge collection container was examined, and through an equipment test confirmed that this method can be applied to actual equipment. It was verified that it is possible to insert piping from the top of the sediment tank and remove sedimentation sludge by suction pump. In the future, the required acceptance conditions of the sedimentation sludge discharge point will be sorted out and sludge canister size and applicability of dehydration treatment will be examined.	When the sedimentation sludge is treated and disposed of as waste, it is discharged to the waste line, so it is necessary to consider the final disposal of sedimentation sludge and apply pretreatment. In the future, if a policy for the final disposal of sludge is presented, the necessary pretreatment within the liquid system will be examined and applied to the sedimentation sludge discharge method. When sedimentation sludge is stored in canisters, it is discharged to the canister line, so it is necessary to apply pretreatment to satisfy the requirements of long-term storage. The requirements for long-term storage of sedimentation sludge will be sorted out and the shape and moisture content of the sludge collection container will be examined.
9	Handling of sedimentation sludge	Assumed properties of sedimentation sludge and the amount generated were arranged based on the results of element tests and the liquid system requirements.	Assumed properties of sedimentation sludge will be shared to both the waste project and the canister project. Issues will be identified respectively to each project.

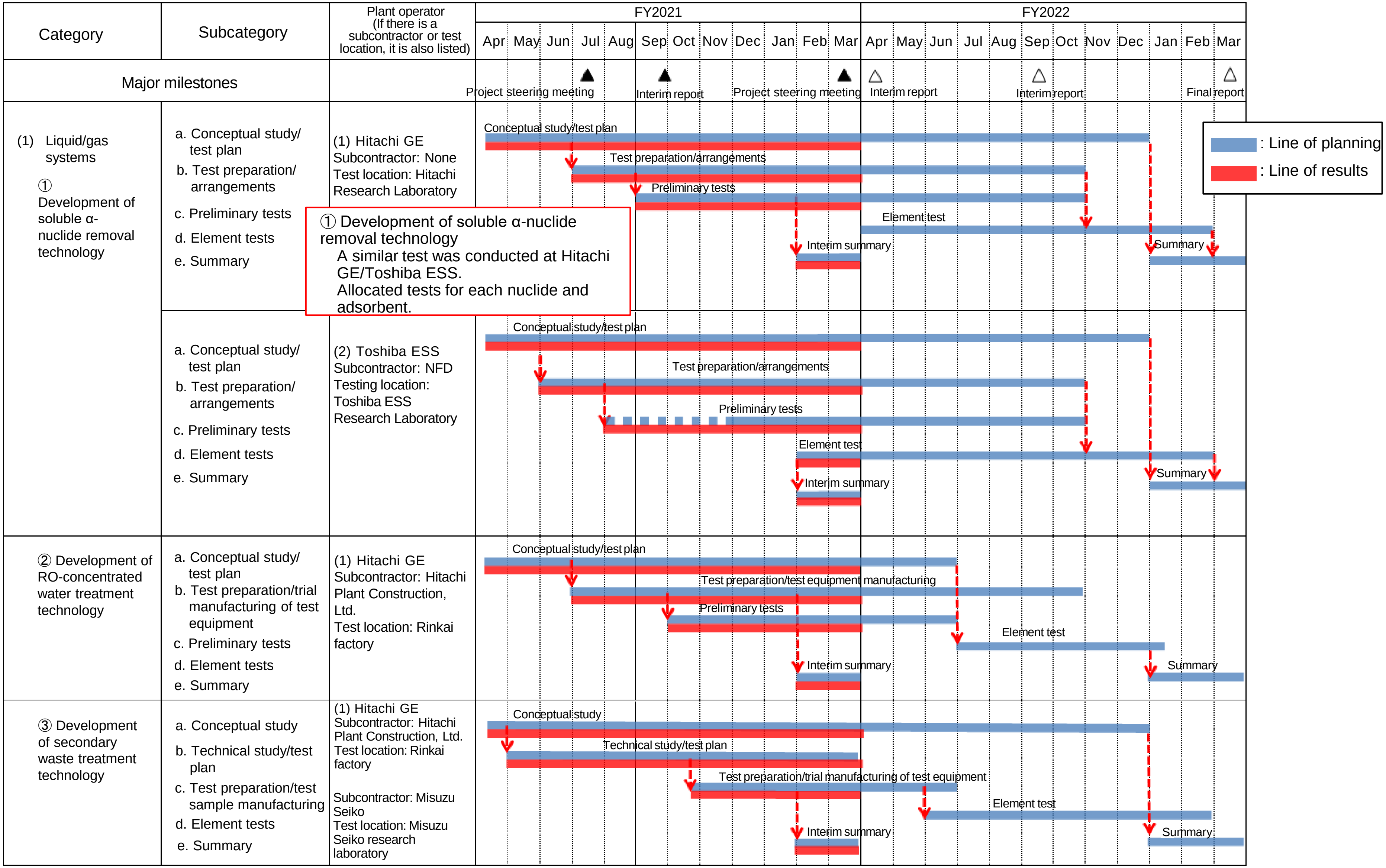
4. Input/output information

This project will be proceeded in collaboration with related projects such as the Development of Technologies for Containing, Transfer and Storage of Fuel Debris project and the R&D for Treatment and Disposal of Solid Waste project.

ID	Demand-side project	Provider-side project	Contents (overview)	Information use
1	Development of Safety Systems (Liquid/Gas Systems, Criticality Control Technology)	Development of Technologies for Containing, Transfer and Storage of Fuel Debris	Acceptance conditions for canisters	Examination of stabilization treatment technology for secondary waste
2	Development of Safety Systems (Liquid/Gas Systems, Criticality Control Technology)	R&D for treatment and disposal of solid waste	Acceptance conditions for waste storage containers	Examination of stabilization treatment technology for secondary waste
3	Development of Safety Systems (Liquid/Gas Systems, Criticality Control Technology)	Development of Analysis and Estimation Technology for Fuel Debris Characterization	Results of examination on formation behavior of fuel debris particles	Examination of properties of particles in circulating cooling water
4	Development of Technologies for Containing, Transfer and Storage of Fuel Debris	Development of Safety Systems (Liquid/Gas Systems, Criticality Control Technology)	Properties of sedimentation sludge generated from liquid gas system	Identification of issues in the handling of fuel debris in slurry or sludge form
5	R&D for treatment and disposal of solid waste	Development of Safety Systems (Liquid/Gas Systems, Criticality Control Technology)	Properties of sedimentation sludge generated from liquid gas system	Identification of issues in the treatment and disposal of radioactive waste in slurry or sludge form

*Also planned to discuss collaboration with TEPCO in the future.

5. Implementation schedule



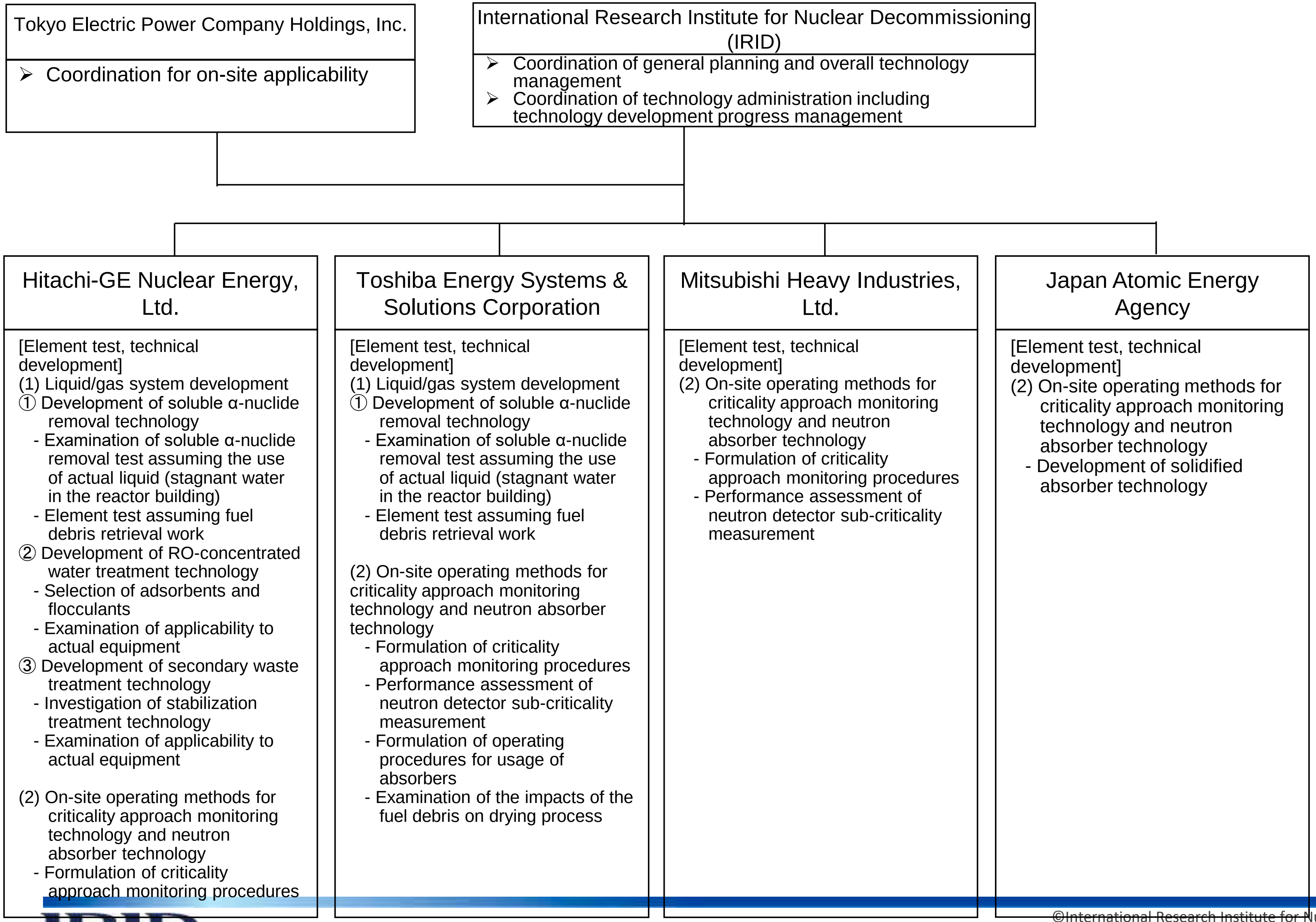
Category	Subcategory	Plant operator (If there is a subcontractor or test location, it is also listed)	FY2021–FY2022 schedule
Major milestones			▲ Project steering meeting (Jul 2021) ▲ Interim report (Sep 2021) ▲ Project steering meeting (Mar 2022) △ Interim report (Apr 2022) △ Interim report (Sep 2022) △ Final report (Mar 2023)
(1) Liquid/gas systems ① Development of soluble α-nuclide removal technology	a. Conceptual study/ test plan b. Test preparation/ arrangements c. Preliminary tests d. Element tests e. Summary	(1) Hitachi GE Subcontractor: None Test location: Hitachi Research Laboratory	Conceptual study/test plan Test preparation/arrangements Preliminary tests Element test Interim summary Summary
	a. Conceptual study/ test plan b. Test preparation/ arrangements c. Preliminary tests d. Element tests e. Summary	(2) Toshiba ESS Subcontractor: NFD Testing location: Toshiba ESS Research Laboratory	Conceptual study/test plan Test preparation/arrangements Preliminary tests Element test Interim summary Summary
② Development of RO-concentrated water treatment technology	a. Conceptual study/ test plan b. Test preparation/trial manufacturing of test equipment c. Preliminary tests d. Element tests e. Summary	(1) Hitachi GE Subcontractor: Hitachi Plant Construction, Ltd. Test location: Rinkai factory	Conceptual study/test plan Test preparation/test equipment manufacturing Preliminary tests Element test Interim summary Summary
③ Development of secondary waste treatment technology	a. Conceptual study b. Technical study/test plan c. Test preparation/test sample manufacturing d. Element tests e. Summary	(1) Hitachi GE Subcontractor: Hitachi Plant Construction, Ltd. Test location: Rinkai factory Subcontractor: Misuzu Seiko Test location: Misuzu Seiko research laboratory	Conceptual study Technical study/test plan Test preparation/trial manufacturing of test equipment Element test Interim summary Summary

① Development of soluble α-nuclide removal technology
A similar test was conducted at Hitachi GE/Toshiba ESS.
Allocated tests for each nuclide and adsorbent.

🟦 : Line of planning
🟥 : Line of results

*Hitachi GE: Hitachi-GE Nuclear Energy, Ltd.
*Toshiba ESS: Toshiba Energy Systems & Solutions Corporation

6. Project organization



Tokyo Electric Power Company Holdings, Inc.

- Coordination for on-site applicability

International Research Institute for Nuclear Decommissioning (IRID)

- Coordination of general planning and overall technology management
- Coordination of technology administration including technology development progress management

Hitachi-GE Nuclear Energy, Ltd.

[Element test, technical development]
(1) Liquid/gas system development
① Development of soluble α-nuclide removal technology
  - Examination of soluble α-nuclide removal test assuming the use of actual liquid (stagnant water in the reactor building)
  - Element test assuming fuel debris retrieval work
② Development of RO-concentrated water treatment technology
  - Selection of adsorbents and flocculants
  - Examination of applicability to actual equipment
③ Development of secondary waste treatment technology
  - Investigation of stabilization treatment technology
  - Examination of applicability to actual equipment

(2) On-site operating methods for criticality approach monitoring technology and neutron absorber technology
  - Formulation of criticality approach monitoring procedures

Toshiba Energy Systems & Solutions Corporation

[Element test, technical development]
(1) Liquid/gas system development
① Development of soluble α-nuclide removal technology
  - Examination of soluble α-nuclide removal test assuming the use of actual liquid (stagnant water in the reactor building)
  - Element test assuming fuel debris retrieval work

(2) On-site operating methods for criticality approach monitoring technology and neutron absorber technology
  - Formulation of criticality approach monitoring procedures
  - Performance assessment of neutron detector sub-criticality measurement
  - Formulation of operating procedures for usage of absorbers
  - Examination of the impacts of the fuel debris on drying process

Mitsubishi Heavy Industries, Ltd.

[Element test, technical development]
(2) On-site operating methods for criticality approach monitoring technology and neutron absorber technology
  - Formulation of criticality approach monitoring procedures
  - Performance assessment of neutron detector sub-criticality measurement

Japan Atomic Energy Agency

[Element test, technical development]
(2) On-site operating methods for criticality approach monitoring technology and neutron absorber technology
  - Development of solidified absorber technology

Development project teams in cooperation with this project

- Development of Fuel Debris Retrieval Method
- Development of Technologies for Containing, Transfer and Storage of Fuel Debris
- R&D on Solid Waste Treatment and Disposal
- Development of Analysis and Estimation Technology for Fuel Debris Characterization
- Development of Technology for Detailed Investigation inside PCV
- Development of Technology for Investigation inside the RPV
- Development of Analysis and Estimation Technology for Fuel Debris Characterization
- Development of Technology for Sorting of Fuel Debris and Radioactive Waste

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Development history

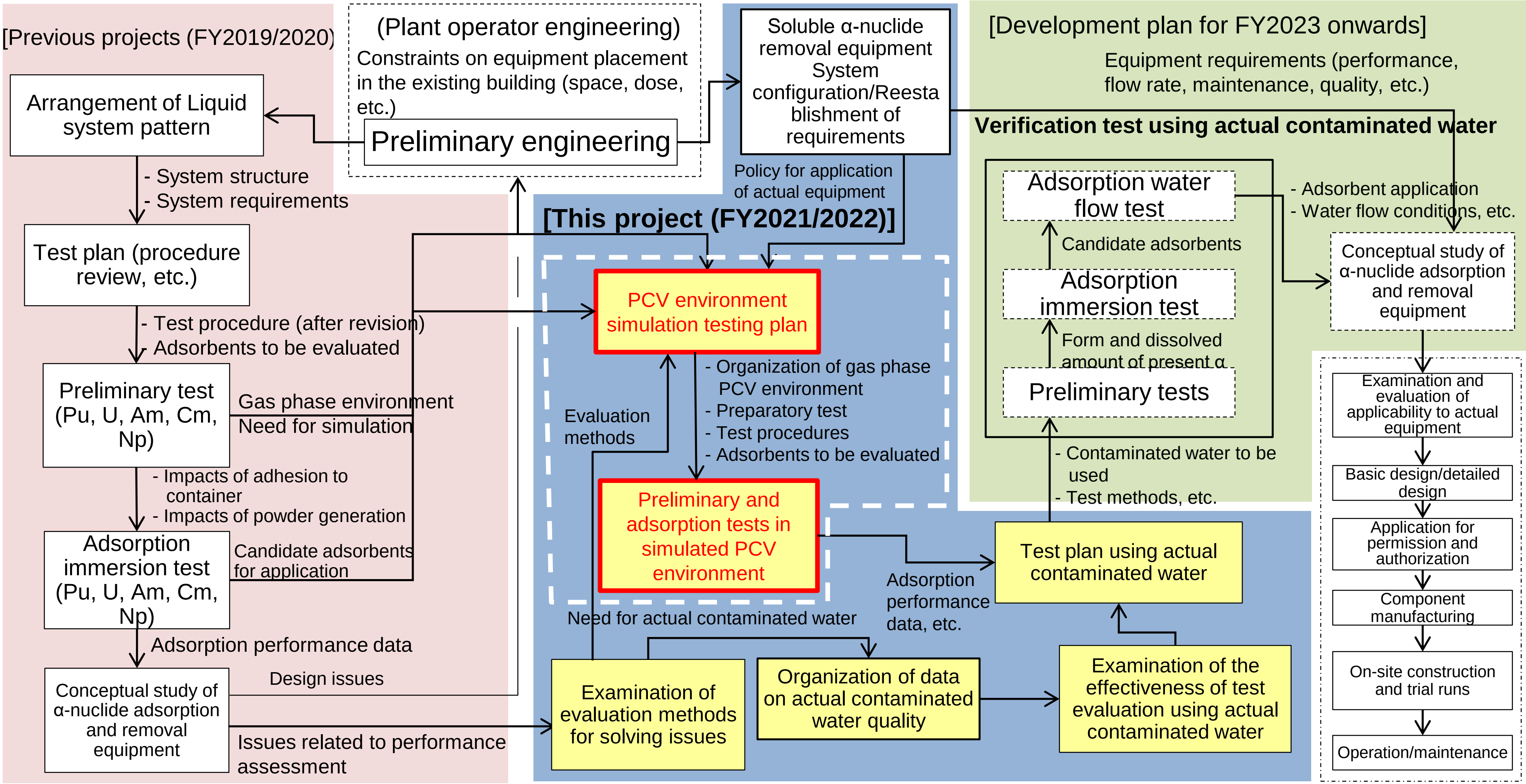
- In the FY2016 subsidy project, “Upgrading of Methods and Systems for Retrieval of Fuel Debris and Reactor Internals,” site boundary exposure doses for liquid system during normal and accident conditions were evaluated. The site boundary exposure dose was below the threshold value in both normal and accident conditions, but in both cases the effects of Pu-based nuclides and daughter product (Pu-238, Pu-241, Am-241) were significant.
- Alpha-emitting nuclides (hereinafter referred to as α-nuclides) at existing nuclear facilities are not nuclides that are discharged into the environment and the exposure due to α-nuclides should be as low as reasonably achievable. Therefore, from the subsidy projects (FY2017/2018) “Upgrading of Methods and Systems for Retrieval of Fuel Debris and Reactor Internals,” examination and evaluation of the effectiveness of removing α-nuclides in liquid system have started. Candidate technologies are selected for each type of α-nuclide (non-soluble α-nuclides and soluble α-nuclides), and element tests are conducted to evaluate the feasibility of the selected technologies.
- Adsorption removal and RO filters have been selected as candidate technologies for removal of soluble α-nuclides; RO filters are capable of removing α-nuclides in principle. On the other hand, although adsorption removal is a proven technology for the Fukushima Daiichi NPS contaminated water treatment, its applicability depends on the adsorbent selected and the form of α-nuclides present in solution.
- The previous project (subsidy project for FY2019 to 2020) dealt with the development of adsorption removal technology and adsorption tests under open-atmospheric conditions were conducted on Pu, U, Am, Cm, and Np α-nuclides (the 5 nuclides targeted for removal). Candidate adsorbents were selected, but their actual effectiveness in the event of PCV water quality fluctuation has not yet been evaluated.
- In addition, in order to verify the adsorption removal technology’s efficacy in removing α-nuclides, a verification test using actual contaminated water is required, but plans for such a test are still under discussion.

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Mapping out of this project

End of FY2021 Subject of report



[Previous projects (FY2019/2020)]

Arrangement of Liquid system pattern

- System structure
- System requirements

Test plan (procedure review, etc.)

- Test procedure (after revision)
- Adsorbents to be evaluated

Preliminary test (Pu, U, Am, Cm, Np)

Gas phase environment
Need for simulation

- Impacts of adhesion to container
- Impacts of powder generation

Adsorption immersion test (Pu, U, Am, Cm, Np)

Candidate adsorbents for application

Adsorption performance data

Conceptual study of α-nuclide adsorption and removal equipment

Design issues

Issues related to performance assessment

(Plant operator engineering)

Constraints on equipment placement in the existing building (space, dose, etc.)

Preliminary engineering

Soluble α-nuclide removal equipment System configuration/Reestablishment of requirements

Policy for application of actual equipment

[This project (FY2021/2022)]

PCV environment simulation testing plan

Evaluation methods

- Organization of gas phase PCV environment
- Preparatory test
- Test procedures
- Adsorbents to be evaluated

Preliminary and adsorption tests in simulated PCV environment

Need for actual contaminated water

Adsorption performance data, etc.

Test plan using actual contaminated water

Examination of evaluation methods for solving issues

Organization of data on actual contaminated water quality

Examination of the effectiveness of test evaluation using actual contaminated water

[Development plan for FY2023 onwards]

Equipment requirements (performance, flow rate, maintenance, quality, etc.)

Verification test using actual contaminated water

Adsorption water flow test

Candidate adsorbents

Adsorption immersion test

Form and dissolved amount of present α

Preliminary tests

- Contaminated water to be used
- Test methods, etc.

- Adsorbent application
- Water flow conditions, etc.

Conceptual study of α-nuclide adsorption and removal equipment

Examination and evaluation of applicability to actual equipment

Basic design/detailed design

Application for permission and authorization

Component manufacturing

On-site construction and trial runs

Operation/maintenance

Figure Overall flow of development of soluble α-nuclide removal technology

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Issues, implementation details and goals to be addressed in this project

[Issues]

- The behavior of α-nuclides in solution under conditions simulating the PCV environment during fuel debris retrieval work has not been verified.
- The impacts of changes in solution water quality on the soluble α-nuclide removal performance has not yet been evaluated.

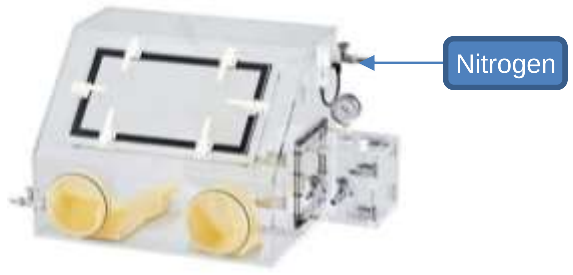
[Implementation details]

- A test equipment was manufactured to simulate the environment during fuel debris retrieval work.
- Preliminary tests were conducted to verify the dissolution behavior of α-nuclides under conditions simulating the environment during fuel debris retrieval work.
- Adsorption tests were conducted to verify the α-nuclide removal performance under conditions simulating the environment during fuel debris retrieval work, and applicable adsorbents are selected.

[Goals]

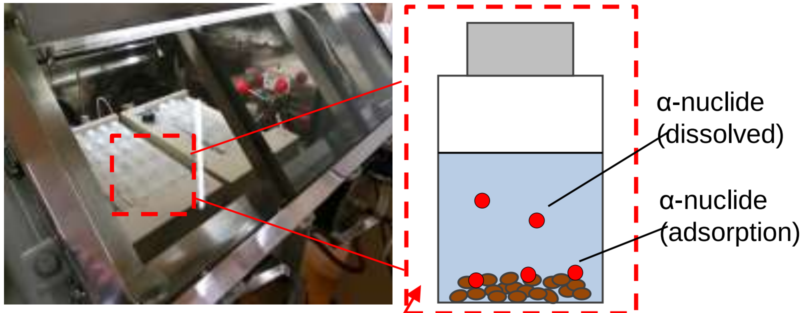
- Data acquisition on α-nuclide adsorption performance in the environment assuming fuel debris retrieval work, and select candidate adsorbents.
- Establishment of a water quality adjustment policy for soluble α-nuclide removal equipment.

