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Research and development activities for cleanup of the Fukushima Daiichi Nuclear Power Station

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ABSTRACT

The Fukushima Daiichi nuclear power station accident and restoration works have produced significant volume of radioactive waste. The waste has very different characteristics from usual radioactive waste produced in nuclear power stations and it requires extensive research and development for management of the waste. R&D works such as analysis of the waste properties, hydrogen generation by radiolysis and diffusion in a storage vessel and corrosion of storage vessels, etc. have been performed for characterization and safe storage of the waste. The detailed R&D plan for processing and disposal waste will be established by the end of FY2012.

INTRODUCTION

Fukushima Daiichi nuclear power station (NPS) consists of six BWRs. Units 1, 2, and 3 were operating at rated power level while 4, 5, and 6 were in cold shutdown when a magnitude 9.0 earthquake occurred offshore of Japan's east coast in the afternoon on March 11, 2011. The operating units auto-scrammed on reactor protection system trip. The earthquake immediately devastated all off-site AC power supply to the station. On-site emergency diesel generators were automatically kicked in to supply power for emergency systems. However, a series of tsunamis with their greatest height of approximately fifteen meter began hitting the site about forty minutes later the earthquake and killed most of on-site emergency AC/DC power supply by flooding diesel generators, switchgear rooms, DC batteries and so forth. Units 1-3 lost core cooling to remove decay heat that lead to fuel degradation and core melt. Hydrogen produced in damaged cores caused explosions blowing off top of reactor buildings in units 1, 3, and 4.

The accident produced radioactive gaseous, liquid, and solid wastes. TEPCO estimated approximately 500PBq of radioactive iodine and cesium was released into the air and the sea after the accident [1]. Significant gaseous releases have stopped since Reactor Pressure Vessel (RPV) temperatures decreased well below 100 °C till the end of 2011 using the temporarily installed core cooling system supplying cooling water to the damaged cores [2].

Approximately 400 tons of cooling water is injected to damaged cores everyday even more than five hundred days after the accident via piping of primary water lines to remove decay heats [3]. Injected cooling water is not lead to any outlet lines after heat removal but spills on Primary Containment Vessel (PCV) floors through accident-made pressure boundary openings, flows through basement floors of reactor/turbine/utility buildings and is pumped out and recycled after decontamination.

Solid wastes consist of secondary wastes from water decontamination treatments such as sludge and filter zeolite, rubble from building explosions, dismantled concrete/metal debris from cleanup work, organic debris from deforestation to make space for waste storages and other cleanup facilities, and radioactive nuclear fuel/debris in damaged units. Total volume of the accumulated rubble and debris have already reached 122,000 m³ [5] and are expected to increase as cleanup work proceeds. Although Fukushima Daiichi station site is one of the largest nuclear power station sites in Japan, it should not be regarded as inexhaustible as the areas set for settlement of nuclear accidents in other countries [6].

Table 1 shows types, dose rates, and volumes of already retrieved accident wastes.

Table 1. Retrieved accident waste

Generation Process	Waste type	Dose rate [mSv/h] ^[7]	Quantity ^[5,8]
Cooling water decontamination	Sludge	Approx. 10 ³	597 