<Illustration of test equipment for simulating a nitrogenous environment during fuel debris retrieval work>



Simple glove box

<Adsorption test>



View of glove box interior

Container for adsorption testing

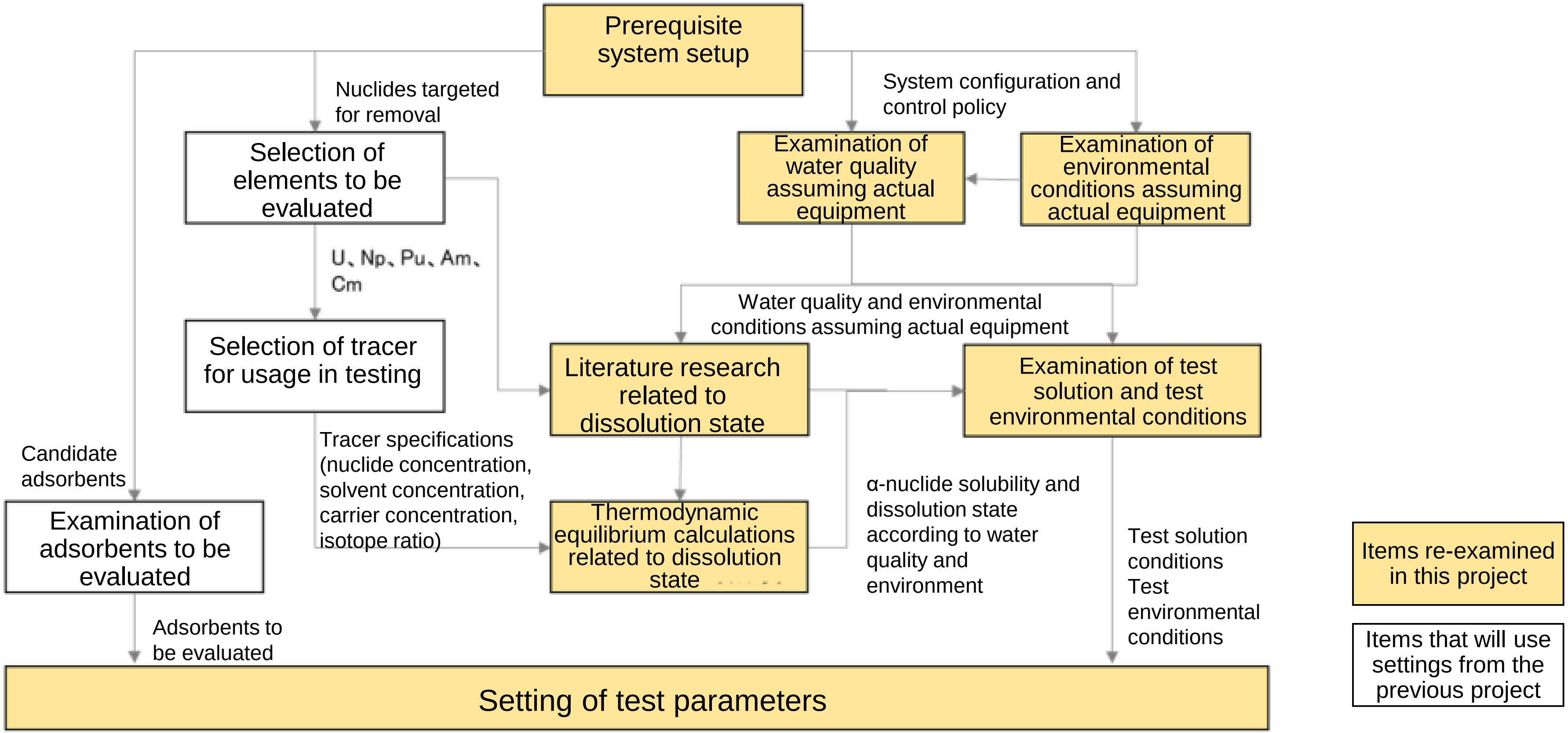
Measure the nuclide concentration in the liquid before and after the test and assess the adsorption amount

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Response flow up to element test parameter setting

After setting up the prerequisite system, the environmental and water quality conditions in the actual equipment were examined. The test environment and test solution conditions were also examined based on the literature research and calculation of thermodynamic equilibrium related to the dissolution state of α-nuclides. Then the test parameters were set according to the adsorbent to be evaluated.



Prerequisite system setup

Nuclides targeted for removal

Selection of elements to be evaluated

U、Np、Pu、Am、Cm

Selection of tracer for usage in testing

System configuration and control policy

Examination of water quality assuming actual equipment

Examination of environmental conditions assuming actual equipment

Water quality and environmental conditions assuming actual equipment

Literature research related to dissolution state

Examination of test solution and test environmental conditions

Candidate adsorbents

Examination of adsorbents to be evaluated

Tracer specifications (nuclide concentration, solvent concentration, carrier concentration, isotope ratio)

Thermodynamic equilibrium calculations related to dissolution state

α-nuclide solubility and dissolution state according to water quality and environment

Test solution conditions
Test environmental conditions

Adsorbents to be evaluated

Setting of test parameters

Items re-examined in this project

Items that will use settings from the previous project

Figure Response flow up to element test planning

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Setting of prerequisite system (liquid/gas systems)

- In the liquid system, in order to control water level and confine the contaminated water in the PCV, water is taken from the D/W (dry well) or S/C (PCV suppression chamber) and fuel debris is cooled by injecting and circulating water taken from D/W or S/C after cooling.
- The gas system manages negative pressure to confine dust and other substances in the PCV, and fills the PCV with nitrogen for hydrogen scavenging and other purposes.
- Although air flows in due to negative pressure control, the gas phase inside the PCV becomes mostly nitrogenous.
- Up until 2020 α-nuclide adsorption tests had been conducted in an atmospheric environment. Testing in a nitrogen environment is necessary in order to further investigate application to actual equipment.

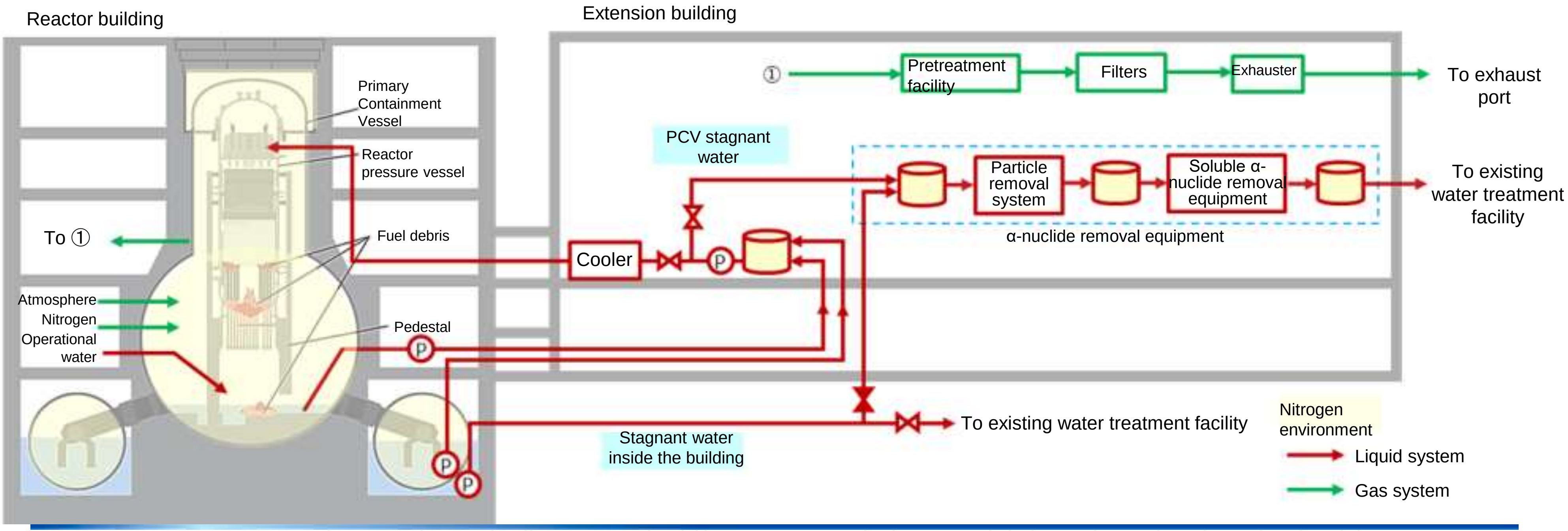


Figure Schematic diagram of liquid/gas systems during fuel debris retrieval (engineering basis)

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Setting of prerequisite system (soluble α-nuclide removal system)

- In the previous project, to study soluble α-nuclide removal system when adsorption/removal is applied, a facility configuration consisting of receiving tank, adsorption tower, monitoring tank, sampling equipment, etc. was examined.
- Adsorption tests were conducted under atmospheric conditions using α-nuclides elements (the nuclides targeted for removal). A tower configuration with attached activated carbon on top and zirconium phosphate/titanic acid in the second stage was proposed. The former is assumed to remove mainly colloidal α-nuclides while the latter is assumed to remove dissolved α-nuclides.
- In this project, similar adsorption tests under a nitrogen atmosphere (simulating the gas phase environment in the PCV) were conducted to evaluate the impacts on the solubility of α-nuclides and adsorption performance, and to investigate whether the tower configuration and adsorbent used in the previous project need to be altered.

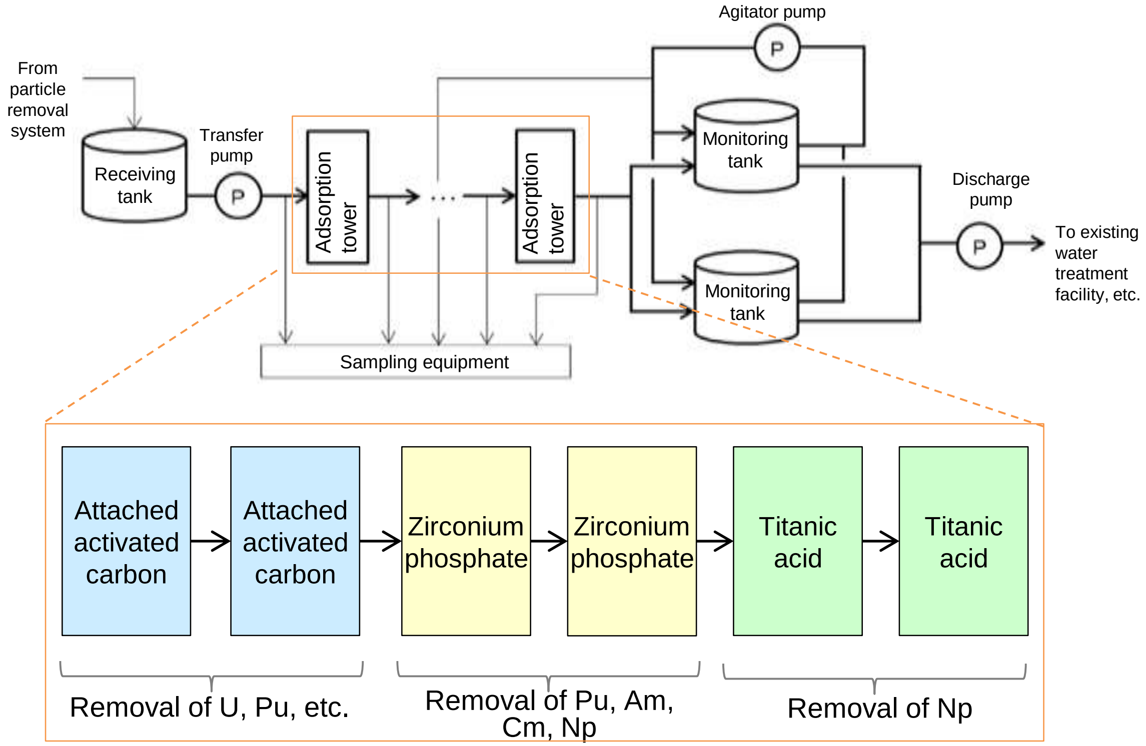


Figure Schematic diagram of soluble α-nuclide removal system and proposed tower configuration*

*Supplementary information on the tower configuration

- The number of towers in each adsorption tower is provisionally set at 2 to enable merry-go-round operation. The number of required towers for each adsorption tower needs to be evaluated in the future.
- In order to reduce the number of adsorption towers as much as possible, attached activated carbon adsorption towers (which have high removal performance for colloidal α-nuclides in addition to soluble α-nuclides) and zirconium phosphate adsorption towers (which have high adsorption performance for multiple soluble α-nuclides) are installed. However, since the Np adsorption performance of the attached activated carbon and zirconium phosphate is only relatively high and the concentration reduction during water flow treatment may be insufficient, a titanic acid adsorption tower should be installed at the post zirconium phosphate stage.

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Examination of environmental conditions assuming actual equipment and policy for applying these conditions to the test environmental conditions

- The following table summarizes the proposed control policy and control values for the gas phase environment of the PCV, based on the analysis status in the plant operator engineering.
- Oxygen concentration is related to the degree of air inflow, and since it is under consideration to set the control value less than 4%, this will be reflected in the conditions in the test evaluation. (Note, however, that the 4% figure is tentative, and not a definite value.)
- The atmospheric component in the PCV gas phase is estimated to be at a concentration less than 1/5 that of the atmosphere, which has an oxygen concentration of about 20%. (Carbon dioxide concentration in the PCV is assumed to be 70 to 80 ppm or less)

Table Proposed control policy and values for environmental items during fuel debris retrieval (gas system)

No.	Environmental item		Control requirements	Proposed control policy	Proposed control value	Remarks
1	Gas phase environment	PCV gas phase pressure	Gas phase confinement (leakage prevention)	Negative pressure is managed by controlling the exhaust flow rate	TBD	If negative pressure control is not feasible, the alternatives are positive pressure control or equalization pressure control.
2		Nitrogen concentration	No	(Nitrogen supply control)	—	Nitrogen concentration itself is not a control target.
3		Hydrogen concentration	Gas phase confinement (fire/explosion prevention)	Nitrogen supply and exhaust flow rate are controlled to keep hydrogen below the lower explosive limit	Less than 2%	This is tentative, not a definite value.
4		Oxygen concentration	Gas phase confinement (fire/explosion prevention)	Nitrogen supply and exhaust flow rate are controlled to prevent the conditions of Zr fire outbreak.	Less than 4%	Concentration increase due to air inflow during negative pressure control. This is tentative, not a definite value.
5		Carbon dioxide concentration	No	Course of events	—	Concentration increase due to air inflow during negative pressure control.
6		Dust concentration	Gas phase confinement (leakage prevention)	Exhaust flow rate is controlled to keep the activity concentration below the specified level	TBD	At the time of full discharge, the concentration is less than or equal to the public exposure dose of 5 mSv.

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Examination of water quality conditions assuming actual equipment and policy for applying these conditions to the test solution conditions (1/3)

- By the start of fuel debris retrieval work, incremental changes in water quality are assumed due to circulation cooling operations in the liquid system and PCV negative pressure control in the gas system.
- Once fuel debris retrieval work begins, it is anticipated that the pH of the liquid phase will increase due to elution of concrete during cutting.
- The increase in pH is also assumed to increase the amount of gas phase carbon dioxide that dissolves into the liquid phase.

Table Evaluation status of water quality of PCV stagnant water and α-nuclide adsorption performance under liquid/gas systems and fuel debris retrieval operating conditions

Operating conditions		**STEP1**	**STEP2**	**STEP3**	**STEP4**
Circulation cooling		**Not conducted**	**Conducted**	**Conducted**	**Conducted**
PCV negative pressure control		**Not conducted**	**Not conducted**	**Conducted**	**Conducted**
Fuel debris retrieval work		**Not conducted**	**Not conducted**	**Not conducted**	**Conducted**
Quality of PCV stagnant water	Overview	RO-treated water is injected from the outside the PCV and then leaks out, so the water quality is basically the same as RO-treated water.	Circulation of RO-treated water may cause elution of concrete, etc. Since the gas phase is in a nitrogen atmosphere, there is no atmospheric impacts.	By managing the PCV with negative pressure, the carbonic acid concentration underwater increases due to gas phase carbon dioxide concentration increase from air inflow into the PCV.	Increased elution of concrete due to fuel debris retrieval work causes a pH increase. Carbonic acid concentration is positively correlated with pH. Injection of boric acid solution as criticality prevention is also hypothesized.
	Estimated pH	Approx. 7	7 to 9	5 to 9	5 to 12
	Inorganic carbon* concentration (estimated)	Approx. 1 ppm	Approx. 1 ppm	Several ppm (Impacts of air inflow)	Several dozen ppm (Air inflow + impacts of pH increase)
α-nuclide adsorption performance evaluation status		(Not applicable)	(Not applicable)	Selection of candidate adsorbents through adsorption tests up to FY2020	Not yet evaluated

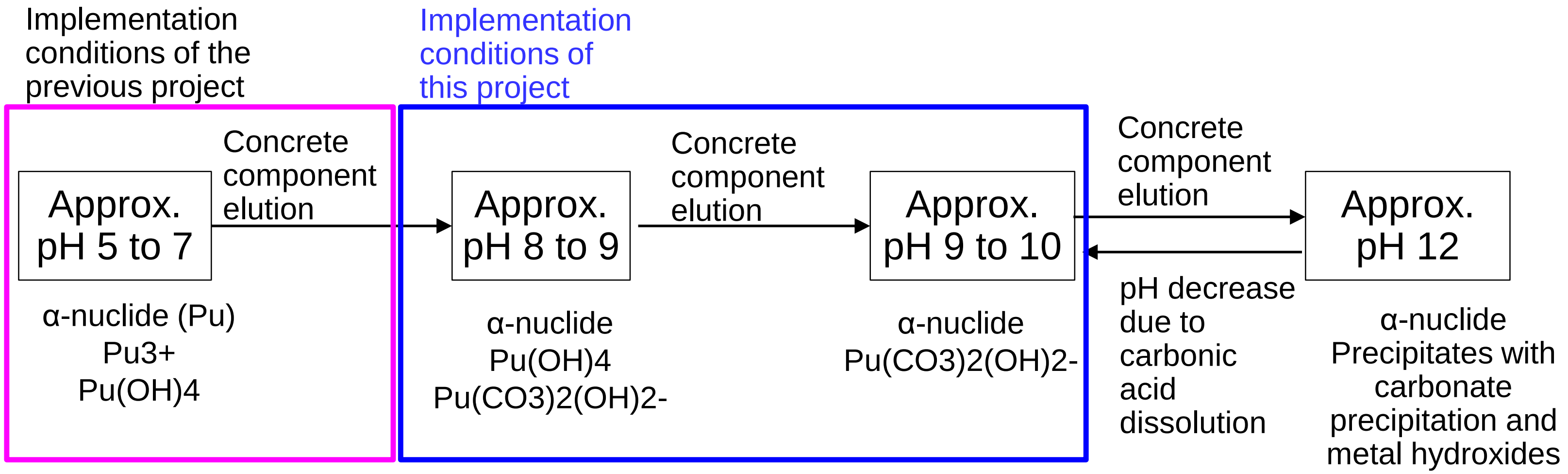
*Collective term for CO_2, HCO_3^- and CO_3^{2-}. The abundance ratio varies with the pH of the solution.

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Examination of water quality conditions assuming actual equipment and policy for applying these conditions to the test solution conditions (2/3)

- The pH range required for estimation was studied because the amount of pH and carbon dioxide elution into the liquid phase are assumed to increase due to elution of concrete components during fuel debris retrieval.
- When calcium from concrete is eluted, the pH of a saturated solution of calcium hydroxide is approximately 12.4, but in the high pH region, carbon dioxide in the gas phase is more soluble, and over time the pH drops to about 9 to 10 due to carbonic acid dissolution.
- When the pH decreases due to carbonic acid dissolution, calcium carbonate precipitates and α-nuclide concentration is assumed to decrease due to coprecipitation, in which case the need for adsorption removal is reduced.
- In order to verify the dissolved α-nuclide adsorption performance, adsorption tests under alkaline conditions (pH 8 to 10) are planned under this project, in which the α-nuclide concentration in the liquid phase is not assumed to decrease significantly.



Implementation conditions of the previous project

Implementation conditions of this project

Approx. pH 5 to 7

α-nuclide (Pu)
Pu3+
Pu(OH)4

Concrete component elution

Approx. pH 8 to 9

α-nuclide
Pu(OH)4
Pu(CO3)2(OH)2-

Concrete component elution

Approx. pH 9 to 10

α-nuclide
Pu(CO3)2(OH)2-

Concrete component elution

pH decrease due to carbonic acid dissolution

Approx. pH 12

α-nuclide
Precipitates with carbonate precipitation and metal hydroxides

Figure Water quality fluctuation scenario in case of concrete component elution

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Examination of water quality conditions assuming actual equipment and policy for applying these conditions to the test solution conditions (3/3)

- There are three factors that contribute to changes in PCV stagnant water quality: elution of seawater components, elution of concrete components, and injection of boric acid solution (sodium pentaborate). The water quality conditions assuming actual equipment with each condition as a parameter are summarized below.
- Concrete component elution-mediated pH increase is assumed to alter the form of α-nuclides. Since it is necessary to understand the impacts of pH on adsorption performance, implementation under two conditions is planned.

Table List of PCV stagnant water quality conditions assuming actual equipment

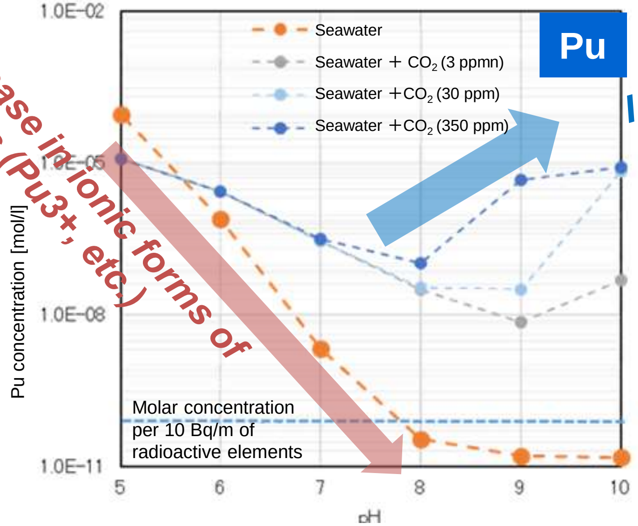
Type of test water	Expected conditions	Seawater component	Concrete component	Boric acid solution component	Conditions							
					Temperature [°C]	Cl ion concentration [ppm]	Ca ion concentration [ppm]	Boron concentration [ppm]	pH	Inorganic carbon concentration [ppm]	SS concentration [ppm]
1	Circulation with water of similar quality to RO-treated water	No	No	No	10 to 40	0	<1	<1	6 to 7	Approx. 1	<1
2-1	Small amount of seawater components eluted into the circulating cooling water	Yes	No	No	10 to 40	20	<1	<1	6 to 7	Approx. 1	<1
2-2	Large amount of seawater components eluted into the circulating cooling water	Yes	No	No	10 to 40	100	Approx. 1	<1	6 to 7	Approx. 1	<1
3-1	Small amount of concrete component eluted into water quality 2-1	Yes	Yes	No	10 to 40	20	Low	<1	8 to 9	Low	<1
3-2	Large amount of concrete component eluted into water quality 2-1	Yes	Yes	No	10 to 40	20	High	<1	9 to 10	High	<1
4-1	Small amount of concrete component eluted into water quality 2-2	Yes	Yes	No	10 to 40	100	Low	<1	8 to 9	Low	<1
4-2	Large amount of concrete component eluted into water quality 2-2	Yes	Yes	No	10 to 40	100	High	<1	9 to 10	High	<1
5	Boric acid solution injected into water quality 2-1 to prevent criticality	Yes	Yes	Yes	10 to 40	20	Low	7000	8 to 9	High	<1

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Investigation and evaluation related to dissolution state

- In the previous project, thermodynamic equilibrium calculations were performed for a system in which carbon dioxide exists in the gas phase. It was evaluated that the solubility form of coordinated carbonate ions (carbonate complexes) as the dissolved form of α-nuclides increases in alkaline conditions.
- The increase in carbonate ion concentration accompanying the rise in pH may cause precipitation formation with Ca (Ca carbonate), a concrete component, accompanied by a decrease in concentration due to coprecipitation of α-nuclides. However, this coprecipitation phenomenon cannot be evaluated by equilibrium calculations, so verification through test evaluation under simulated environment assuming actual equipment is required.

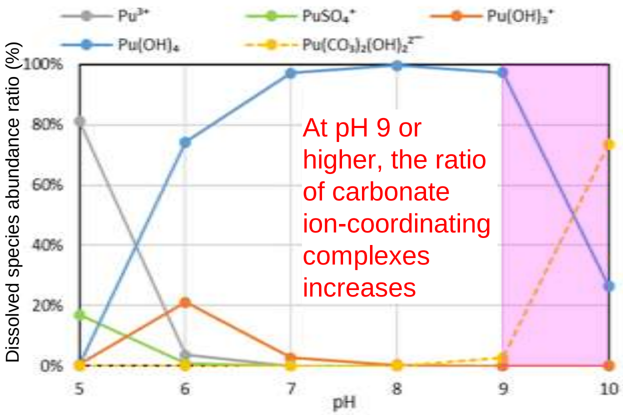


Decrease in ionic forms of nuclides (Pu3+, etc.)

Increase in carbonic acid-coordinating nuclides ($Pu(CO3)2(OH_3^{2-})$)

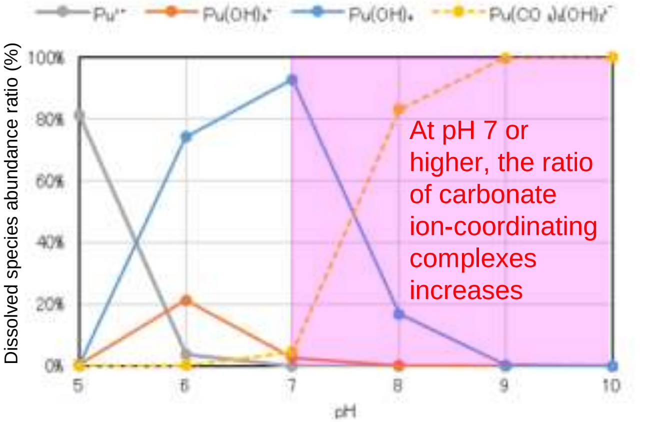
Figure Results of evaluating Pu solubility based on thermodynamic equilibrium calculation with condition of carbon dioxide concentration in gas phase as a parameter (50-fold diluted seawater)

(1) Carbon dioxide concentration in gas phase: 3 ppm



At pH 9 or higher, the ratio of carbonate ion-coordinating complexes increases

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



At pH 7 or higher, the ratio of carbonate ion-coordinating complexes increases

Figure Results of evaluating dissolved form of Pu based on thermodynamic equilibrium calculation with condition of carbon dioxide concentration in gas phase as a parameter (50-fold diluted seawater)

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

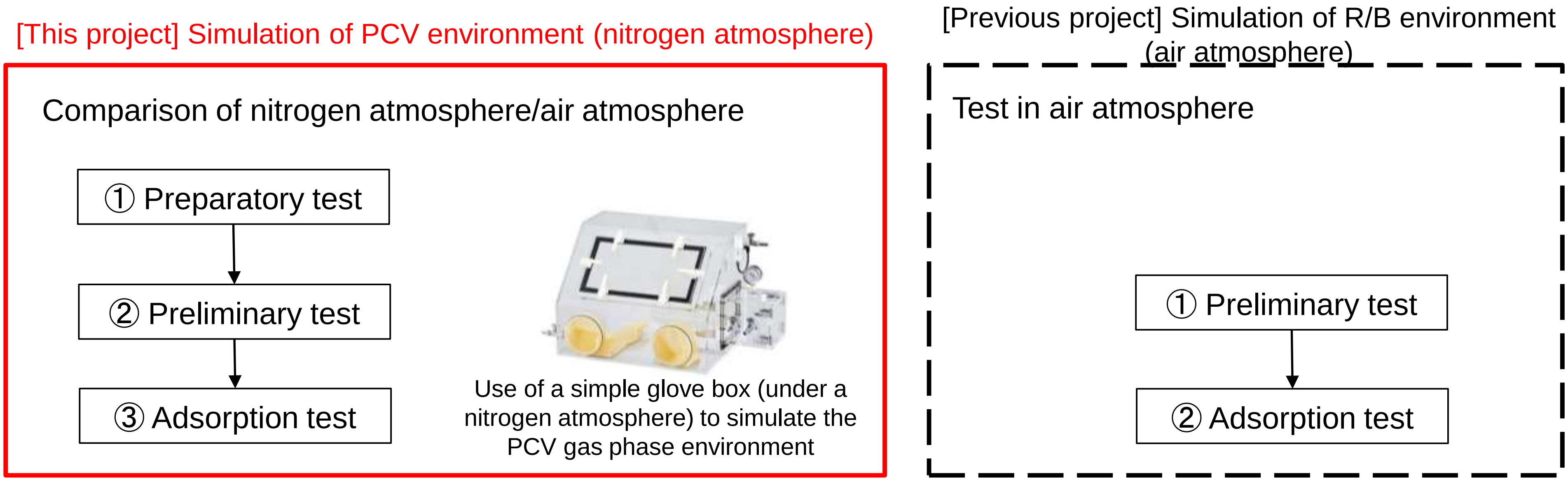
- Overall test plan

- In the previous project, the soluble α removal test was conducted in an environment simulating the air atmosphere inside the R/B, whereas in this project the test simulated the nitrogen atmosphere environment inside the PCV.
- The adsorption test was conducted after completing the preparatory and preliminary tests. The outline and objectives of each test are as follows.

① Preparatory test: Verification of feasibility of controlling the nitrogen atmosphere in the simple glove box (verification of feasibility of maintaining an oxygen concentration of 4% or less)

② Preliminary test: Verification of the impacts of α-nuclide in the liquid phase adhering to containers as well as the impacts of precipitation formation (verification of concentration reduction factors other than adsorbents)

③ Adsorption test: Verification of soluble α-nuclide adsorption performance by immersion testing



[This project] Simulation of PCV environment (nitrogen atmosphere)

Comparison of nitrogen atmosphere/air atmosphere

① Preparatory test → ② Preliminary test → ③ Adsorption test

Use of a simple glove box (under a nitrogen atmosphere) to simulate the PCV gas phase environment

[Previous project] Simulation of R/B environment (air atmosphere)

Test in air atmosphere

① Preliminary test → ② Adsorption test

Figure Overall test implementation flow

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Test system and preparatory test results

- The diagram below (Figure A) shows the test system for evaluating simulation of the internal PCV environment (nitrogen atmosphere).
- In order to investigate radiation control, a simple glove box (simple GB) was installed in the hood of the laboratory, and a nitrogen-filled line from an N2 cylinder is connected to create a nitrogen atmosphere inside the simple GB.
- By verifying the impacts of air inflow into the simple GB, it was confirmed that it takes about 10 days to exceed the 4% oxygen concentration control target, and that the atmosphere can be maintained even when human work is unavailable (e.g., nights and holidays). (Figure B)
- In addition to periodically checking the oxygen concentration in the simple GB and filling it with nitrogen as appropriate, it was also decided to adopt a policy of adding nitrogen prior to collecting liquid from test samples, since the GB atmosphere is strongly affected each time the lid of the test sample container is opened.
- Temperature control in the simple GB is difficult due to space constraints. Therefore, temperature was measured and recorded during the test and data necessary to investigate any impacts from temperature difference was collected.

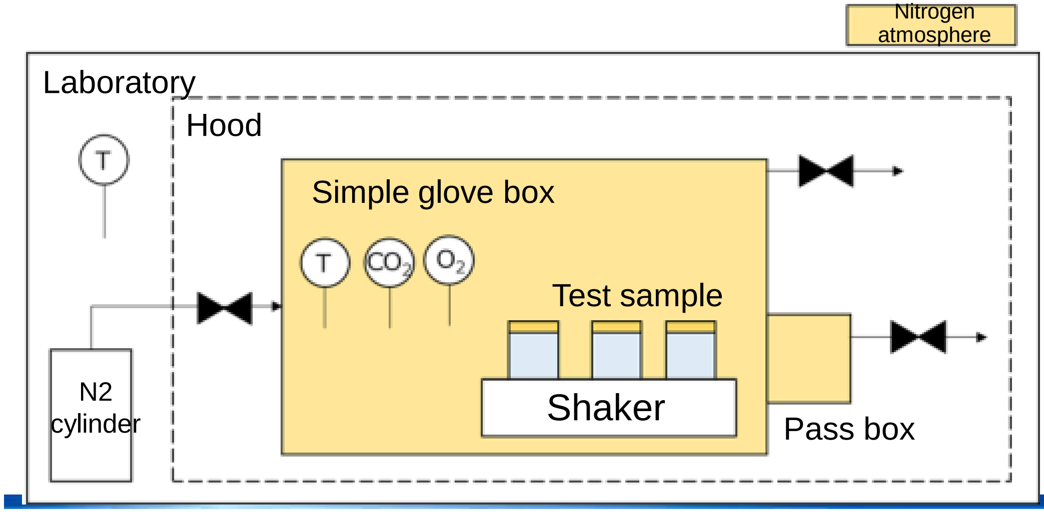


Figure A Test system simulating a nitrogen atmosphere

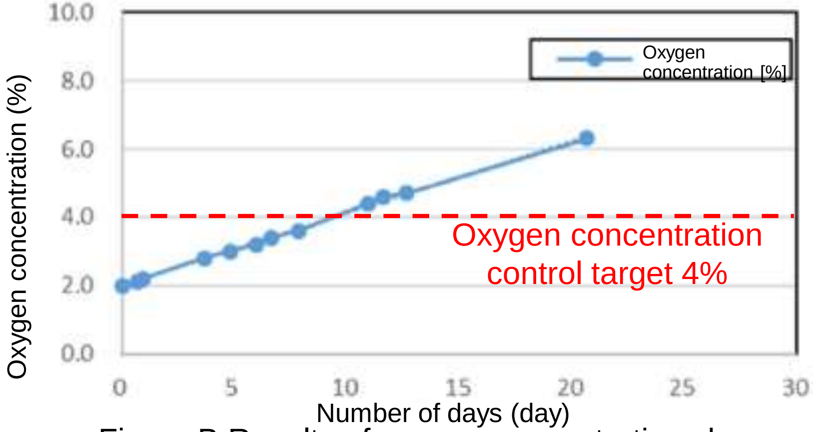


Figure B Results of oxygen concentration changes verified in a simple GB

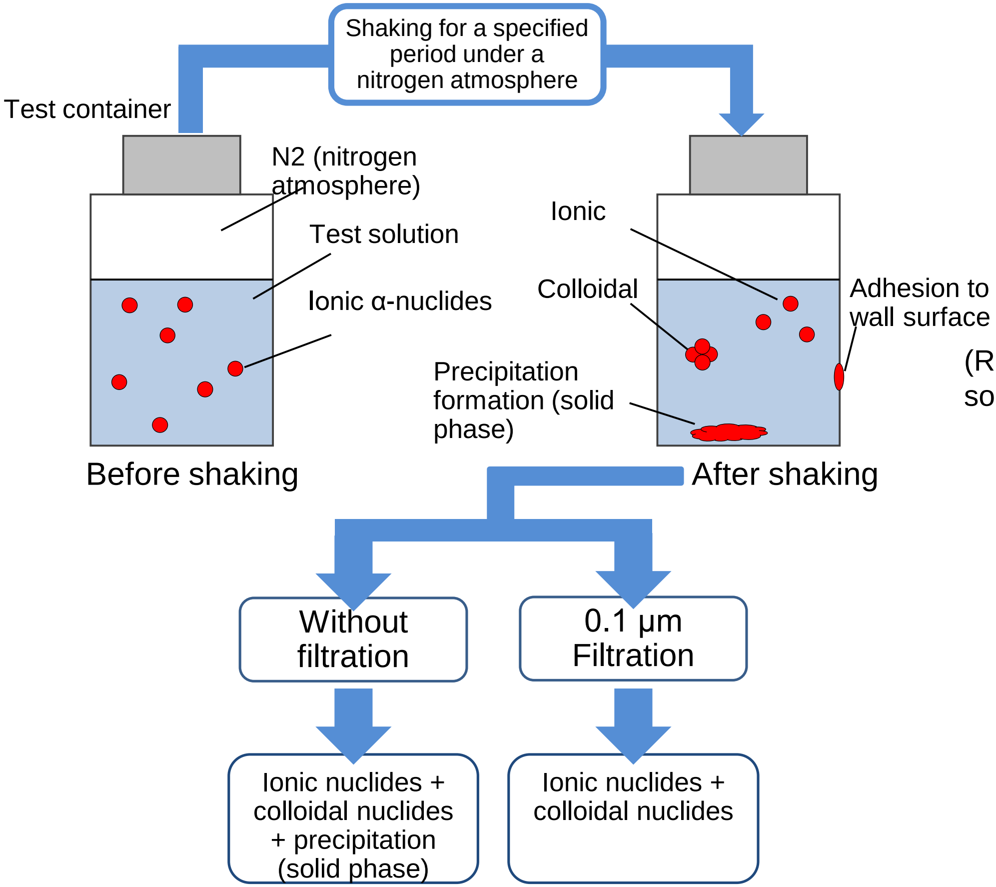
7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Preliminary test procedures

The preliminary test procedures, established based on the preparatory test results, are shown below. The test procedures are the same as in the previous project, and the test environment was performed under a nitrogen atmosphere.

Assuming operation in a simple GB under a nitrogen atmosphere



Shaking for a specified period under a nitrogen atmosphere

Test container

N2 (nitrogen atmosphere)

Test solution

Ionic α-nuclides

Before shaking

Ionic

Colloidal

Precipitation formation (solid phase)

Adhesion to wall surface

After shaking

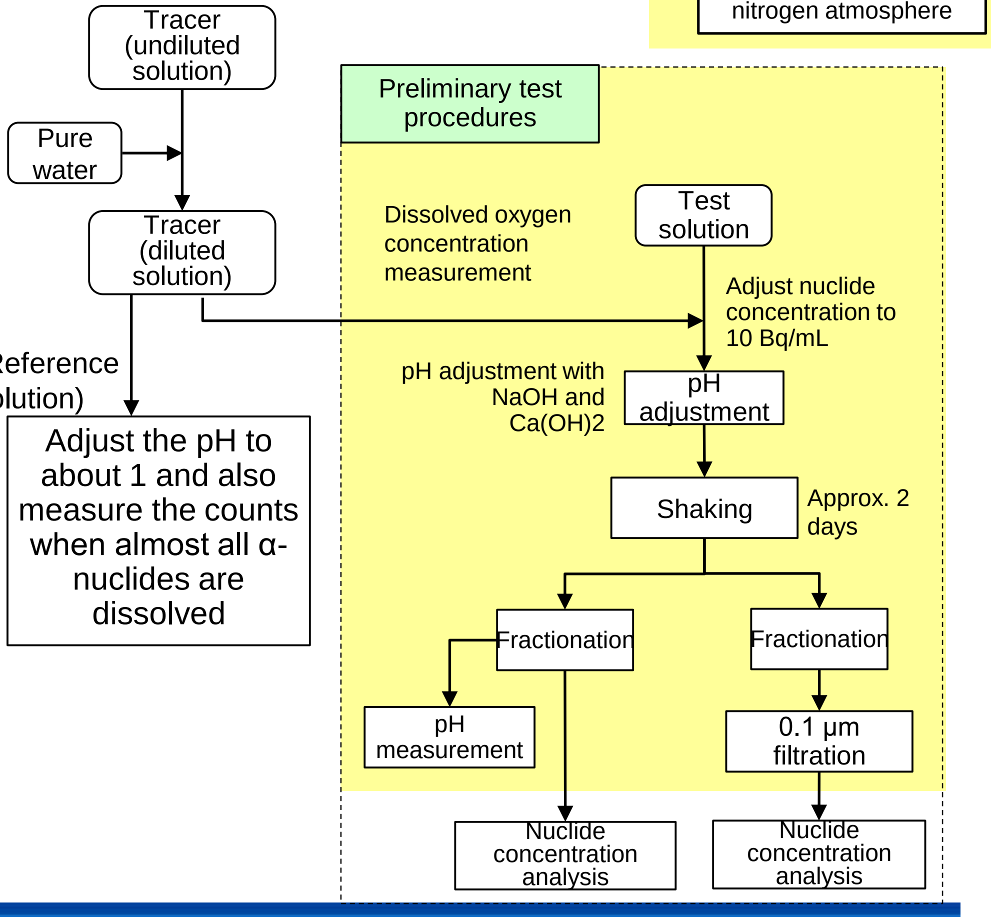
Without filtration

0.1 µm Filtration

Ionic nuclides + colloidal nuclides + precipitation (solid phase)

Ionic nuclides + colloidal nuclides

Figure Diagram of preliminary test



Tracer (undiluted solution)

Pure water

Tracer (diluted solution)

(Reference solution)

Adjust the pH to about 1 and also measure the counts when almost all α-nuclides are dissolved

Preliminary test procedures

Dissolved oxygen concentration measurement

Test solution

Adjust nuclide concentration to 10 Bq/mL

pH adjustment with NaOH and Ca(OH)2

pH adjustment

Shaking

Approx. 2 days

Fractionation

Fractionation

pH measurement

0.1 µm filtration

Nuclide concentration analysis

Nuclide concentration analysis

Figure Preliminary test procedures

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Adsorption test procedures

The adsorption test procedures, established based on the preparatory test results, are shown below.

The test procedures are the same as in the previous project, and the test environment was performed under a nitrogen atmosphere.

Assuming operation in a simple GB under a nitrogen atmosphere

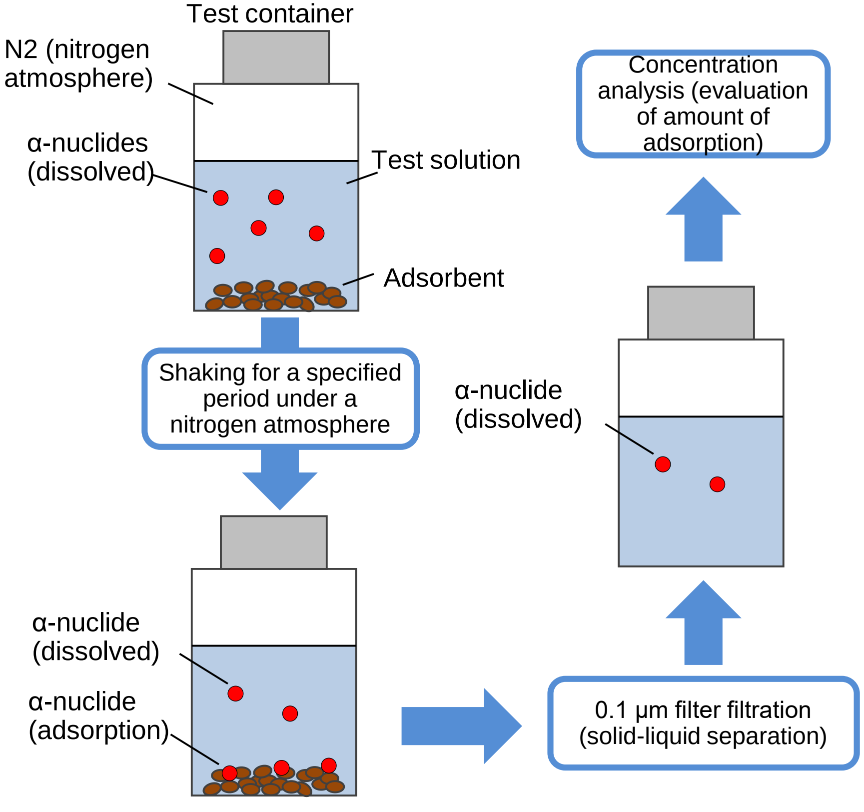
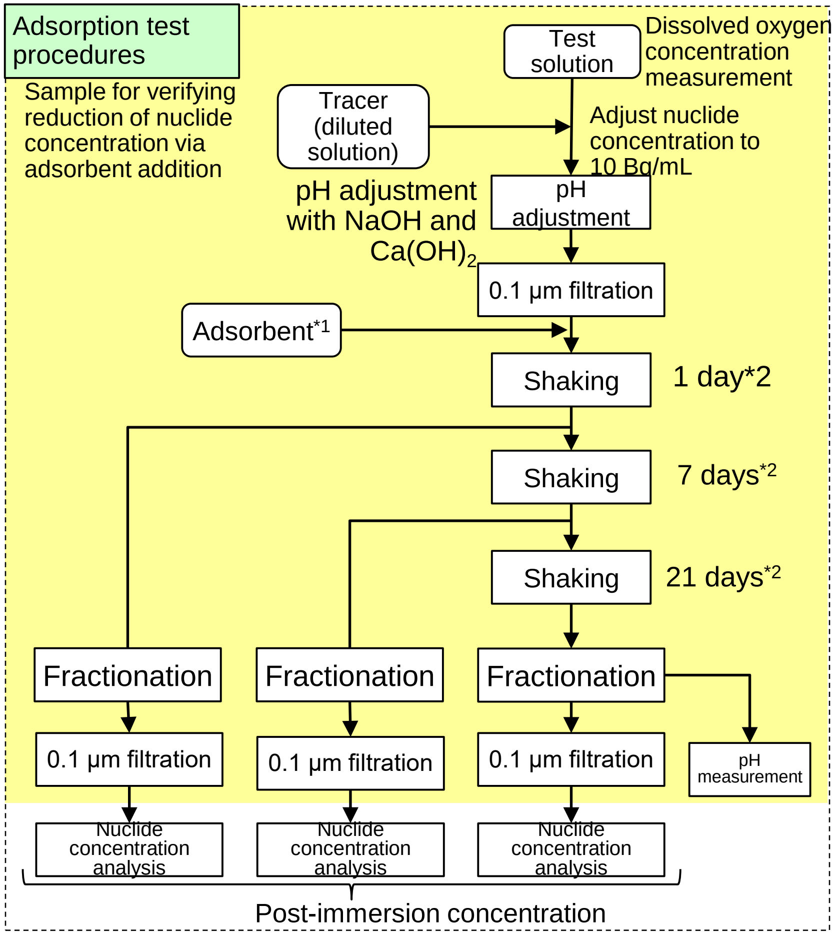


Figure Diagram of adsorption test



*1: Some adsorbents are pre-washed to reduce pH fluctuations from addition of adsorbents.

*2: Equilibrium attainment is ensured by setting a 3-pattern shaking period.

Figure Adsorption test procedures

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Preliminary test parameters

Considering the PCV gas phase environmental conditions (nitrogen environment) and the alkaline conditions during concrete cutting, the following test conditions were set.

Table Test parameters for preliminary test

No.	Test parameters	Setting requirements	Remarks
1	Nuclide	Am, Cm, Pu, Np, U	Based on the results of exposure assessment at the site boundary, it is highly necessary to remove these particular α-nuclides
2	Nuclide concentration	10 Bq/mL	Particular concentrations that can be handled by the test facility and that allow for analysis of removal rates
3	Gas phase environment	Nitrogen atmosphere	Required conditions given the anticipated gas phase environment inside the PCV during fuel debris retrieval Based on preparatory test results, the atmosphere is controlled using a simple GB with an oxygen concentration of 4% as the reference value
4	Material of test container	PP, PFA	Adsorption tests are conducted under two conditions, containers with a low container adhesion impacts are used PP: Polypropylene, PFA: Fluoropolymer.
5	Water quality conditions	See the table below	

Table Water quality conditions for preliminary test

No.	Water quality conditions	pH	Remarks
1	1000-fold diluted seawater	6 to 7	The policy is to use the results of condition implementation from the previous project, and to utilize as-yet unimplemented conditions.
2	200-fold diluted seawater	6 to 7	
3	1000-fold diluted seawater ＋Ca(OH)2	Approx. 9	- Assumed to be operated near the analysis value of the chloride ion concentration in the stagnant water up to the present (19 ppm) due to the low elution of seawater components - Assuming a rise in pH due to elution of concrete components
4		Approx. 10	
5	200-fold diluted seawater ＋Ca(OH)2	Approx. 9	- Assumed to be operated near the upper control limit (100 ppm) due to high elution of seawater components - Assuming a rise in pH due to elution of concrete components
6		Approx. 10	
7	Sodium pentaborate water (7000 ppm as B)	8 to 9	Conditions for evaluating the impacts of sodium pentaborate injection

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Adsorption test parameters

The proposed adsorption test conditions are shown in the table below. Based on the results of the preliminary tests to be conducted in the future, water quality conditions, adsorbent conditions, and shaking period conditions to be implemented will be selected.

Table Test parameters for adsorption test

No.	Test parameters	Setting requirements	Remarks
1	Nuclide	Am, Cm, Pu, Np, U	Based on the results of exposure assessment at the site boundary, it is highly necessary to remove these particular α-nuclides
2	Nuclide concentration	10 Bq/mL	Particular concentrations that can be handled by the test facility and that allow for analysis of removal rates
3	Gas phase environment	Nitrogen atmosphere	Required conditions given the anticipated gas phase environment inside the PCV during fuel debris retrieval Based on preparatory test results, the atmosphere is controlled using a simple GB with an oxygen concentration of 4% as the reference value
4	Shaking period	1, 7, and 21 days	Evaluation of near-equilibrium state by shaking for 7 and 21 days. Judgment whether the adsorption rate is remarkably low by shaking for a short time.
5	Material of test container	PP or PFA	Based on the preliminary test results, the type of container to be used is selected. PP: Polypropylene, PFA: Fluoropolymer.
6	Adsorbent	see remarks	Adsorbents (attached activated carbon, activated carbon, titanic acid, silicic titanic acid, zirconium phosphate) that have obtained high adsorption performance in the evaluation under atmospheric environmental conditions up to the previous project are targeted.
7	Water quality conditions	See the table below	

Table Water quality conditions for adsorption test*

No.	Water quality conditions	pH	Remarks
1	1000-fold diluted seawater ＋Ca(OH)2	Approx. 9	- Assumed to be operated near the analysis value of the chloride ion concentration in the stagnant water up to the present (19 ppm) due to the low elution of seawater components - Assuming a rise in pH due to elution of concrete components
2		Approx. 10	
3	200-fold diluted seawater ＋Ca(OH)2	Approx. 9	- Assumed to be operated near the upper control limit (100 ppm) due to high elution of seawater components - Assuming a rise in pH due to elution of concrete components
4		Approx. 10	
5	Sodium pentaborate water (7000 ppm as B)	8 to 9	Conditions for evaluating the impacts of sodium pentaborate injection

*Since all conditions cannot be implemented due to restrictions in the test process, the plan is to narrow down the water quality conditions to be implemented based on the preliminary test results.

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Preliminary test progress

- Using Cm, 1000-fold diluted seawater and 200-fold diluted seawater were adjusted to pH 9 and 10 with calcium hydroxide, and shaken for 2 days in two types of containers (PP, PFA).
- Since the counts are reduced by 10 to 30% without filtration, it is estimated that part of Cm adheres to the container.
- In addition, it is estimated that part of the Cm changed into particles from the decrease in the count after 0.1 μm filtration.
- Adhesion to containers and formation of particles were verified, but since Cm exists in a dissolved state to some extent, it was verified that an adsorption test was required. Since there is no big difference in the results between PP and PFA, PP containers will be used in the adsorption test.

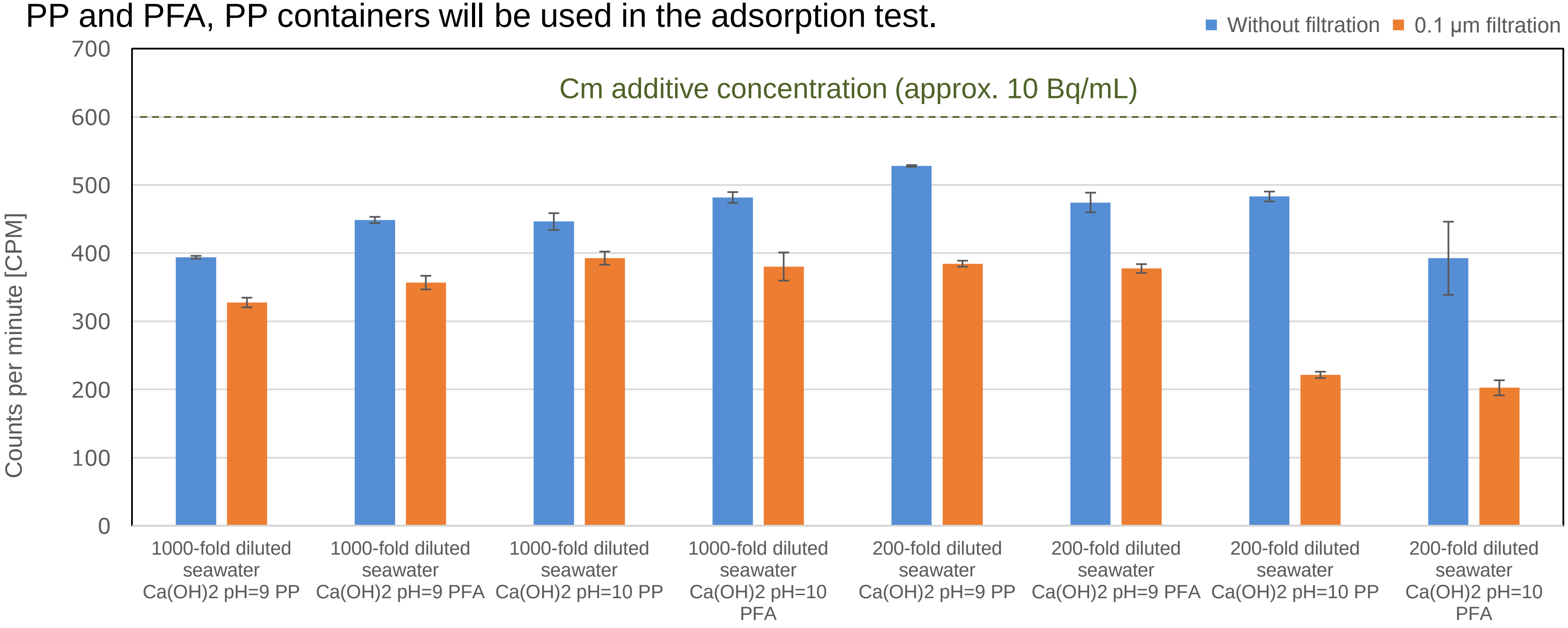


Figure Preliminary test results using Cm

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Preliminary test progress (2)

- Based on 1000-fold diluted seawater and 200-fold diluted seawater, Am was used to prepare solutions adjusted to pH 9 and 10 with calcium hydroxide and B=7000 ppm with sodium pentaborate, and shaken for 2 days.
- The counts were lower than that of the reference solution under the unfiltered conditions of 1000-fold diluted seawater pH 9,10 and 200-fold diluted seawater pH 10, and it is estimated that part of Am adheres to the container. In addition, it is estimated that part of Am changed into particles from the decrease in the count after 0.1 μm filtration.
- Adhesion to containers and formation of particles were verified for some liquids, but since Am exists in a dissolved state to some extent, it was verified that an adsorption test was required.

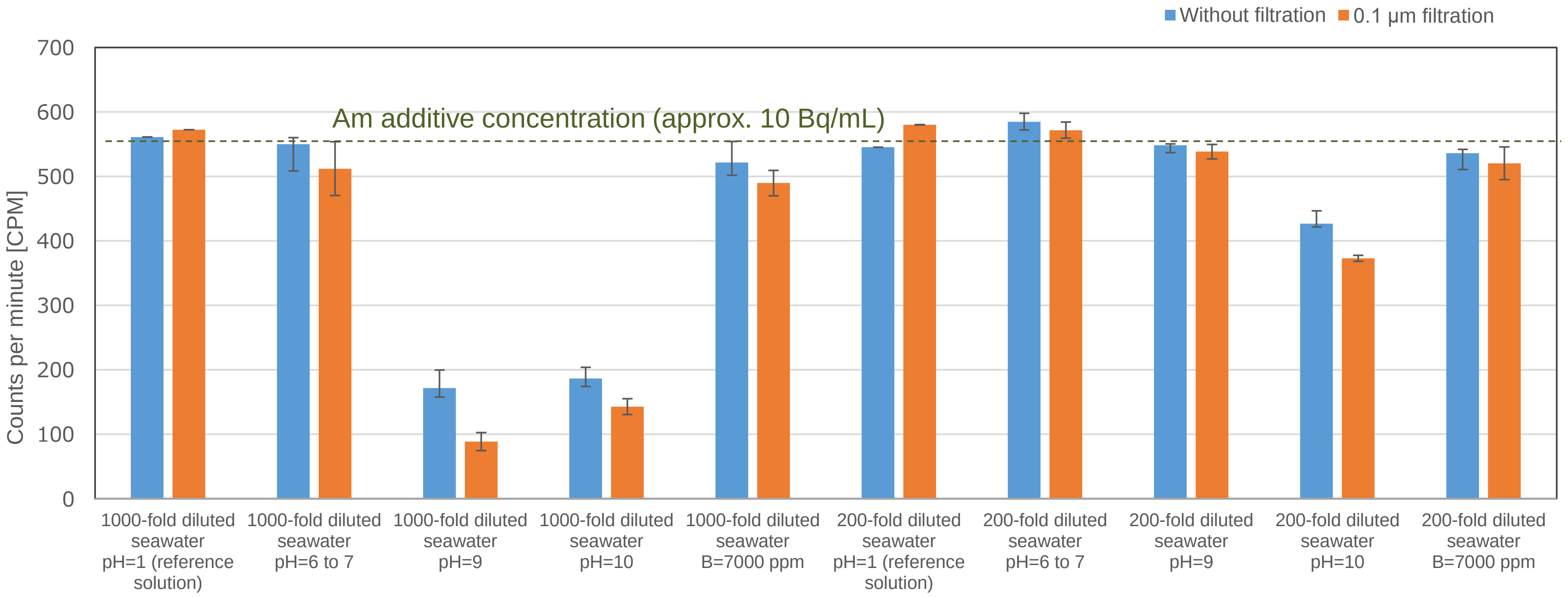


Figure Preliminary test results using Am

7. Implementation details

① Development of soluble α-nuclide removal technology (element test assuming fuel debris retrieval work)

- Summary

[Results for this year]

- ✓ Based on the control policy for the gas phase environment inside the PCV during fuel debris retrieval, a policy was determined to control the oxygen concentration at 4% or less as a simulation of the test environment, and it was verified that is possible to control the atmosphere inside the simple glove box during the preparatory test.
- ✓ Based on the gas-phase atmosphere control policy verified in the preparatory test, the test procedures for the preliminary test and the adsorption test were devised in the simple glove box with nitrogen atmosphere.
- ✓ Some preliminary tests have been started, and Am and Cm exist in a dissolved state to some extent in a nitrogen atmosphere and alkaline conditions (pH 9, 10), so it was verified that there is a high need for nuclide removal and an adsorption test.

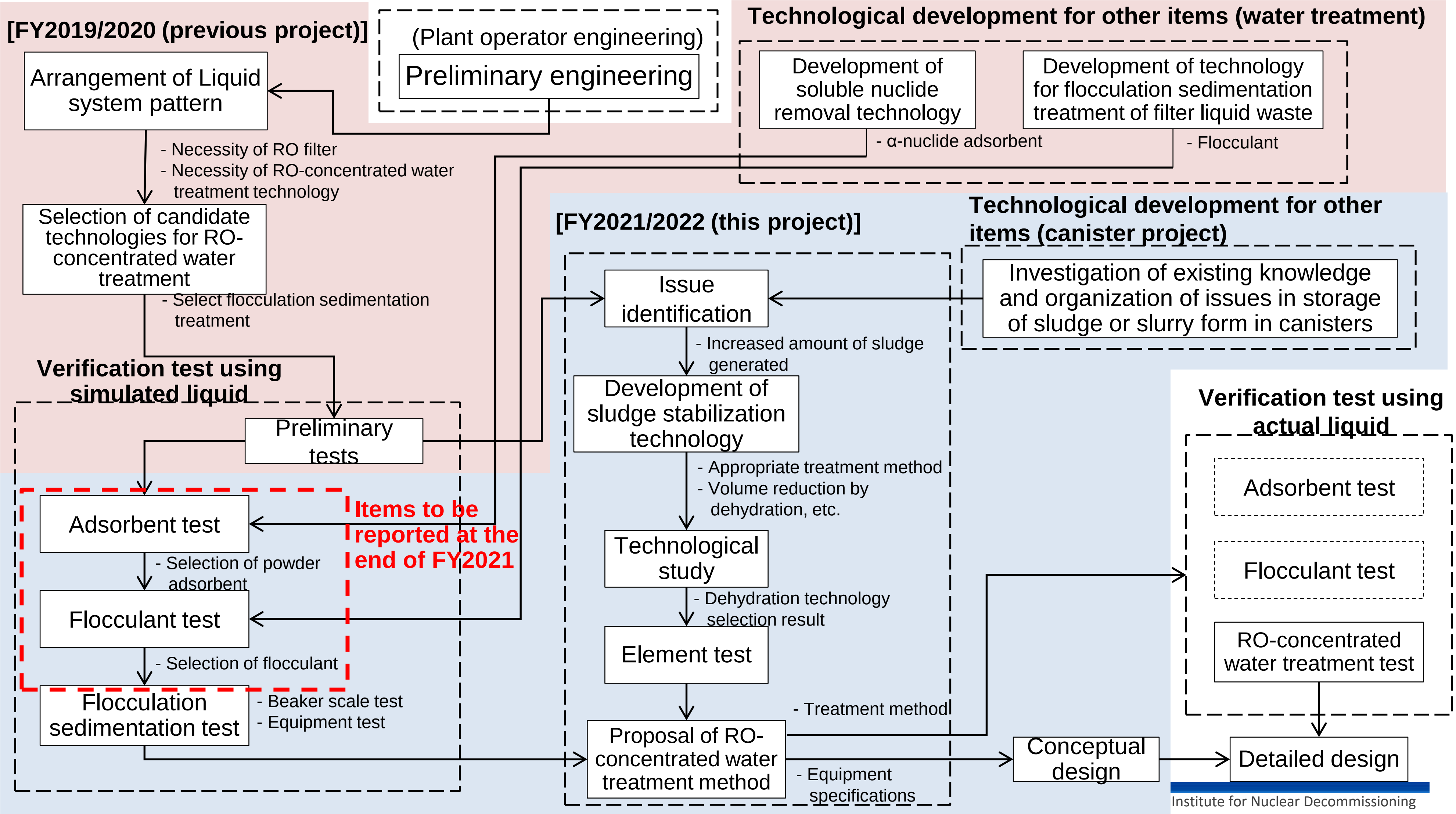
[Future plans]

- ✓ Preliminary tests for Pu, U, and Np will be implemented using the proposed test procedures to verify the behavior of α-nuclides under each water quality condition.
- ✓ Based on the results of the preparatory and preliminary tests, conditions for adsorption tests will be examined.
- ✓ An adsorption test will be conducted to evaluate the adsorption performance of each candidate adsorbent for α-nuclides.
- ✓ Based on the results of the adsorption test, the conceptual system design will be reviewed and a test plan using actual contaminated water will be devised.

7. Implementation details

② Development of RO-concentrated water treatment technology

- Flowchart of examination



[FY2019/2020 (previous project)]

(Plant operator engineering)

Preliminary engineering

Arrangement of Liquid system pattern

- Necessity of RO filter
- Necessity of RO-concentrated water treatment technology

Selection of candidate technologies for RO-concentrated water treatment

- Select flocculation sedimentation treatment

Verification test using simulated liquid

Preliminary tests

Adsorbent test

- Selection of powder adsorbent

Flocculant test

- Selection of flocculant

Flocculation sedimentation test

- Beaker scale test
- Equipment test

Items to be reported at the end of FY2021

Technological development for other items (water treatment)

Development of soluble nuclide removal technology

- α-nuclide adsorbent

Development of technology for flocculation sedimentation treatment of filter liquid waste

- Flocculant

[FY2021/2022 (this project)]

Issue identification

- Increased amount of sludge generated

Development of sludge stabilization technology

- Appropriate treatment method
- Volume reduction by dehydration, etc.

Technological study

- Dehydration technology selection result

Element test

Proposal of RO-concentrated water treatment method

- Treatment method
- Equipment specifications

Technological development for other items (canister project)

Investigation of existing knowledge and organization of issues in storage of sludge or slurry form in canisters

Verification test using actual liquid

Adsorbent test

Flocculant test

RO-concentrated water treatment test

Conceptual design

Detailed design

7. Implementation details

② Development of RO-concentrated water treatment technology

[Issues]

- As a treatment method for RO-concentrated water generated from RO filters, flocculation sedimentation is being examined, and it is necessary to establish treatment procedures.
- Powder adsorbents and flocculants that can be used for RO- concentrated water treatment have not been selected, and data on their nuclide adsorption performance and flocculation sedimentation performance are insufficient.

[Implementation details]

- Candidates for powder adsorbents and flocculants were selected based on literature research and past results of use in the existing water treatment facility.
- The nuclide removal performance of the powder adsorbent was evaluated by adsorption test.
- A beaker scale flocculation sedimentation test evaluates the flocculant performance of a flocculant on a powder adsorbent.

[Goals]

- Evaluation on the nuclide removal performance of the powder adsorbent and the flocculation sedimentation performance of the flocculant.
- Selection of applicable powder adsorbent and flocculant candidates.
- Development of treatment procedures for RO-concentrated water.

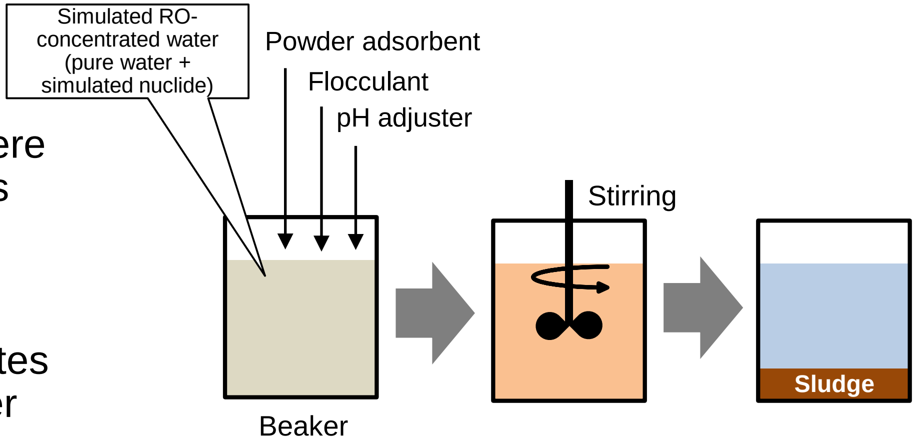
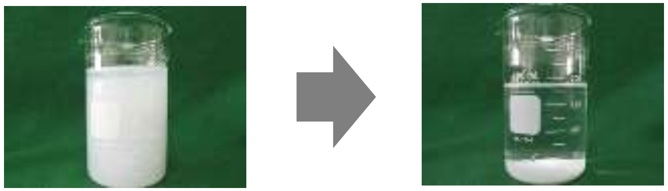


Figure RO-concentrated water treatment element test image


Before flocculation sedimentation treatment

After flocculation sedimentation treatment

Figure RO-concentrated water treatment element test

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ System configuration of sludge collection system

Solid components were collected from liquid waste generated from non-soluble nuclide removal equipment and RO filter equipment.

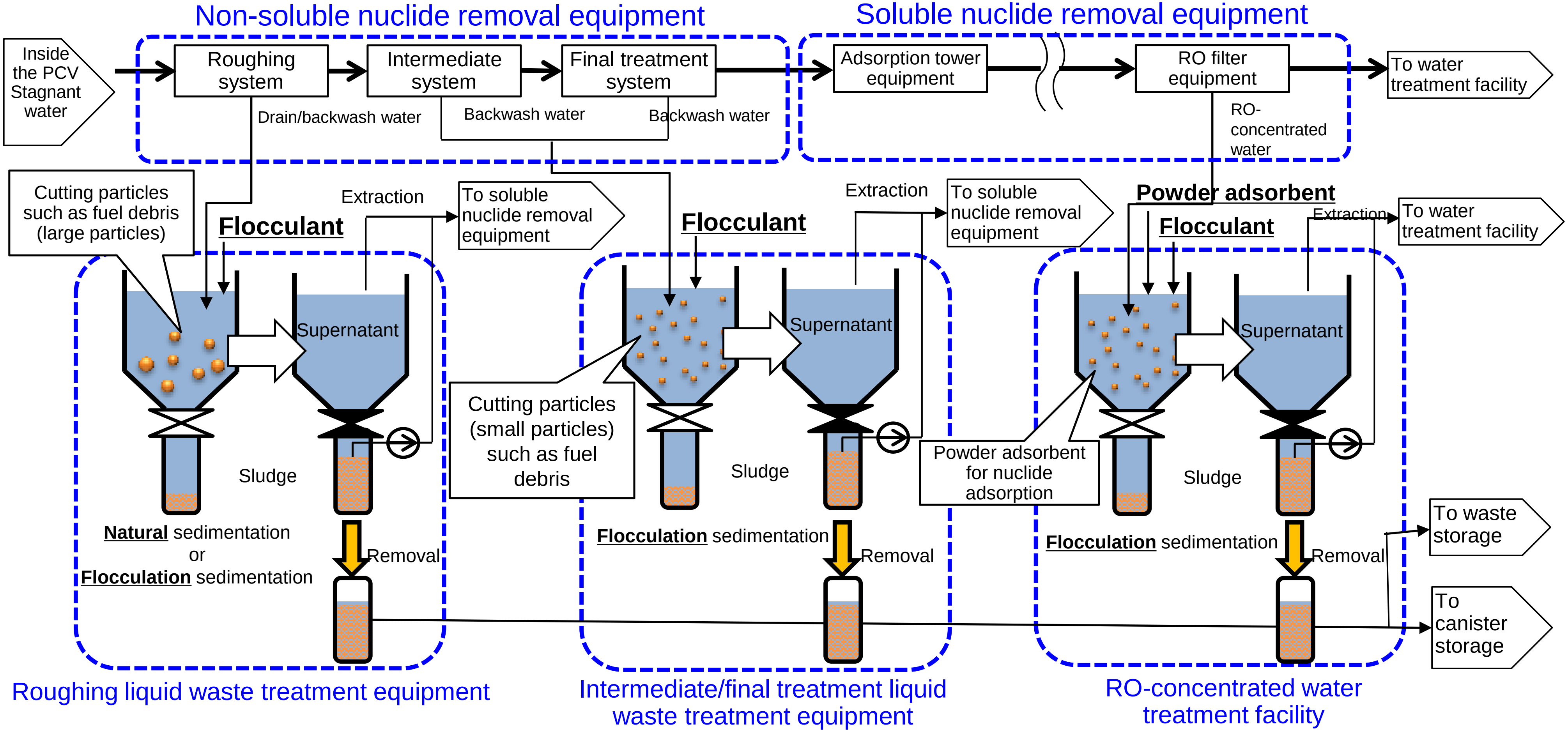


Figure System configuration concept of sludge collection system

7. Implementation details

② Development of RO-concentrated water treatment technology

- **Sludge collection system requirements**

The liquid to be treated and treatment requirements for each sedimentation equipment are shown below.

Table System requirements for each sedimentation equipment

*Trial calculation based on the water quality conditions set in the subsidy project

No.	Items	Roughing liquid waste treatment equipment	Intermediate/final treatment liquid waste treatment equipment	RO-concentrated water treatment facility
1	Water to be treated	Drain water and backwash water generated from roughing components	Backwash water generated from intermediate components and final treatment components (It is assumed that the intermediate system will be in bypass operation during fuel debris retrieval work)	RO-concentrated water generated from RO filter equipment
2	Contained particle component (for flocculation sedimentation)	Molten fuel, core structures, concrete components, other solids (Particle size 100 to several tens of μm)	Molten fuel, core structures, concrete components, other solids (Particle size several tens to 0.1 μm)	Powder adsorbent for nuclide adsorption
3	Particle concentration	Thousands to 10000 ppm From filter element test results	Thousands to 10000 ppm From filter element test results	Hundreds to 10000 ppm From adsorbent element test results
4	Treatment flow rate	Intermittent drain: 100 m^3/year Regular drain: 1300 m^3/year From filter element test results	0.1 μm UF filter: 10 m^3/year 0.05 μm UF filter: 100 m^3/year From filter element test results	8 to 11 m^3/day Fuel debris retrieval water Calculated assuming an RO filter concentration ratio of 2 to 3 times at (22 m^3/day)
5	Treatment flow rate requirements	Intermittent drain: 1.0 m^3/day or more Regular drain: 8.2 m^3/day or more Flow rate to enable treatment at 80% equipment availability	0.1 μm UF filter: 0.063 m^3/day or more 0.05 μm UF filter: 0.63 m^3/day or more Flow rate to enable treatment at 80% equipment availability	14 m^3/day or more Flow rate to enable treatment at 80% equipment availability
6	Operating method	Batch treatment Assuming completion of 1 batch treatment in 1 day		

7. Implementation details

② Development of RO-concentrated water treatment technology

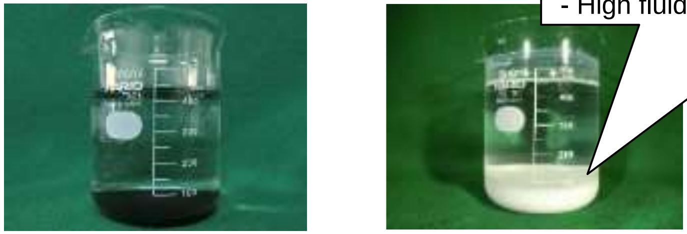
[Issue] A large amount of flocculated sediment (hereinafter referred to as sedimentation sludge) are generated from a sedimentation tank.

Assuming that the sludge collection container is a unit can size (Φ200 x H400 mm), the required number of times of discharging is calculated as shown in the table below

Table Number of discharge of sludge collection container

Liquid waste to be treated	Number of collection containers [pieces/day]
Roughing liquid waste	1
Final treatment liquid waste	8
RO-concentrated water	111

Sedimentation sludge properties
- The amount generated is about 10 to 15 vol% of the initial liquid volume
- The moisture content is about 90%
- High fluidity



RO-concentrated water treatment test
*Magnetite 1000 ppm

Filter liquid waste flocculation sedimentation test
*SiO_2 10000 ppm

Figure Appearance of liquid waste after flocculation sedimentation treatment

- ➢ The amount of sedimentation sludge generated from the sedimentation tank for filter liquid waste treatment is relatively small, and it is estimated that the number of times it is discharged is on the order that can be applied to the actual equipment
- ➢ The amount of sedimentation sludge generated from the sedimentation tank for RO-concentrated water treatment is enormous and needs to be discharged an enormous number of times, so improvement is necessary

Proposed plan

① Selection of adsorbent with higher removal performance
② Examination of optimizing the amount of powder adsorbent to be added
③ Examination of optimizing the amount of flocculant to be added
④ Review of sedimentation treatment procedures and equipment
⑤ Examination of volume reduction treatment of sedimentation sludge by dehydration

① to ④ ⇒ Examine via element tests
⑤ ⇒ Perform element tests after literature research and theoretical study
*Examined in Item ③

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ Adsorbent test

[Purpose] Selection of adsorbents applicable to RO-concentrated water treatment
[Implementation details] The adsorption performance of the powder adsorbent was evaluated by an agitated batch test.

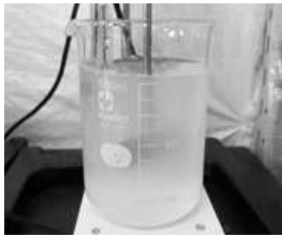


Figure Adsorbent test (titanic acid)

Examination of test system

When evaluating the adsorption performance of adsorbents, it is common to conduct an immersion batch test, but since RO-concentrated water is agitated and treated in a sedimentation tank, a test system that simulates the actual equipment was adopted.

Selection of powder adsorbent

The adsorbents used in this test were selected as shown in the table below, referring to the results of literature research, the results of use in the Fukushima Daiichi NPS, and the adsorption rate data obtained in tests up to fiscal 2020. Adsorbents reported to have high removal performance against α-nuclides (Pu, Am, etc.) were selected.

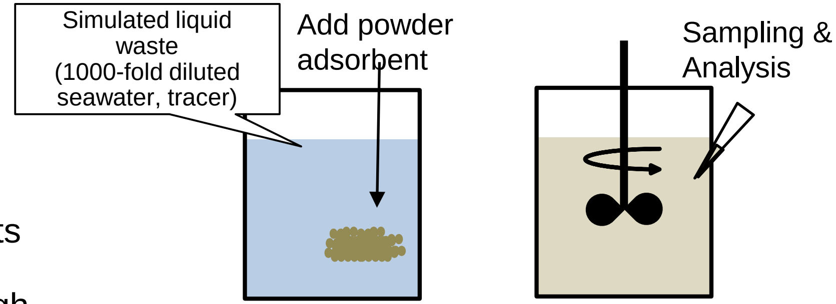


Figure Stirring type adsorption test Image

Table Selection results of powder adsorbent*

No.	Adsorbent type	Particle size	Remarks
1	Titanic acid	4.08 μm	There is the use results at the Savannah River Site in the United States (batch treatment)
2	Titanium silicate	8.55 μm	There are the use results in the Fukushima Daiichi NPS
3	Activated carbon	Several μm	There are the use results in the Fukushima Daiichi NPS
4	Hematite	1.32 μm	Iron oxide-based adsorbent. There is a report of high Kd (partition coefficient) for α-nuclides such as Pu.
5	Magnetite	1.96 μm	An iron oxide-based adsorbent similar to hematite.

*Since HOT test is difficult with this test method, all tests are performed as COLD test. For α-nuclides (Pu, etc.) that are difficult to simulate with a COLD tracer, item ① soluble adsorption rate data is referred.

*Selected from α-nuclide adsorbents Selected from adsorbents that cannot be formed into granules and are difficult to fill in adsorption towers

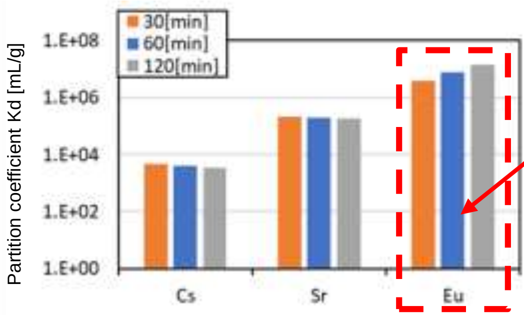
*Particle size of activated carbon not yet measured (to be implemented in the future)

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ Adsorbent test results

Test conditions
Test solution: 1000-fold diluted seawater, pH: 7
Tracer: Cs (0.1 ppm), Sr (0.1 ppm), Eu (0.5 ppm)
Adsorbents: right figure (particle size: several tens to several μm)
Concentration of adsorbent added: 20 to 500 ppm
Stirring conditions: 150 rpm × 30, 60, 120 min



Verified decrease in tracer concentration even after 30 minutes under the condition that the amount of adsorbent added was small

However, in almost all test cases, verified that adsorption equilibrium was reached within 30 minutes

Figure Adsorbent test results (titanic acid 20 ppm) Change in partition coefficient over time

Eu removal performance of Kd=1E6 to 7 was verified by titanic acid and titanium silicate.

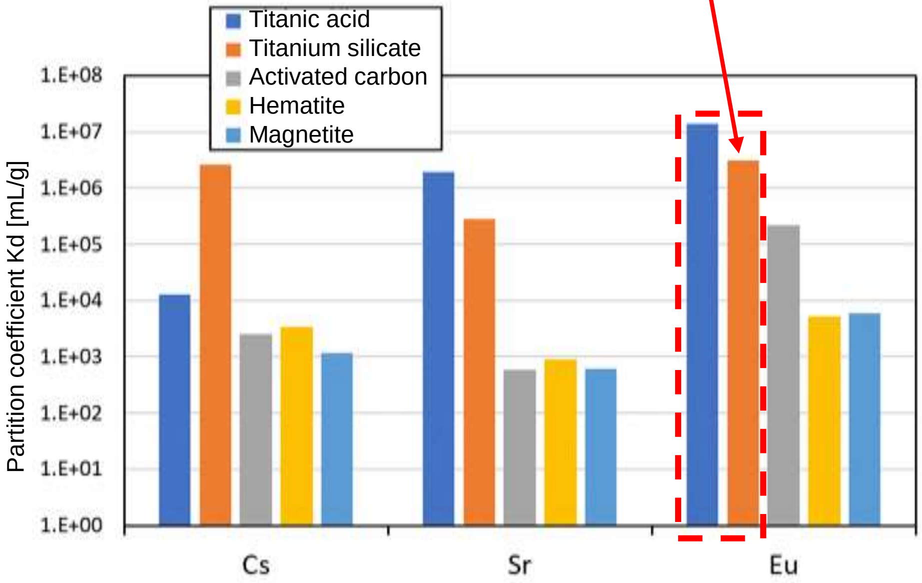


Figure Adsorbent test results: Partition coefficient

Titanic acid has a Kd of about 1E+7 and has the highest Eu removal performance.
Titanium silicate has a Kd of 1E+6 to 7 and has the second highest Eu removal performance after titanic acid.
Activated carbon has a Kd of about 1E+5 to 6, and hematite and magnetite both have a relatively low Kd of about 1E+3 to 4.

In addition, the Kd of titanic acid with respect to Sr and the titanium silicate with respect to Cs and Sr are comparable (compared to the Kd of Eu), and the Kd with respect to Cs and Sr of activated carbon, hematite and magnetite are relatively low.

The adsorption rate is fast and it is assumed that the adsorption equilibrium will be reached within 30 minutes.

7. Implementation details

② Development of RO-concentrated water treatment technology

- **Adsorbent test results**

The additive amount required to satisfy the removal requirement (DF100) was evaluated.

Evaluation of required additive amount of powder adsorbent

No.	Adsorbent	Partition coefficient [mL/g] [*1]			Additive amount[*2]	Treatment time [*3]
		Cs	Sr	Eu (Am)		
1	Titanic acid	1.3.E+04	1.9.E+06	1.4.E+07	<50 ppm	<30 min
2	Titanium silicate	2.6.E+06	2.8.E+05	3.2.E+06	50ppm	
3	Activated carbon	2.5.E+03	5.8.E+02	2.2.E+05	200 ppm<	
4	Hematite	3.5.E+03	9.1.E+02	5.2.E+03	1000 ppm<	
5	Magnetite	1.1.E+03	6.0.E+02	5.9.E+03	1000 ppm<	

[*1] Results with relatively high removal performance are shown in red. No.3 is a relatively high result, but is inferior to No.1 and 2, so it is shown in light red.

[*2] In this test, the tracer concentration was set to be higher than the actual water quality, so it is possible that the amount of adsorbent added can be reduced for liquid waste with lower concentration. It is necessary to evaluate the adsorption isotherm separately and evaluate the partition coefficient in each concentration region.

[*3] If the solution can be uniformly agitated, the treatment time is assumed to be short (approximately 30 min) because the adsorption rate in the initial stage is fast.

Titanic acid (No. 1) and titanium silicate (No. 2) have high removal performance against Eu, and the required additive amount was evaluated as several tens of ppm.
Since it is possible to reduce it from the 1000 ppm that was assumed in the past, it is thought that the amount of sludge generated can be reduced accordingly.

It was verified that the iron oxide-based (No. 4, 5) have a relatively low Eu removal rate.
→ Compared to titanic acid, etc., the amount to be added must be increased (several thousand ppm or more).

Since the removal rate for Cs and Sr is low, it is possible to selectively remove only α-nuclides

When selectively removing only α-nuclides → Select activated carbon (magnetite)
When removing total activity including α-nuclides → Select titanic acid and titanium silicate

7. Implementation details

② Development of RO-concentrated water treatment technology

- **Adsorbent selection policy**

  In addition to the removal performance of α-nuclides, the removal performance of Cs and Sr is considered to select the adsorbent.

Liquid system configuration plan 1

The liquid system shall be a system capable of selectively removing only α-nuclides and not removing Cs and Sr as much as possible

- The α-activity concentration shall be reduced to a level that can be discharged to the existing water treatment facility
- β and γ nuclides such as Cs and Sr are removed by the existing water treatment facility
- If only α-nuclides are sludged and captured, the dose of sludge can be kept low
- The shielding function required for sedimentation equipment and sedimentation sludge treatment equipment can be kept low, contributing to the reduction of worker exposure

Candidate powder adsorbents: activated carbon, (titanic acid, magnetite)

Adsorbents with high removal rate of α-nuclide (Eu) and low removal rate of Cs and Sr are selected
Considering the Eu removal rate, there is a concern that the additive amount must be increased (several hundred ppm)
→The amount of sludge generated increases

Even when retrieving fuel debris, if the Cs and Sr concentrations are at a level that can be treated with the existing water treatment facility, the only activity that needs to be reduced in the liquid system is α-nuclides. If a powder adsorbent capable of adsorbing and removing Cs and Sr is used for RO-concentrated water treatment, a strong shielding function will be required for the equipment itself, leading to an increase in the scale of the equipment. For the above reasons, it is assumed that it is necessary to select a powder adsorbent capable of selectively removing only α-nuclides. As the powder adsorbent for selective removal of α-nuclides, select a powder adsorbent that has high Eu removal performance and low adsorption performance for Cs and Sr.

7. Implementation details

② Development of RO-concentrated water treatment technology

- **Adsorbent selection policy**

  In addition to the removal performance of α-nuclides, the removal performance of Cs and Sr are considered to select the adsorbent

Liquid system configuration plan 2

The liquid system shall be a system capable of reducing the concentration of total activity, including α-activity

- The total activity concentration including α-activity concentration shall be reduced to a level that can be discharged to the existing water treatment facility
- Since Cs and Sr, which are dominant as activity, are also sludged and captured, the dose of the sedimentation sludge will be high
- From the viewpoint of worker exposure prevention, the level of shielding function required for sedimentation equipment and sedimentation sludge treatment equipment will increase

Candidate powder adsorbents: Titanic acid *, titanium silicate

*Titanic acid has a low Cs removal rate

Adsorbents with high removal rates for α-nuclides (Eu), Cs, and Sr are selected
Due to the high Eu removal rate, the additive amount is evaluated to be relatively small (<several tens of ppm)

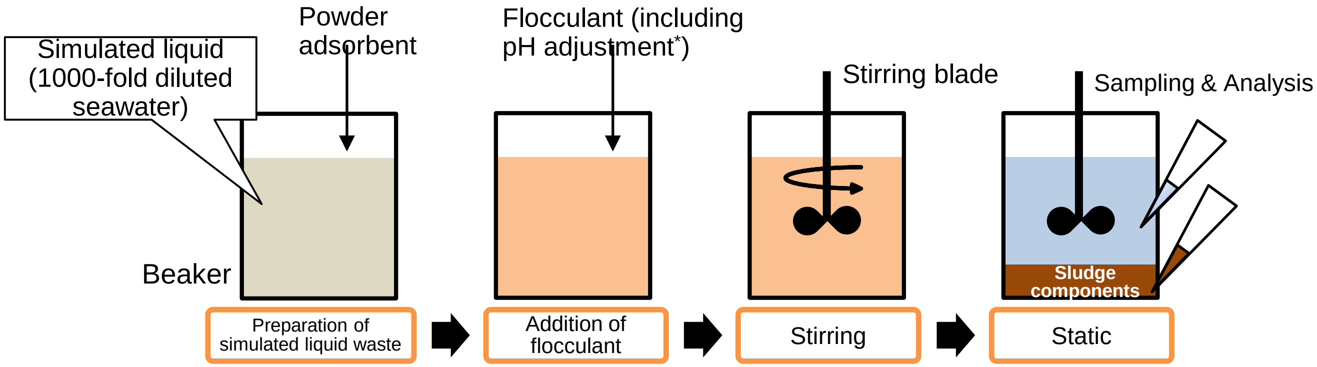
If the total activity concentration is set as a system requirement for being discharged to the existing water treatment facility, it will be necessary to reduce the concentrations of Cs and Sr, which are assumed to be predominantly radioactive, in addition to α-nuclides. Therefore, it is assumed that it is necessary to select a powder adsorbent that can reduce the total activity. As a powder adsorbent for total activity removal, select a powder adsorbent with high removal performance for all of Cs, Sr, and Eu.

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ Flocculant test

[Purpose] Selection of flocculants applicable to RO-concentrated water treatment
[Implementation details] Flocculation sedimentation treatment for the simulated liquid to which the powder adsorbent is added was performed.



Simulated liquid (1000-fold diluted seawater)

Powder adsorbent

Flocculant (including pH adjustment*)

Stirring blade

Sampling & Analysis

Beaker

Sludge components

Preparation of simulated liquid waste ➡ Addition of flocculant ➡ Stirring ➡ Static

Figure Flocculant test Image

Figure Flocculant test Appearance of equipment

*The tracer removal rate is not evaluated because the main item to be verified is whether the powder adsorbent flocculates and settles. Therefore, no tracer is added.

*All of the flocculants selected this time are acidic reagents, and it is necessary to adjust to the neutral region after addition, so sodium hydroxide solution is used.

Examination of test system

A stirring jar test similar to the adsorbent test is adopted.
It simulates a scaled-down system of treatment in an actual flocculation sedimentation tank.

Selection of flocculant

Flocculant was selected as shown in the table on the right based on the results of the literature research and the results of FY2020 test.
In addition to the flocculation sedimentation performance, redundancy with respect to the ionic strength of liquid waste was also considered.

Table Results of flocculant selection

No.	Flocculant	Remarks
1	High base PAC	There are reports of high treatment performance for liquid waste with low ionic strength
2	Sulfuric acid Aluminum	Preliminary project test verifies high treatment performance for liquid waste containing boric acid

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ Flocculant test results

The amount of sedimentation sludge generated after the flocculation sedimentation treatment was less dependent on the type of flocculant, and largely dependent on the type of powder adsorbent.

Amount of sedimentation sludge generated

Conventionally, when the amount of powder adsorbent added was 1000 ppm, the amount of sludge generated was assumed to be about 10 vol%

Titanic acid, activated carbon, magnetite → All results were found in 0.2 to 0.3 vol%.

Titanium silicate, hematite →The results of order of several vol% were frequently found(varies depending on test conditions).

Table Flocculant test conditions (adsorbent/flocculant)

Powder adsorbent*[1]		Flocculant*[2]			
Types	Concentration [ppm]	Types	Concentration [ppm]	Types	Concentration [ppm]
Titanic acid	50	High base PAC	100	Aluminum sulfate	20
Titanium silicate	50		1000		200
Activated carbon	200		1000		50
Hematite	500		1000		1000
Magnetite	500		1000		50

*1 Conditions set based on adsorbent test results

*2 Conditions set based on the preparatory test

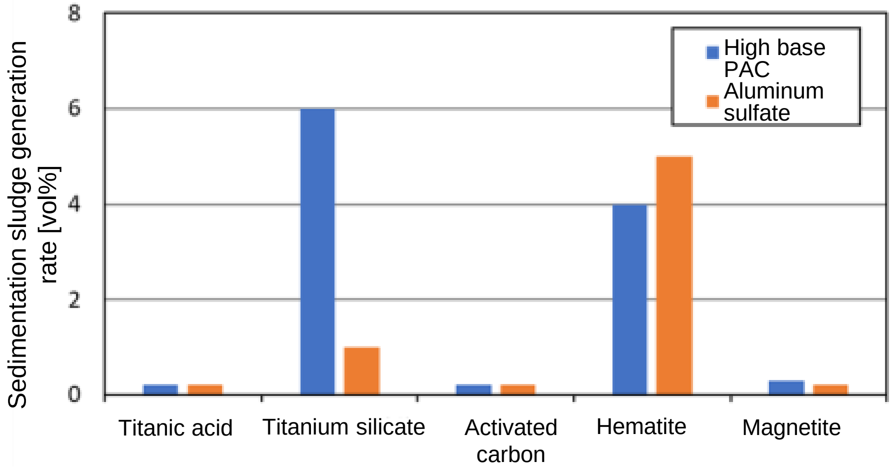


Figure Flocculant test results Sedimentation sludge volume

There was no significant difference between high-base PAC and aluminum sulfate in terms of the amount of sedimentation sludge generated.

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ Flocculant test results

- A large percentage of particles settle within 10 minutes after flocculation sedimentation treatment
- It was evaluated that it is possible to reduce the SS concentration to about 10 ppm by adding a flocculant.
- It was difficult to reduce the SS concentration to 1 ppm or less without depending on the amount of powder adsorbent added.

  *Because the turbidity measurement results were converted to SS concentration, there is a possibility that the region of 10 ppm or less could not be evaluated appropriately

- It was verified that the less the SS components, the narrower the adjustment range of the flocculant concentration.

→ It will be difficult to adjust the amount of flocculant added in actual equipment operation.

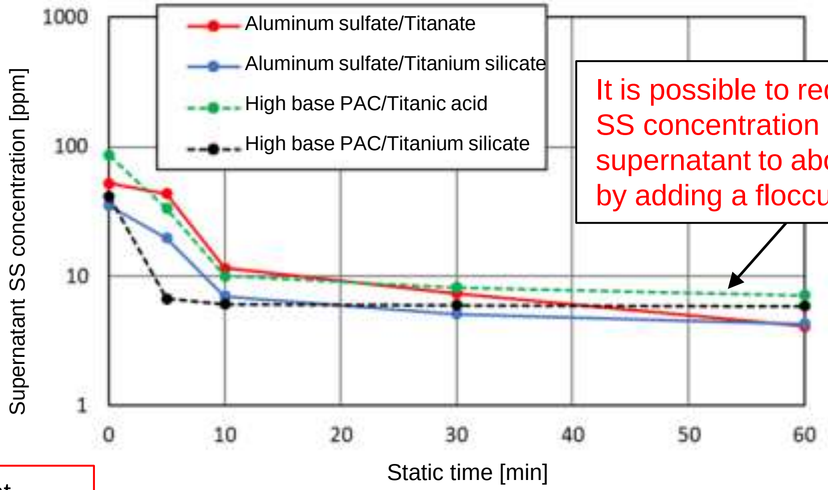
Table Flocculant test conditions (adsorbent/flocculant)

Powder adsorbent*1		Flocculant*2			
Types	Concentration [ppm]	Types	Concentration [ppm]	Types	Concentration [ppm]
Titanic acid	50	High base PAC	100	Aluminum sulfate	20
Titanium silicate	50		1000		200
Activated carbon	200		1000		50
Hematite	500		1000		1000
Magnetite	500		1000		50

*1 Conditions set based on adsorbent test results

*2 Conditions set based on the preparatory test

The figure on the right shows the transition of SS concentration



It is possible to reduce the SS concentration of the supernatant to about 10 ppm by adding a flocculant

Figure Flocculant test results Transition of SS concentration (titanic acid, titanium silicate)

No difference was observed in sedimentation behavior of particles between high base PAC and aluminum sulfate. Aluminum sulfate was selected as the flocculant because it was verified to have high flocculation and sedimentation properties in the previous project, even when water quality fluctuated due to boric acid solution conditions.

7. Implementation details

② Development of RO-concentrated water treatment technology

➢ Examination on the discharge of supernatant to the existing water treatment facility

Acceptance requirements for the existing water treatment facility were examined mainly based on the past results of accepting stagnant water in the process main building (PMB).

Table Tentative set values for acceptance requirements for the existing water treatment facility*

Items	Set value	Test results	Remarks
Cl	<700 ppm	<17 ppm	It is assumed that if the concentration of each ion in the circulating cooling water is below the assumed water quality conditions, the concentration in the supernatant water will not exceed this item. However, regarding the Cl concentration, it is necessary to examine the possibility of treating high Cl concentration stagnant water such as torus room water.
Ca	<50 ppm	-	
Mg	<30 ppm	-	
SO_4	-	Scheduled to acquire data	When aluminum sulfate is used, most of the sulfate ions remain in the supernatant.
pH	7 to 8.5	6.7 to 7.2	In the process of flocculation sedimentation treatment, the pH of the supernatant is adjusted to about pH 7, so it is assumed that this item will be satisfied.
SS concentration	<20 ppm	<10 ppm	Since it is assumed that the SS concentration can be reduced to about 10 ppm or less by flocculation sedimentation treatment, the SS concentration reference is assumed to satisfy the acceptance requirements. However, since the particles remaining in the supernatant have a high amount of activity, it is necessary to separately evaluate whether they meet the requirements for acceptance based on the activity concentration.
Total α concentration	<7.4E+0 Bq/L	99% removal rate against initial concentration (after 0.45 μm filtration)	Set from the maximum α concentration of the inlet water of the existing water treatment facility. Issues for future study. It is assumed that it can be evaluated from the total α concentration of the input water and the removal performance and additive amount of the powder adsorbent. Evaluation is also necessary from the viewpoint of public exposure dose (DF100) and announcement density.

It is estimated that the supernatant water after the flocculation sedimentation treatment can be discharged to the existing water treatment facility, considering the assumed treatment results of the existing water treatment facility. However, removal of residual particles in the supernatant water, such as by filtration, may be necessary.

*Primarily set based on the past results of accepting PMB stagnant water in FY2018.
Acceptance requirements for the existing water treatment facility during the fuel debris retrieval work is an issue for future study

7. Implementation details

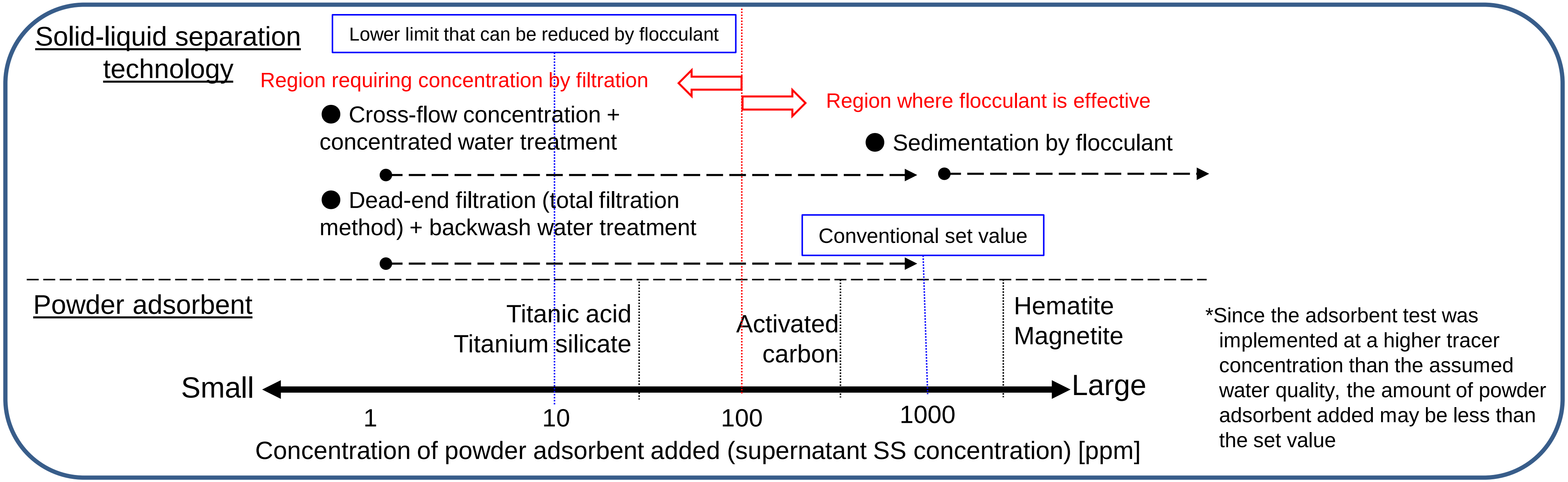
② Development of RO-concentrated water treatment technology

➢ Examination of solid-liquid separation of powder adsorbent when the amount added is small

It was suggested that the amount of powder adsorbent added could be reduced from the conventional set value (1000 ppm). Since flocculants are not applicable to the treatment of liquid waste with a small amount of SS, a system that concentrates the SS components as a pretreatment was examined.

Concentration of powder adsorbent added	Solid-liquid separation technology
Thousands to hundreds of ppm	→ Solid-liquid separation by flocculant is effective
Hundreds to several tens of ppm	→ Solid-liquid separation by flocculant (concentration treatment if necessary)
Several tens of ppm or less	→ Concentration treatment + flocculant

*It is assumed that cross-flow filtration and dead-end filtration are effective for concentration treatment



Solid-liquid separation technology

Lower limit that can be reduced by flocculant

Region requiring concentration by filtration

Region where flocculant is effective

● Cross-flow concentration + concentrated water treatment

● Sedimentation by flocculant

● Dead-end filtration (total filtration method) + backwash water treatment

Conventional set value

Powder adsorbent

Titanic acid
Titanium silicate

Activated carbon

Hematite
Magnetite

Small ← → Large

1, 10, 100, 1000

Concentration of powder adsorbent added (supernatant SS concentration) [ppm]

*Since the adsorbent test was implemented at a higher tracer concentration than the assumed water quality, the amount of powder adsorbent added may be less than the set value

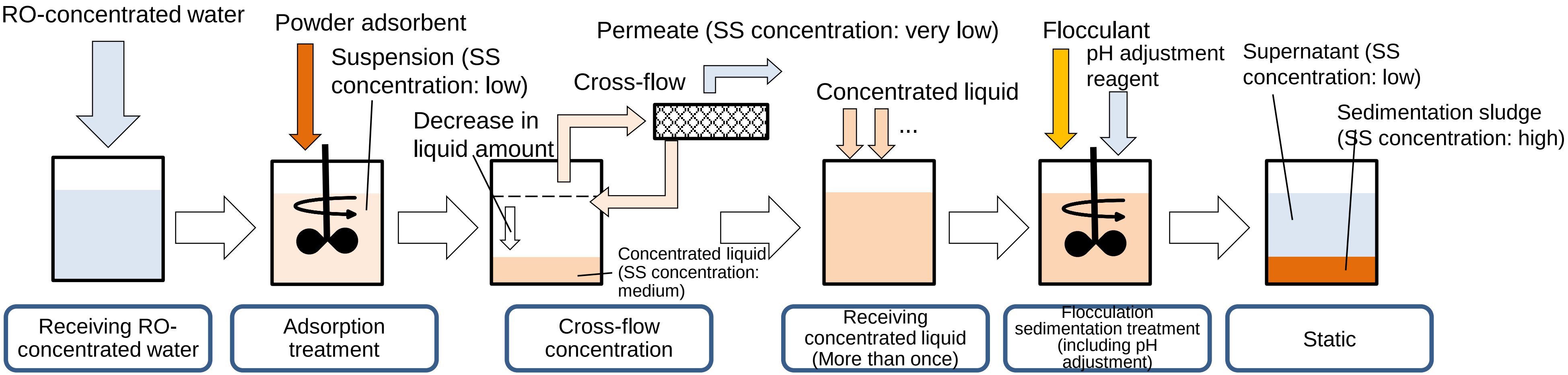
By concentrating the SS components through concentration treatment, solid-liquid separation using a flocculant is possible while reducing the amount of liquid waste

7. Implementation details

② Development of RO-concentrated water treatment technology

- **Examination of solid-liquid separation of powder adsorbent when the amount added is small**

Application of concentration treatment to RO-concentrated water treatment



RO-concentrated water

Powder adsorbent

Suspension (SS concentration: low)

Decrease in liquid amount

Cross-flow

Permeate (SS concentration: very low)

Concentrated liquid (SS concentration: medium)

Concentrated liquid

…

Flocculant

pH adjustment reagent

Supernatant (SS concentration: low)

Sedimentation sludge (SS concentration: high)

Receiving RO-concentrated water

Adsorption treatment

Cross-flow concentration

Receiving concentrated liquid (More than once)

Flocculation sedimentation treatment (including pH adjustment)

Static

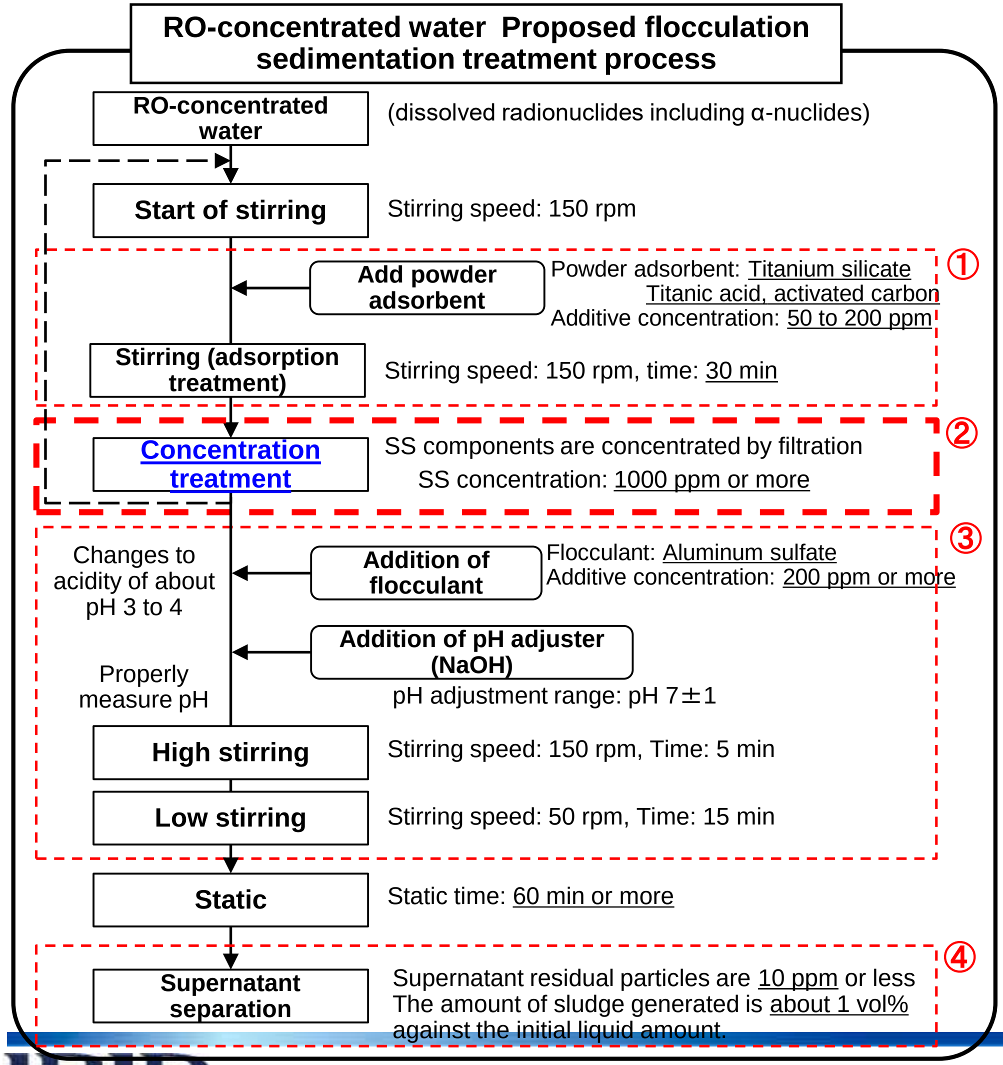
Figure Proposed procedures for RO-concentrated water treatment method when cross-flow concentration is applied

If the concentration of powder adsorbent added is low (dozens of ppm or less), cross-flow filtration is performed after adsorption treatment to separate moisture to reduce the liquid amount and concentrate SS components (approx. 1000 ppm or more).
After that, the concentrated liquid is subjected to flocculation sedimentation treatment to separate into supernatant and sedimentation sludge.
In addition, since the liquid amount is reduced by the concentration treatment, it can be assumed that the concentrated liquid for multiple times can be subjected to the flocculation sedimentation treatment at once. Therefore, the concentrated liquid is discharged into a tank for flocculation sedimentation treatment, and the treatment is performed after securing a certain amount of liquid.

⇒ Mass balance (water volume, sludge volume) in the system using each concentration treatment will be evaluated in the future.

7. Implementation details

② Development of RO-concentrated water treatment technology



RO-concentrated water Proposed flocculation sedimentation treatment process

RO-concentrated water (dissolved radionuclides including α-nuclides)

Start of stirring Stirring speed: 150 rpm

①

Add powder adsorbent Powder adsorbent: Titanium silicate
Titanic acid, activated carbon
Additive concentration: 50 to 200 ppm

Stirring (adsorption treatment) Stirring speed: 150 rpm, time: 30 min

②

Concentration treatment SS components are concentrated by filtration
SS concentration: 1000 ppm or more

③

Changes to acidity of about pH 3 to 4

Addition of flocculant Flocculant: Aluminum sulfate
Additive concentration: 200 ppm or more

Addition of pH adjuster (NaOH)
pH adjustment range: pH 7±1

Properly measure pH

High stirring Stirring speed: 150 rpm, Time: 5 min

Low stirring Stirring speed: 50 rpm, Time: 15 min

Static Static time: 60 min or more

④

Supernatant separation Supernatant residual particles are 10 ppm or less
The amount of sludge generated is about 1 vol% against the initial liquid amount.

(1) Adsorption treatment
Soluble nuclides are adsorbed by adding a powder adsorbent and stirring for a certain period of time.

(2) **Concentration treatment** (adding reflecting the examination results)
Powder adsorbents are concentrated, such as by cross-flow filtration or dead-end filtration.
* Receiving RO-concentrated water → ① → ② are repeated to secure a certain amount of concentrated liquid.

(3) Flocculation sedimentation treatment
A flocculant is added to the concentrated liquid, adjusted to a neutral region with a pH adjusting reagent, and agitated for a certain period of time to cause flocculation and sedimentation of the powder adsorbent. (RO-concentrated water sedimentation sludge generation)

(4) Discharge of supernatant and sedimentation sludge
Supernatant: Filtration to remove residual particles after extraction.
Sedimentation sludge: Extracted by pump or discharged in a collection container.

The volume reduction of sedimentation sludge by applying dehydration treatment will be examined in the development item 3 "Development of secondary waste treatment technology."

7. Implementation details

② Development of RO-concentrated water treatment technology

- Summary

[Results for this year]

- ✓ Adsorbent tests were implemented, and powder adsorbents with high removal performance against α-nuclides were selected. Activated carbon was evaluated as highly applicable for selective removal of α-nuclides, and titanic acid and titanium silicate were evaluated as highly applicable for removing total activity including α-nuclides.
- ✓ A flocculant test was implemented, and aluminum sulfate was selected as a flocculant for flocculation and sedimentation of the powder adsorbent. In addition, data on the supernatant and the amount of sedimentation sludge generated and its properties were obtained.
- ✓ The water quality conditions under which solid-liquid separation by flocculant is effective were organized. As a treatment method for low SS concentration liquid waste, which is difficult to separate solid-liquid using flocculant, a system applying concentration treatment was devised.
- ✓ Based on the results of this fiscal year, treatment procedures for RO-concentrated water were devised.

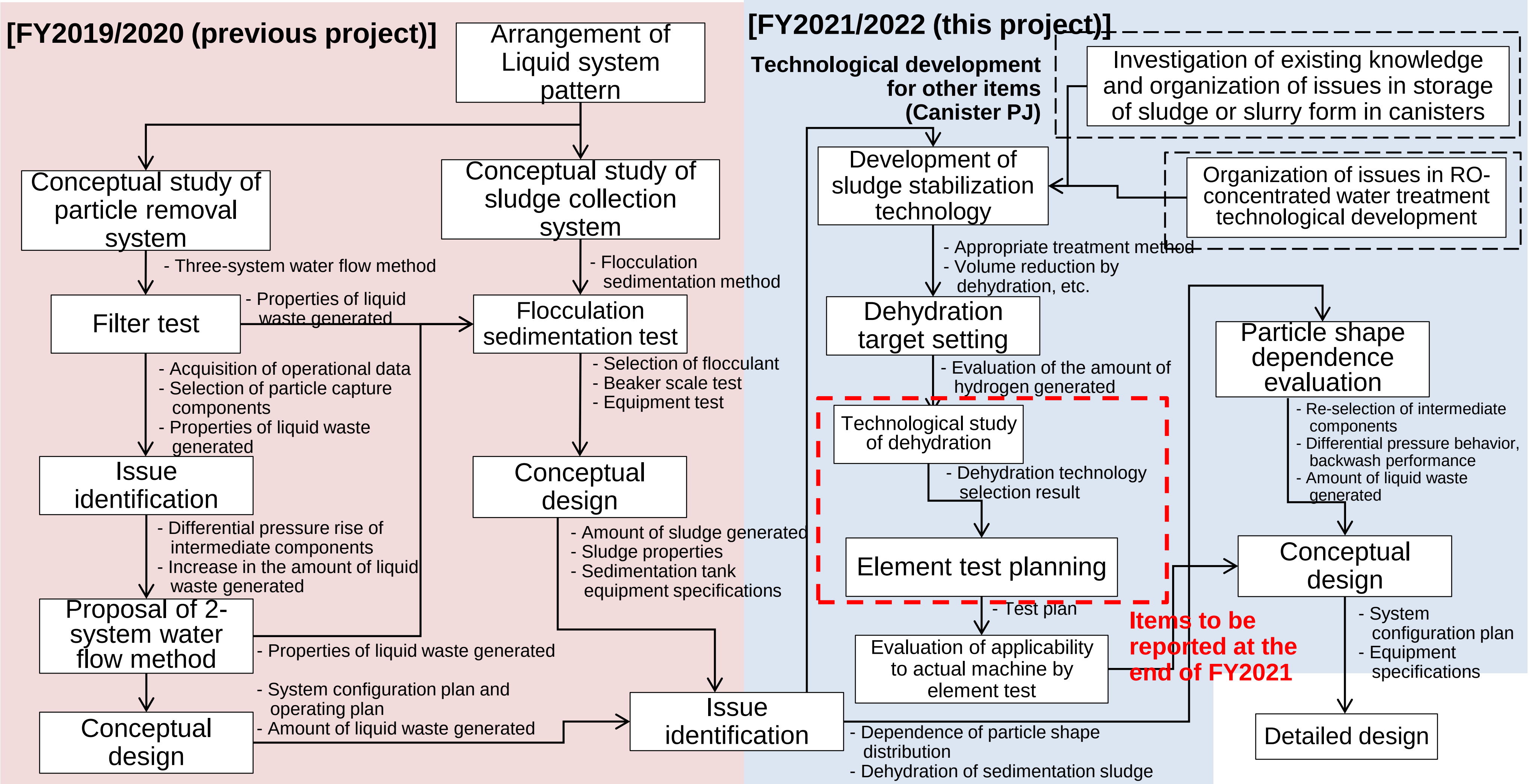
[Future plans]

- ✓ Beaker-scale RO-concentrated water treatment tests and equipment tests will be implemented to obtain test data.
- ✓ Based on the results of each test, a method for treating RO concentrated water will be devised, and a conceptual system design for the system will be implemented.

7. Implementation details

③ Development of secondary waste treatment technology

- Flowchart of examination



[FY2019/2020 (previous project)]

Arrangement of Liquid system pattern

Conceptual study of particle removal system

- Three-system water flow method

Filter test

- Properties of liquid waste generated
- Acquisition of operational data
- Selection of particle capture components
- Properties of liquid waste generated

Issue identification

- Differential pressure rise of intermediate components
- Increase in the amount of liquid waste generated

Proposal of 2-system water flow method

- Properties of liquid waste generated
- System configuration plan and operating plan
- Amount of liquid waste generated

Conceptual design

Conceptual study of sludge collection system

- Flocculation sedimentation method

Flocculation sedimentation test

- Selection of flocculant
- Beaker scale test
- Equipment test

Conceptual design

- Amount of sludge generated
- Sludge properties
- Sedimentation tank equipment specifications

Issue identification

- Dependence of particle shape distribution
- Dehydration of sedimentation sludge

[FY2021/2022 (this project)]

Technological development for other items (Canister PJ)

Investigation of existing knowledge and organization of issues in storage of sludge or slurry form in canisters

Organization of issues in RO-concentrated water treatment technological development

Development of sludge stabilization technology

- Appropriate treatment method
- Volume reduction by dehydration, etc.

Dehydration target setting

- Evaluation of the amount of hydrogen generated

Technological study of dehydration

- Dehydration technology selection result

Element test planning

- Test plan

Evaluation of applicability to actual machine by element test

Items to be reported at the end of FY2021

Particle shape dependence evaluation

- Re-selection of intermediate components
- Differential pressure behavior, backwash performance
- Amount of liquid waste generated

Conceptual design

- System configuration plan
- Equipment specifications

Detailed design

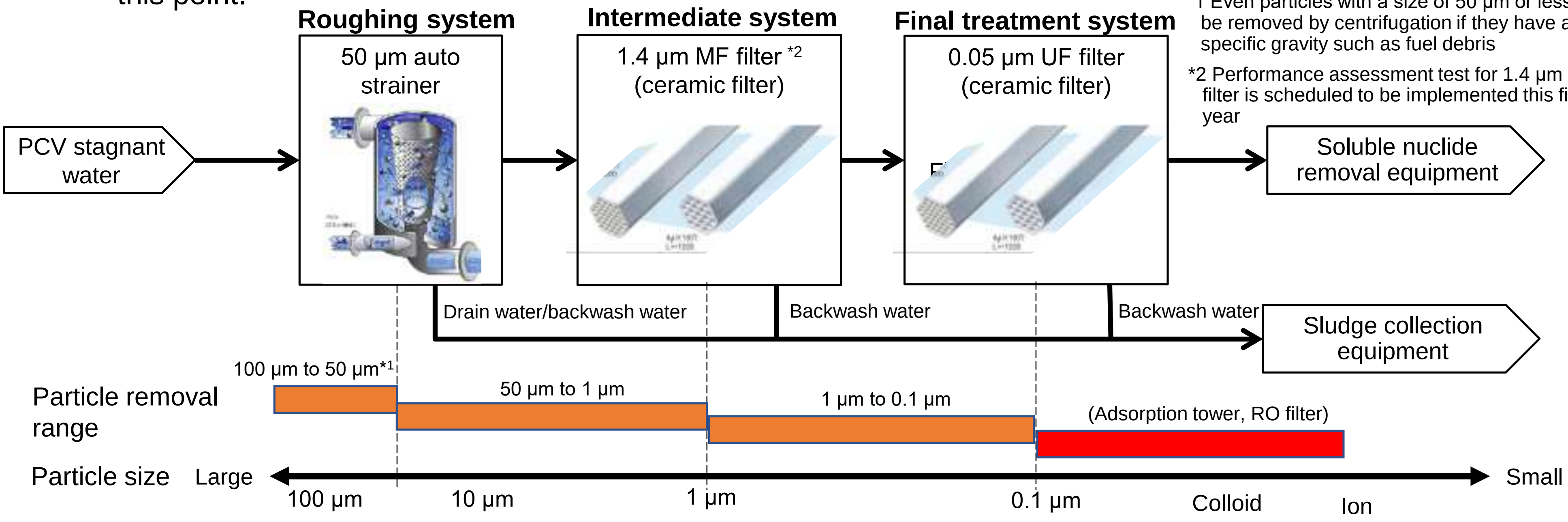
7. Implementation details

③ Development of secondary waste treatment technology

1) Characterization of liquid waste generated from particle removal system

- **System configuration of particle removal system**

The system configuration of particle removal system was examined based on the results of development up to this point.



Roughing system

50 µm auto strainer

Intermediate system

1.4 µm MF filter [*2] (ceramic filter)

Final treatment system

0.05 µm UF filter (ceramic filter)

PCV stagnant water → Roughing system → Intermediate system → Final treatment system → Soluble nuclide removal equipment

Drain water/backwash water

Backwash water

Backwash water

→ Sludge collection equipment

Particle removal range

100 µm to 50 µm[*1]

50 µm to 1 µm

1 µm to 0.1 µm

(Adsorption tower, RO filter)

Particle size Large ← → Small

100 µm

10 µm

1 µm

0.1 µm

Colloid

Ion

[*1] Even particles with a size of 50 µm or less can be removed by centrifugation if they have a large specific gravity such as fuel debris

[*2] Performance assessment test for 1.4 µm MF filter is scheduled to be implemented this fiscal year

Figure Conceptual diagram of the particle removal equipment system configuration

It is assumed that the fuel debris processing work performed in the PCV will atomize the processed matter, and particles of approximately 100 µm or less will float in the liquid and migrate to the liquid system.
Particles of 50 µm or larger are removed by the auto strainer in the roughing system. Particles with a high specific gravity (e.g., fuel debris, core structures) are also removed, where their centrifugal separation effect is significant. Small particles that have passed through the roughing system are removed by the MF and UF filters in the intermediate and final treatment systems, with the entire system removing particles down to 0.1 µm.
Captured particles are discharged by drain/backwash, and the liquid waste is treated in the sludge collection equipment.

7. Implementation details

③ Development of secondary waste treatment technology

1) Characterization of liquid waste generated from particle removal system

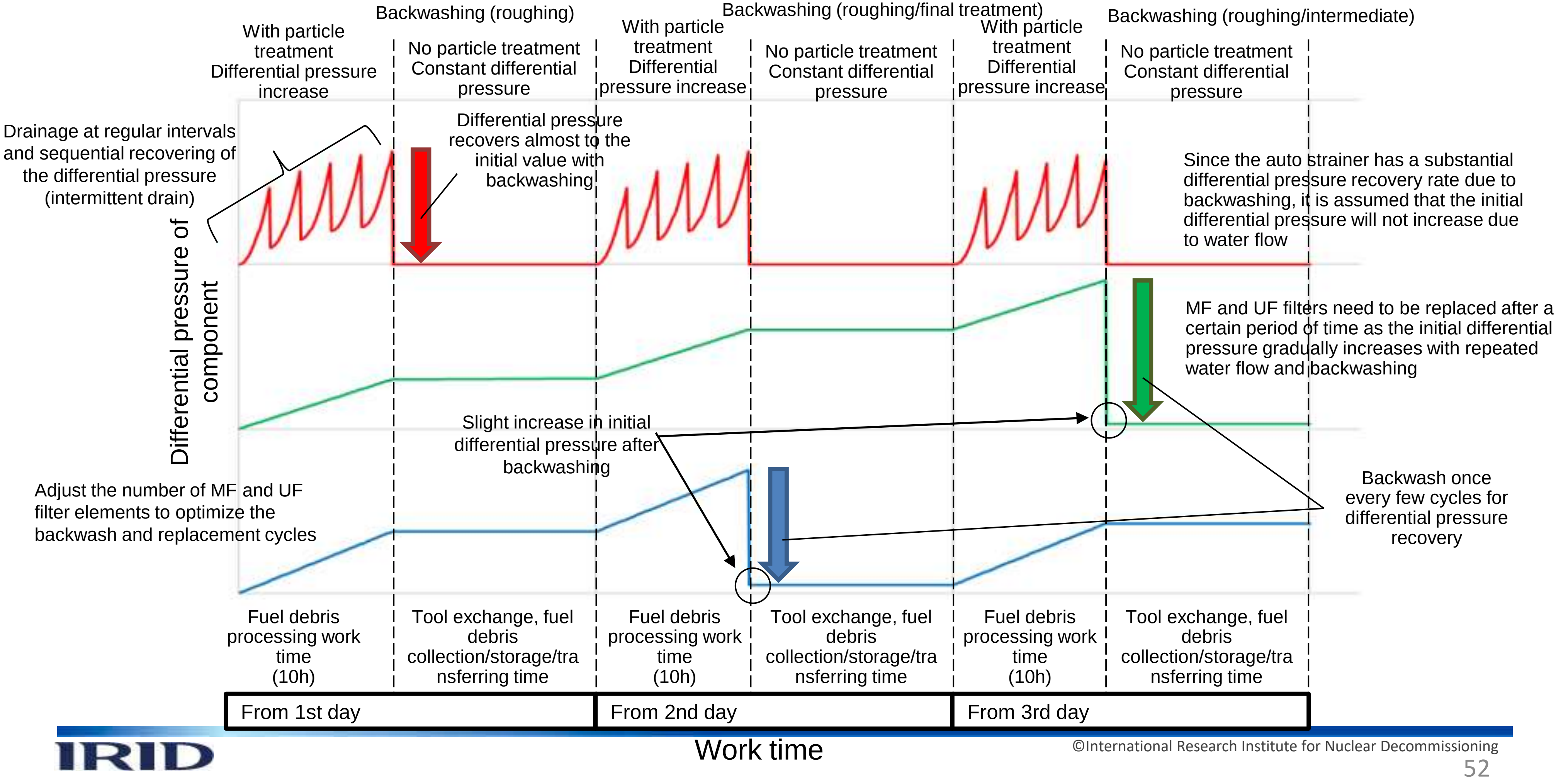
- **Examination of water flow cycle during fuel debris retrieval work**

3-system configuration (Roughing→ Intermediate → Final treatment)

🟥 : Roughing; 50 μm auto strainer

🟩 : intermediate; 1.4 μm MF filter

🟦 : Final treatment; 0.05 μm UF filter



Drainage at regular intervals and sequential recovering of the differential pressure (intermittent drain)

Differential pressure recovers almost to the initial value with backwashing

Since the auto strainer has a substantial differential pressure recovery rate due to backwashing, it is assumed that the initial differential pressure will not increase due to water flow

MF and UF filters need to be replaced after a certain period of time as the initial differential pressure gradually increases with repeated water flow and backwashing

Slight increase in initial differential pressure after backwashing

Adjust the number of MF and UF filter elements to optimize the backwash and replacement cycles

Backwash once every few cycles for differential pressure recovery

7. Implementation details

③ Development of secondary waste treatment technology

1) Characterization of liquid waste generated from particle removal system

- **MF filter water flow test**

[Issue]

The intermediate filters such as an auto strainer, a sintered metal filter, and a bag filter were selected and their applicability was evaluated by filter tests. However, both resulted in a significant increase in the component differential pressure during water flow due to the small effective filtration area and the small contribution of centrifugal separation effect to particles of only a few μm. As such, a highly applicable intermediate filter component has not yet been selected at this time.

[Purpose]

Evaluation of the applicability of a MF filter with pore size of 1.4 μm (ceramic filter) as an intermediate filter.

[Implementation details]

Implement a filter water flow test using a 1.4 μm MF filter to obtain operational data.

1.4 μm MF filter

- It was verified that the UF filter selected as the final treatment filter has a very large effective filtration area, leading to a gentle increase in differential pressure
- It is assumed that off-the-shelf identical ceramic filters with pore size of up to 1.4 μm can remove particles in the intermediate filter range
- The investigation confirmed that ceramic filters with pore sizes of 1.4 μm or larger are not available, or that only tube types with a small filtration area are available

7. Implementation details

③ Development of secondary waste treatment technology

1) Characterization of liquid waste generated from particle removal system

- **Particle shape dependence evaluation test**

[Issue]

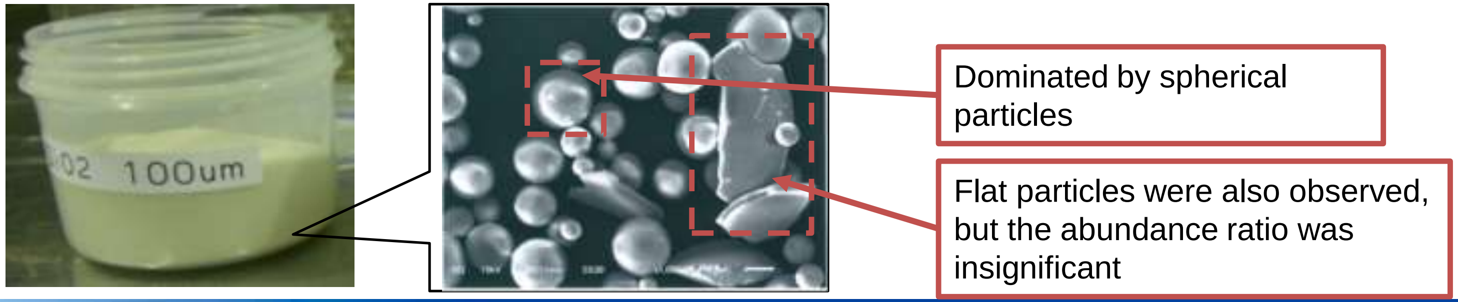
The effect of particle shape on filter performance has not been evaluated for the selected filter components in each system. All filter tests conducted up to the previous project used spherical particles, and there is a possibility that the filter performance was not evaluated in a conservative manner.

[Purpose]

Evaluation of the particle shape dependence of the filter performance for the selected filter components in each system.

[Implementation details]

A filter water flow test using spherical particles and non-spherical particles was conducted to evaluate particle size dependence by comparative examination.
In addition, properties of the backwash water, which are the input conditions to the sludge collection equipment were examined.



Dominated by spherical particles

Flat particles were also observed, but the abundance ratio was insignificant

Figure Particles used in the filter test of the previous project

7. Implementation details

③ Development of secondary waste treatment technology

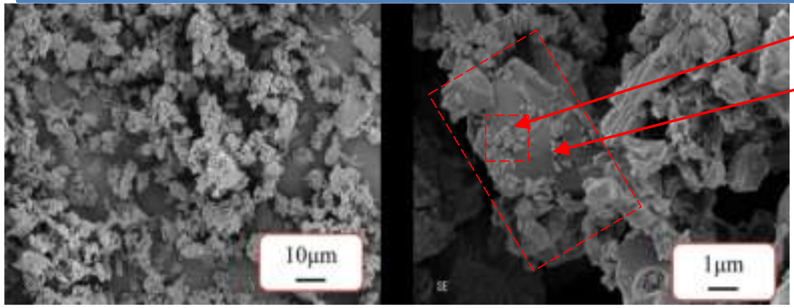
1) Characterization of liquid waste generated from particle removal system

- **Non-spherical particles used in filter test**

  Simulant particles were selected based on the assumption that the cutting debris generated by machining process is passed through a filter.

Simulant target

Powders generated by machining process test of simulant fuel debris in the implementation of the characterization project*



Relatively small particles: <1 μm

Relatively large particles: several μm

Figure. SEM image of a sample after cutting test of simulant fuel debris implemented in the characterization project (excerpt)

Particle size distribution

Formation of agglomerates of <1 μm powders on the surface of a large particle of the order of several to 10 μm

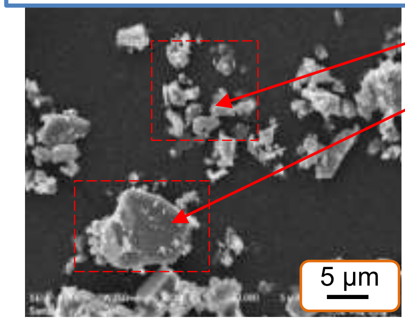
Particle Shape

Dominated by Block-like particles

*FY2018 Subsidy Project of “Development of Analysis and Estimation Technology for Fuel Debris Characterization”

Simulant particles

Non-spherical particles used in this study



Relatively small particles: <1 μm

Relatively large particles: several μm

Figure Simulant particles SEM image

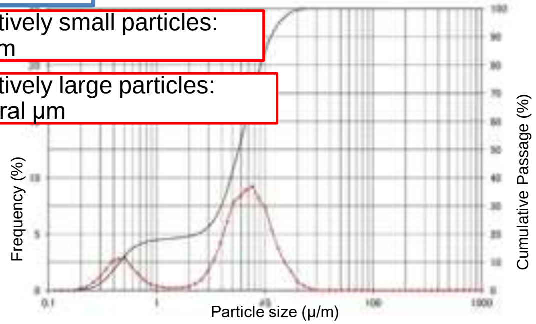


Figure Simulant particles Particle size distribution

Particle size distribution and shape are simulated based on the test report of the characterization project

Silica sand particles with a shape and particle size that are relatively similar to those used by particle manufacturers are used

Sizing to be performed as necessary

7. Implementation details

③ Development of secondary waste treatment technology

1) Characterization of liquid waste generated from particle removal system

- **Test system for water flow test**

For filter components, particle capture components of each system selected up to the previous project were used.
The 1.4 µm MF filter, which has been evaluated as having high applicability based on literature research, was only be used for the intermediate filter.

Main line: →

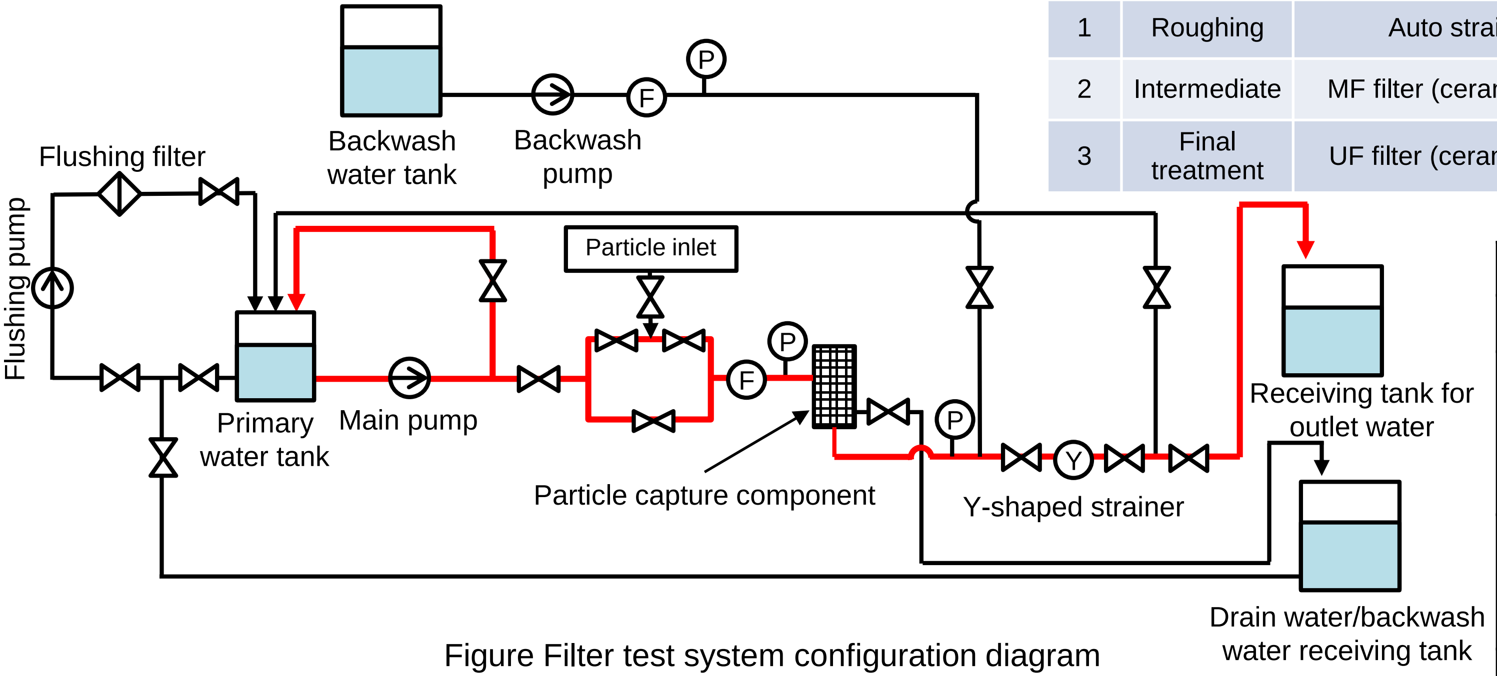


Figure Filter test system configuration diagram

Table list of particle capture components used

No.	System	Particle capture component	Filter mesh size
1	Roughing	Auto strainer	50 µm
2	Intermediate	MF filter (ceramic filter)	1.4 µm
3	Final treatment	UF filter (ceramic filter)	0.05 µm

Table Evaluation Items

No.	Items
1	Particle capture rate
2	Time of water flow until backwash pressure is reached (Differential pressure rising trend)
3	Backwashing performance (Differential pressure recovery performance)
4	Backwash water properties

In this test, the performance of each filter was evaluated, and property data on the liquid waste was obtained.

7. Implementation details

③ Development of secondary waste treatment technology

1) Characterization of liquid waste generated from particle removal system

- **Filter element replacement cycles**

  Replacement cycles for the particle capture components are assumed to be as follows

  Roughing system components: 2 times/year → Rate-limiting corrosion of materials due to chlorine

  Intermediate system components: Evaluation for this year* → Rate-limiting increase in initial differential pressure

  Final treatment system components: 5 to 8 times/year → Rate-limiting increase in initial differential pressure

  *It is assumed that it can be operated for a longer period of time because the pore size is larger than that of the final treatment system components

- **Filter element replacement**

  It is necessary to evaluate worker exposure due to residual particles in the housing during the replacement

  Fundamentally, a cartridge system will be adopted, and it is assumed that the entire housing will be replaced.

  The housing should be sufficiently shielded to prevent exposure to workers.

  Another possible method is to replace the elements remotely, but the feasibility of full remote control processing needs to be studied.

- **Examination of the amount of filter liquid waste and secondary waste generated**

  This shows the filter specifications and approximate results of the amount of liquid waste and secondary waste generated which was evaluated from the test results up to FY2020

Summary of filter performance assessment results based on previous element test results

System	Particle capture component		Number of required base	Number of elements required	Rated flow	Filtering accuracy	Removal efficiency	Approximate dimensions (per reactor)	Required area dimensions (per reactor)	Replacement frequency	Amount of secondary waste generated	Number of times drained/back washed	Amount of liquid waste generated
			Base/1 system	Number	m^3/h	µm or more	% or higher	Φ__ m × __ mH	__ mL × __ mW	times/year	kg/year	times/day	m^3/year
Roughing	Auto strainer	Intermittent drain	1	1	10 to 30	50	99	Φ0.5 m × 1.0 mH	1.4 mL × 1.4 mW	2	11	2.5	167
		Continuous drain	1	1	10 to 30	50	99	Φ0.5 m × 1.0 mH	1.4 mL × 1.4 mW	2	11	Continuous drain	1300
	Liquid cyclone		1	1	10	40	80	Φ0.4 m × 1.1 mH	1.6 mL × 1.6 mW	-	-	Continuous drain	2000
Intermediate	MF filter	1.4 µm				Scheduled to be tested in FY2022							
Final treatment	UF filter	0.1 µm	2	33	6.5	0.1	99	Φ0.21 m × 1.3 mH	3.0 mL × 1.6 mW	8	314	0.34	11
		0.05 µm	3	46	<23	0.05	99	Φ0.3 m × 1.4 mH	1.4 mL × 1.4 mW	5	253	0.23	98.6

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

[Issues]

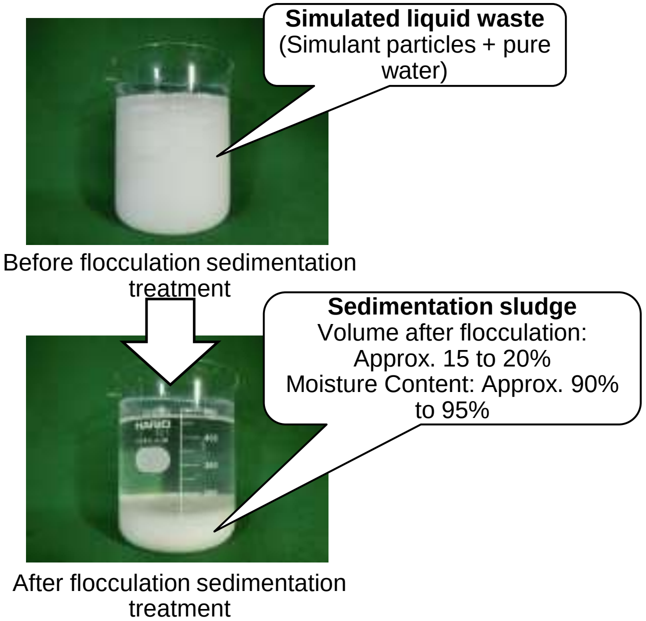
- The amount of sedimentation sludge generated from the sludge collection equipment is high due to the high moisture content, etc., so it is necessary to reduce the volume by dehydration.
- Since dehydration technology applicable to sedimentation sludge has not yet been selected, it is necessary to select a candidate technology.

[Implementation details]

- Assumed properties of sedimentation sludge and supernatant were organized to select an applicable dehydration treatment technology through literature research, etc.
- An element test plan was developed to evaluate the applicability of the selected candidate technology for actual equipment.

[Goals]

- Selection of candidates for the sedimentation sludge dehydration treatment technology.
- Planning of an element test for evaluation of applicability to actual equipment.



Simulated liquid waste
(Simulant particles + pure water)

Before flocculation sedimentation treatment

Sedimentation sludge
Volume after flocculation: Approx. 15 to 20%
Moisture Content: Approx. 90% to 95%

After flocculation sedimentation treatment

Simulant drain water and backwash water flocculation sedimentation test

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

[Implementation details] Dehydration technologies that can be applied to sedimentation sludge were selected through literature research and element tests.
The applicability of the selected dehydration technologies for actual equipment was examined through element tests.

The sedimentation sludge for the dehydration technology to be developed shall be the sedimentation sludge generated from the sedimentation tank for RO-concentrated water treatment, where the amount generated is enormous.

Implementation items
1. Coordination with the canister project (connection conditions, etc.)
2. Consolidation of assumed properties of sedimentation sludge based on the previous project results
3. Evaluation of the amount of hydrogen generated based on sedimentation sludge properties
4. Setting dehydration target (moisture content after dehydration)
5. Investigation of existing dehydration technologies and theoretical study of their applicability
6. Proposal of test plan that can evaluate the applicability to the actual equipment
7. Implementation of applicability evaluation test on the actual equipment

The following items were coordinated among project teams: Project of Development of Technology for Containing, Transfer and Storage of Fuel Debris*1 and project of Development of Safety Systems (liquid and gas systems)*2.
○ Liquid system - Canister storage line interface requirements
○ Sharing of development results
  *Canister PJ: Control policy for collection, transfer, and storage
  Safety system (liquid and gas systems) PJ: Properties of sedimentation sludge at the time of discharge, etc.
○ Classification of the future development contents of both projects

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∗1 Project of Development of Technology for Containing, Transfer and Storage of Fuel Debris: Canister PJ
∗2 Project of Development of Safety Systems (liquid and gas systems: Safety System PJ

7. Implementation details

③ Development of secondary waste treatment technology

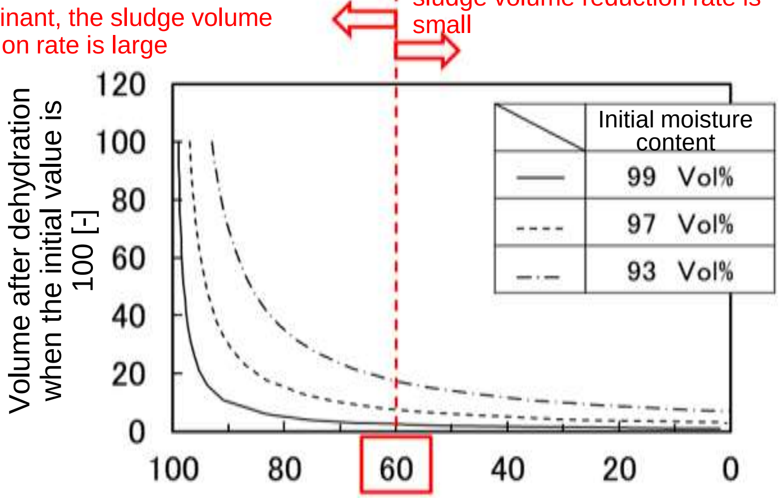
2) Selection of dehydration technology for sedimentation sludge

[Implementation details] The volume reduction of sedimentation sludge due to dehydration was evaluated.

The volume reduction behavior of sedimentation sludge was evaluated when moisture was removed from the assumed moisture content of approximately 90%.

In the region where supernatant water and interstitial water decrease is dominant, the sludge volume reduction rate is large

In the region where the reduction in interstitial water results in gaps, the sludge volume reduction rate is small

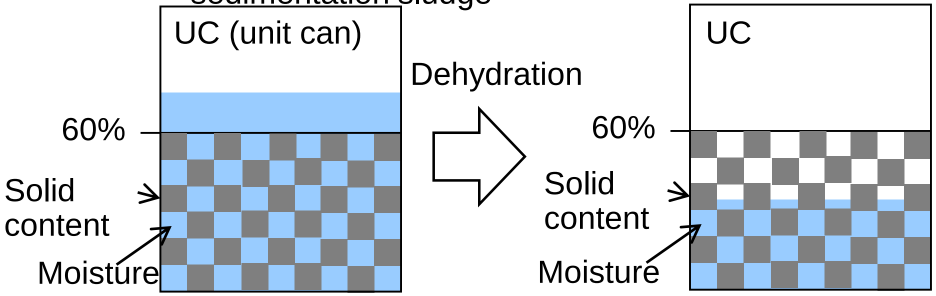


Water content ratio [vol%]

Figure Relationship between moisture content and volume of sludge

Appearance of sedimentation sludge

Water content ratio below 60% is unlikely to contribute much to the sludge volume reduction, as the space occupied by moisture results in gaps



(a) Water content ratio of 60% or more

(b) Water content ratio of 60% or less

Figure Illustration of sludge volume reduction behavior

If the initial moisture content is 90 vol% or more, the volume can be reduced to nearly 20 vol% by dehydration treatment to a moisture content of 60 vol%.
Volume reduction by dehydration has a small effect in the region where the water content is 60 vol% or less, because the volume reduction reaches its limit.

Tentatively set the target value for the water content after dehydration treatment at 60 vol%*, and examine dehydration treatment technology.

*Sludge content rate of 40 vol%

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

[Implementation details] The amount of hydrogen generated from sedimentation sludge during the transferring period was evaluated.

The amount of hydrogen generated was evaluated based on the collection, transfer, and storage conditions set in the canister PJ.
⇒ The results are shown on the next page

■ Collection, transfer, and storage conditions

(1) Fuel debris: 100% UO^2 with maximum burnup in the Fukushima Daiichi NPS
(2) Waste filling rate: 30 vol% of fuel debris
(3) During transferring
  - Transferring period: 1 week
  - Gas vent (opening the coupler of the canister)
    Case 1 Canister: Vent, Transfer cask: Sealed
    Case 2 Canister: Sealed, Transferring cask: Sealed
(4) During storage
  - Gas Vent Canister: Vent

■ Assumed properties of sedimentation sludge

(a) Sedimentation sludge generated from the sedimentation tank for filter liquid waste treatment: fuel debris content 100%
(b) Sedimentation sludge generated from the sedimentation tank for RO-concentrated water treatment: fuel debris content 0.1%*

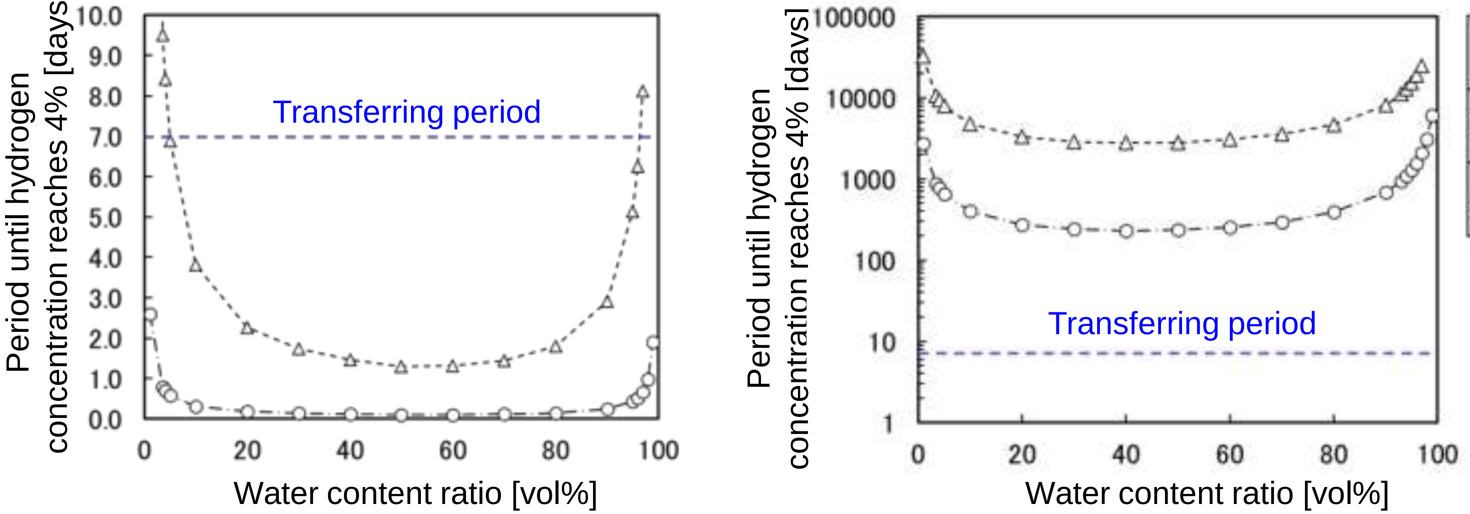
*Rate setting based on the removal rate of nuclides (DF100) in the particle removal equipment and soluble nuclide removal equipment, the assumed concentration rate (2 to 4 times) in the RO filter, etc.

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

[Implementation details] The amount of hydrogen generated from sedimentation sludge during the transferring period was evaluated.



(a) Filter liquid waste sedimentation sludge

(b) RO-concentrated water sedimentation sludge

		Canister	Transfer cask
○	Period required for canister to reach 4% hydrogen concentration	Closed	Closed
△	Period required for transfer cask to reach 4% hydrogen concentration	Opened	Closed

Canister
Gas phase: 0.0133 m^3
Sludge: 0.0115 m^3
Transfer cask
Gas phase: 1.82 m^3
Sludge: 0.16 m^3

Figure. Evaluation of sludge moisture content and amount of hydrogen generated

In both cases, it was verified that the amount of hydrogen generated was the largest when the amount of water and fuel debris content were around 40% to 60%.

If the filter liquid waste sedimentation sludge is dehydrated from the initial moisture content of about 90%, there is a concern that the risk of hydrogen explosion will increase unless the water content is reduced to a few percent or less.

It is unlikely that the RO-concentrated water sedimentation sludge will reach the explosive limit of hydrogen concentration during the transferring period regardless of the water content.

Even if RO-concentrated water sedimentation sludge is stored for a long period, the amount of hydrogen generated would not reach the explosion limit, so the need for dehydration treatment is low from the viewpoint of the amount of hydrogen generated.

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

[Implementation details] Proven technologies were selected as the sludge collection treatment technology for overseas nuclear facilities.

Table Results of sludge dehydration technological study at overseas nuclear power sites

No.	Treatment method	Advantages	Disadvantages	Examples/Ready-made products
1	MF, UF filters (DE) filtration	High DF, low cost.	Requires Backwash and backwash water treatment.	-
2	Cross-flow (CF) filtration	Suitable for radioactive colloid removal.	Clogging due to fouling. Requires concentrated water treatment.	Used in recent PWR liquid waste treatment. Equipment available for treatment at 300 GPM
3	RO concentration	Suitable for concentration of CF filtered water.	Clogging due to fouling.	Used in recent PWR liquid waste treatment.
4	Evaporator	High DF.	Requires scaling treatment, high equipment cost.	-
5	Cartridge filter	Space-saving, low cost.	Low waste collection efficiency.	Solids Collection Filter (SCF) with shielding function for treating large volumes, is used to remove S/P pool sludge, etc.
6	Decompression/thermal dehydration (In-Drum dryer)	Waste can be reduced by 85 to 95%.	Treatment speed depends on drying level.	Mobile equipment

➡ Cartridge filters, which are assumed to be able to dehydrate sedimentation sludge to the target moisture content of 60%, as well as decompression and thermal dehydration will be selected as the dehydration technology for sedimentation sludge.

➡ For other candidate technologies, applicability as concentration treatment technology and secondary nuclide removal technology, which are assumed to have high applicability for each technology, will be examined.

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

[Implementation details] Proven technologies were selected as the sludge collection treatment technology for overseas nuclear facilities.
For sludge collection in accident reactors and weapon legacy systems, the technology used is selected according to the sludge content.

*UK: Atomic Weapons Establishment (AWE), US: TMI, Hanford Site, etc.

Table Investigation of cases of dehydration technologies applied by sludge content

No.	Amount of sludge	Target case	Sludge removal, dehydration, and drying methods
1	Miniscule amount	SNF pool water	Cartridge filter filtration
2	Miniscule amount	Radioactive liquid waste	Cartridge filter filtration + evaporator, RO concentration
3	Small amount (sub-percent)	Sludge during reprocessing, saltcake	Cross-flow filter filtration
4	Medium amount (a few percent)	Long-term corroded SNF pool	Dehydration through filters
5	Large amount (several tens of percent)	Research reactor waste Concentrates No. 2 to 4	(Solidified as is)

When the sludge content is small, there are many cases where filtration with a cartridge filter is adopted. If the sludge content is slightly low, cross-flow filtration is used to concentrate it to a few percent. However, raising the concentration to several tens of percent is difficult.
If the sludge content is high, instead of dehydration treatment, solidification treatment such as cement solidification or vitrification is adopted depending on the activity level and final disposal method. If the treatment/disposal method is undecided, it is stored for a long period of time, making use of facilities such as underwater storage.
In many cases of overseas sites, the sludge content is increased by cross-flow concentration, drying treatment, etc., and then solidified.

7. Implementation details

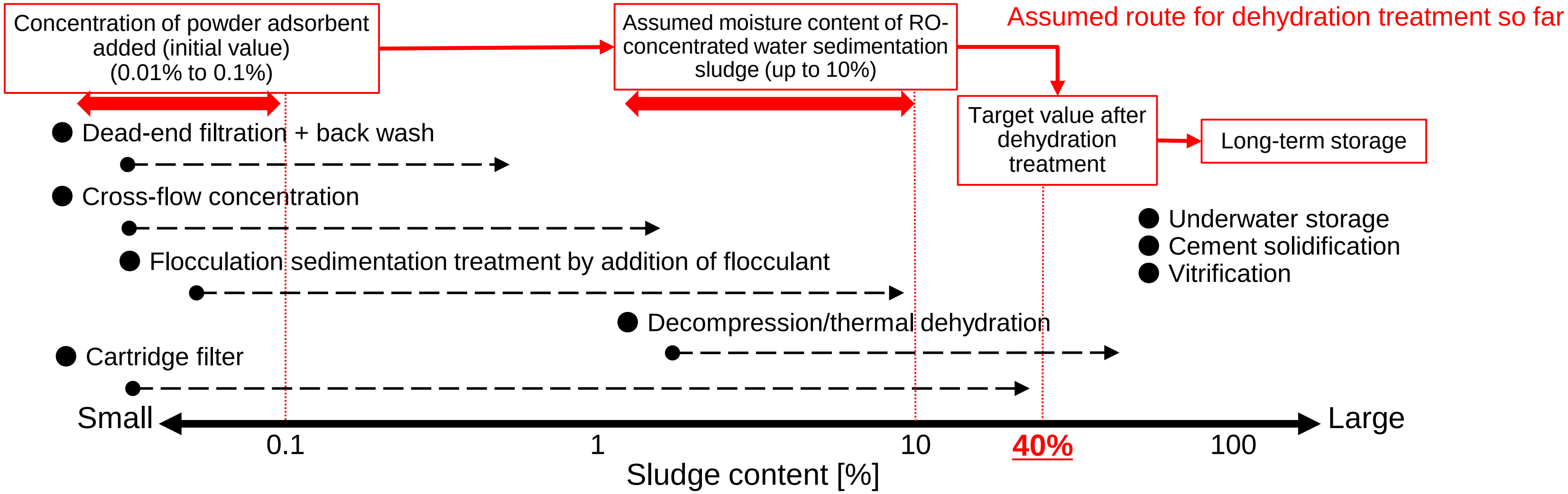
③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

➢ Examination of RO-concentrated water treatment system

The case results show that in many cases, appropriate dehydration methods have been adopted for each sludge content region, and the sludge content has been concentrated to the target sludge content.

Selection of RO-concentrated water sedimentation sludge dehydration technology



The following two proposals were examined as treatment processes for RO-concentrated water.

Plan 1:	① Flocculation sedimentation treatment	→	② Cartridge filter filtration	→	③ Long-term storage
Plan 2:	① Flocculation sedimentation treatment	→	② Decompression/thermal dehydration	→	③ Long-term storage

* For plan 2, since it is difficult to treat large volumes with decompression/thermal dehydration equipment, a system that reduces the amount of liquid by concentration treatment (cross-flow, etc.) as a pretreatment was also examined.

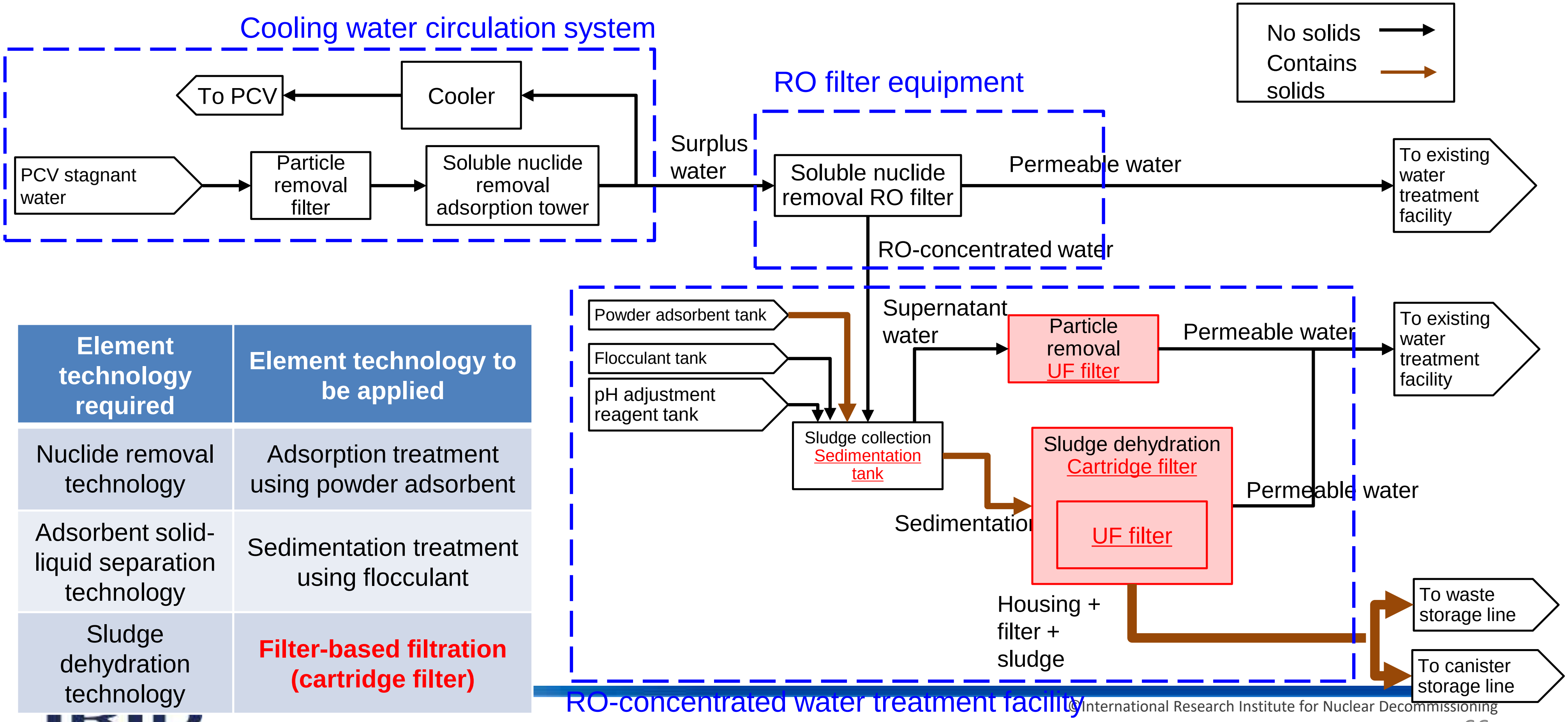
7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

- **Examination of RO-concentrated water treatment system**

Plan 1: ① Flocculation sedimentation treatment → ② Cartridge filter filtration → ③ Long-term storage



Element technology required	Element technology to be applied
Nuclide removal technology	Adsorption treatment using powder adsorbent
Adsorbent solid-liquid separation technology	Sedimentation treatment using flocculant
Sludge dehydration technology	**Filter-based filtration (cartridge filter)**

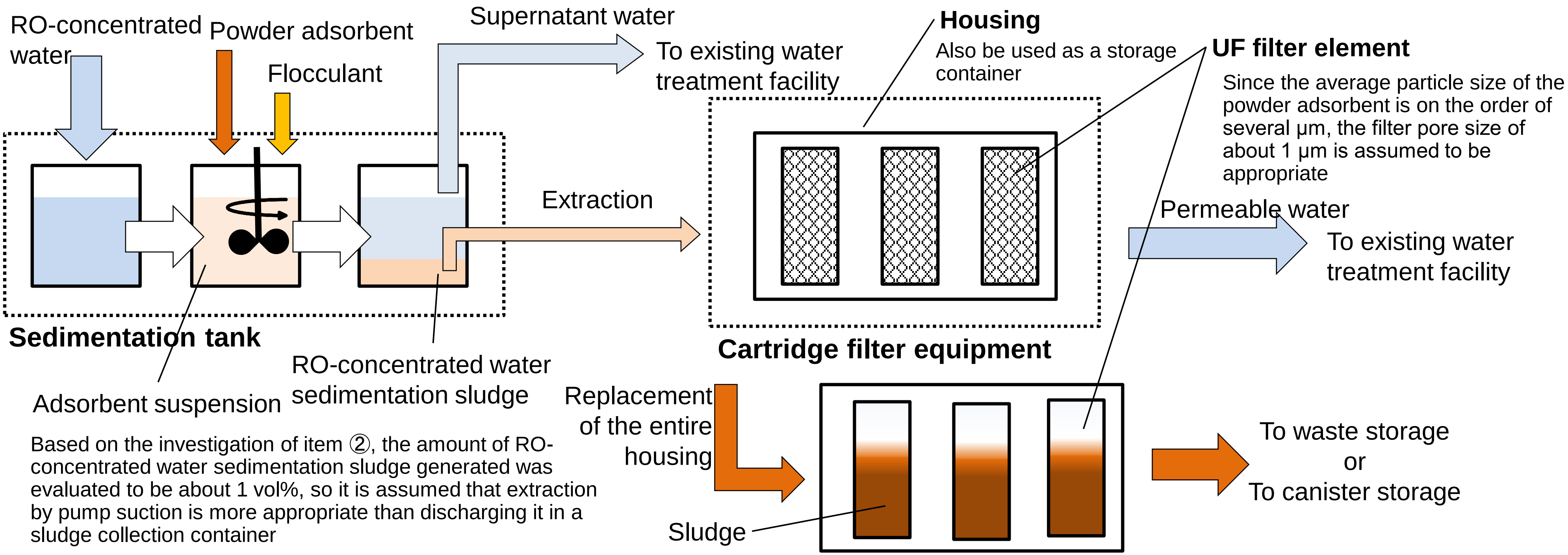
7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

➢ Examination of RO-concentrated water treatment system

Plan 1: ① Flocculation sedimentation treatment → ② Cartridge filter filtration → ③ Long-term storage



Since the average particle size of the powder adsorbent is on the order of several μm, the filter pore size of about 1 μm is assumed to be appropriate

Based on the investigation of item ②, the amount of RO-concentrated water sedimentation sludge generated was evaluated to be about 1 vol%, so it is assumed that extraction by pump suction is more appropriate than discharging it in a sludge collection container

*Verified in the FY2020 test that it has fluidity due to its high moisture content of about 90% and can be extracted with a pump

The structure of cartridge filters can be designed to be replaced as a module.
By handling the housing as a storage container, long-term storage is possible as it is.
Since it is not necessary to retrieve the element, worker exposure can be reduced if the housing has a shielding function.
It is also simpler to operate than element replacement and can therefore be expected to be operated remotely.

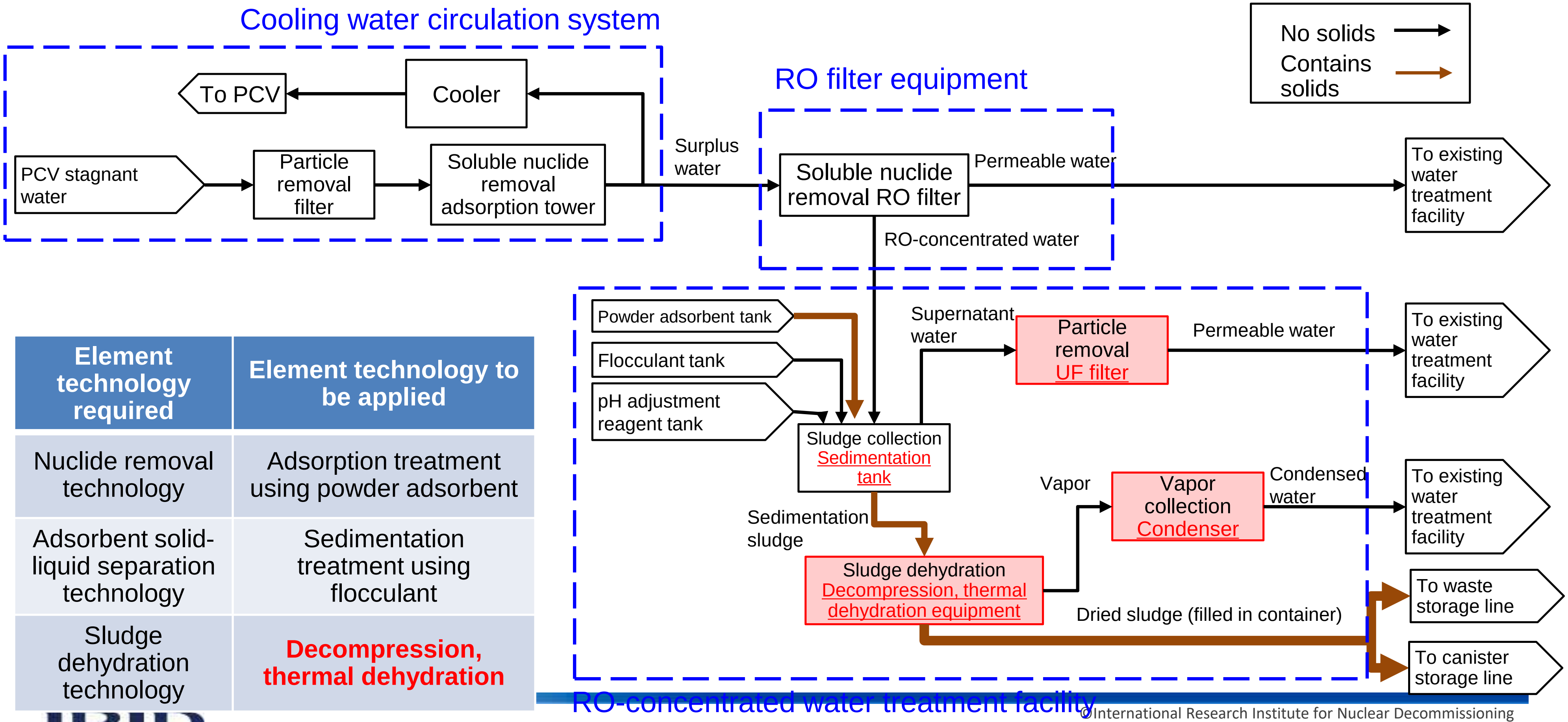
7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

- **Examination of RO-concentrated water treatment system**

Plan 2: ① Flocculation sedimentation treatment → ② Decompression/thermal dehydration → ③ Long-term storage



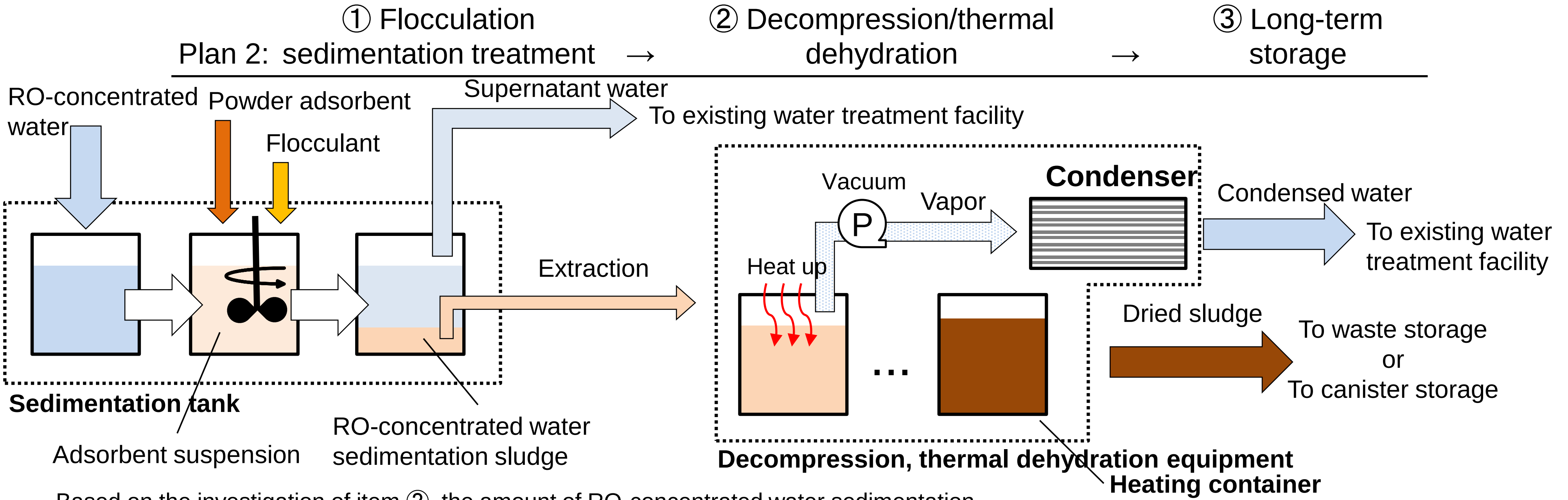
Element technology required	Element technology to be applied
Nuclide removal technology	Adsorption treatment using powder adsorbent
Adsorbent solid-liquid separation technology	Sedimentation treatment using flocculant
Sludge dehydration technology	**Decompression, thermal dehydration**

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

- **Examination of RO-concentrated water treatment system**



Based on the investigation of item ②, the amount of RO-concentrated water sedimentation sludge generated was evaluated to be about 1 vol%, so it is assumed that extraction by pump suction is more appropriate than discharging it in a sludge collection container

*Verified in the FY2020 test that it has fluidity due to its high moisture content of about 90% and can be extracted with a pump

RO-concentrated water sedimentation sludge is transferred to the container for dehydration treatment, and the water is evaporated by decompression and heating.
The generated vapor is collected and condensed in the condenser and discharged to the existing water treatment facility.
Although dried sludge remains on the container side, it is assumed that the dead space inside the container is large due to the volume reduction.
Therefore, the cycle of receiving sedimentation sludge→ dehydration treatment→ residual dried sludge is repeated to increase the filling rate of dried sludge in the container. After it is filled to a certain level, the entire container is discharged and stored for a long period of time.
By giving the container a shielding function, worker exposure can be reduced.

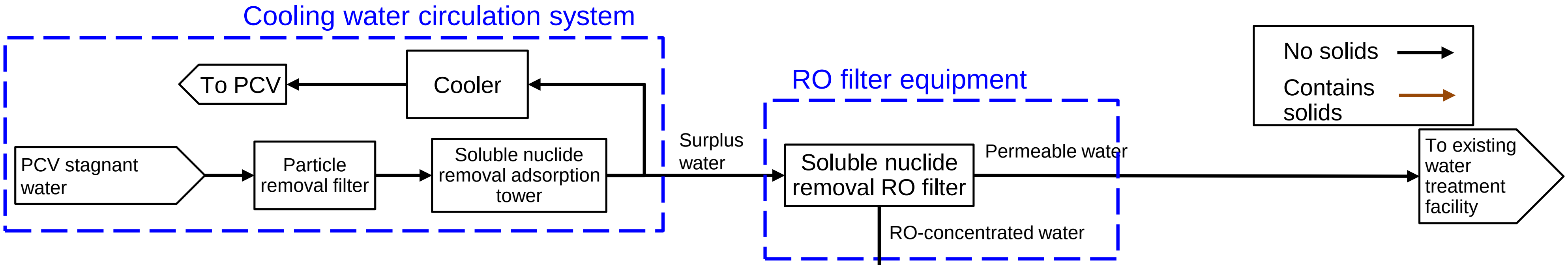
7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

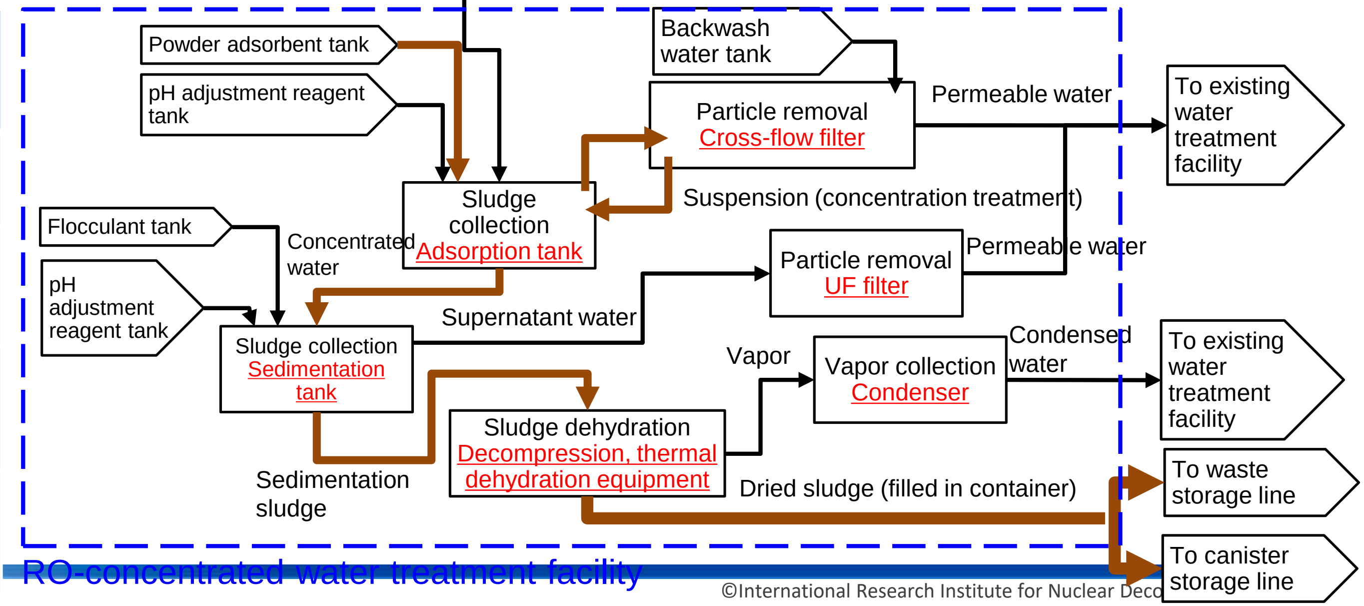
- **Examination of RO-concentrated water treatment system (when adopting cross-flow concentration)**

① Cross-flow concentration → ② Flocculation sedimentation treatment → ③ Decompression, thermal dehydration → ④ Long-term storage



Cooling water circulation system
To PCV
Cooler
PCV stagnant water
Particle removal filter
Soluble nuclide removal adsorption tower
Surplus water
RO filter equipment
Soluble nuclide removal RO filter
Permeable water
RO-concentrated water
To existing water treatment facility
No solids →
Contains solids →

Element technology required	Element technology to be applied
Nuclide removal technology	Adsorption treatment using powder adsorbent
Solid content concentration technology	**Cross-flow filtration**
Adsorbent solid-liquid separation technology	Sedimentation treatment using flocculant
Sludge dehydration technology	Decompression, thermal dehydration



Powder adsorbent tank
pH adjustment reagent tank
Backwash water tank
Particle removal Cross-flow filter
Permeable water
To existing water treatment facility
Suspension (concentration treatment)
Sludge collection Adsorption tank
Flocculant tank
pH adjustment reagent tank
Concentrated water
Particle removal UF filter
Permeable water
Supernatant water
Sludge collection Sedimentation tank
Vapor
Vapor collection Condenser
Condensed water
To existing water treatment facility
Sludge dehydration Decompression, thermal dehydration equipment
Sedimentation sludge
Dried sludge (filled in container)
To waste storage line
To canister storage line
RO-concentrated water treatment facility

7. Implementation details

③ Development of secondary waste treatment technology

2) Selection of dehydration technology for sedimentation sludge

- **Examination on element test of sludge dehydration technology**

  The applicability of candidate technologies that were selected by the literature research was verified through element test.

Sludge dehydration technology	Concept	Items to be verified in element test	Possibility of dehydration up to the target value of 60%	Element test Necessity
Cartridge filter	Depending on the sludge properties (particle size distribution, moisture content, etc.), it is necessary to select an appropriate filter element (pore size, filtration area), filtration conditions, etc.	- Selection of filtration method and conditions - Dependence of dehydration performance on sedimentation sludge properties	Possible	Required
Decompression, thermal dehydration	It is assumed that the dehydration treatment can be performed up to the target value without greatly depending on the sludge properties (particle size distribution, particle properties)	—	Possible	Not required
(Solidification treatment)	Solidification treatment is considered to be effective from the viewpoint of stabilization of sludge type waste, but the concept of final disposal of waste in the Fukushima Daiichi NPS is currently under examination. Since solidification treatment should be performed according to the burial requirements, it is not examined in this development, and long-term storage after dehydration treatment is estimated.	—	—	—

Element tests for cartridge filters were conducted to obtain operational data.

*System using decompression and thermal dehydration will also be examined with reference to literature data, etc.

- **Small-scale test (assuming 0.1 to 1L scale)**

  A filtration method which the sedimentation sludge moisture content after dehydration treatment satisfies the target value of 60 vol% and the amount generated is at a level that can be discharged to a later stage equipment was selected.

  Tests for Vacuum filtration and pressure filtration using RO-concentrated water sedimentation sludge simulants were conducted. Information on average cake resistance, wet-dry mass ratio, etc. to evaluate whether the requirements for throughput and dehydration effect can be satisfied. (Test content under consideration)

7. Implementation details

③ Development of secondary waste treatment technology

- Summary

1) Characterization of liquid waste generated from the particle removal system

[Results for this year]

- ✓ A 1.4 μm MF filter was adopted as the intermediate filter. Non-spherical particles were selected to evaluate particle shape dependence of filter apparatus for each system. A test method of filter test was devised to evaluate the performance of each filter.

[Future plans]

- ✓ A filter water flow test (both scheduled for FY2022) will be conducted to evaluate the applicability of the 1.4 μm MF filter (intermediate component) and the particle shape dependence of each filter apparatus.

2) Selection of dehydration technology for sedimentation sludge

[Results for this year]

- ✓ The required dehydration performance was examined from the viewpoint of the amount of hydrogen generated and volume reduction, and the sludge dehydration target was tentatively set at a moisture content of 60 vol%.
- ✓ Based on literature research, cartridge filters and decompression and thermal dehydration were selected as dehydration treatment methods applicable to sedimentation sludge.

[Future plans]

- ✓ Element tests will be planned and conducted to examine the applicability of cartridge filters.
- ✓ The sludge dehydration treatment system that was selected candidate technologies (cartridge filters, decompression, thermal dehydration) through literature research will be studied.

8. Specific objectives for achieving the purposes of technological developments

(1) Liquid/gas systems	
① Development of soluble α-nuclide removal technology - Examination of soluble α-nuclide removal test assuming use of actual liquid	In order to verify the effectiveness and feasibility of the soluble α- nuclide removal equipment, the test method, test equipment and test plan that assume the use of actual liquid shall be specified. (TRL at the end of the project: Level 3)
- Element test assuming fuel debris retrieval work	Regarding the soluble α-nuclide removal equipment, the α-nuclide adsorption performance data in the environment assumed during fuel debris retrieval work shall be obtained, and candidate adsorbents shall be selected. At that time, a water quality adjustment policy shall be established. (TRL at the end of the project: Level 4)
② Development of RO-concentrated water treatment technology - Selection of adsorbent and condensing agent	Regarding the RO-concentrated water treatment facility, the nuclide removal performance of the powder adsorbent and the flocculation sedimentation performance of the flocculant shall be evaluated and the powder adsorbent and flocculant shall be selected. (TRL at the end of the project: Level 4)
- Examination of applicability to actual equipment	Regarding the RO-concentrated water treatment facility, the conceptual design of the treatment method and equipment shall be implemented from the test using the sedimentation tank. (TRL at the end of the project: Level 4)
③ Development of secondary waste treatment technology - Investigation of pretreatment technologies	A candidate stabilization treatment technology shall be selected for the sludge generated from the flocculation sedimentation tank. At that time, pretreatment technology for liquid waste such as supernatant water shall also be selected. (TRL at the end of the project: Level 3)
- Examination of applicability to actual equipment	The applicability of the pretreatment technology to the actual equipment shall be verified based on the results of element tests. In addition, the conceptual design of the pretreatment equipment shall be established. (TRL at the end of the project: Level 4)

*TRL: Technology readiness levels

End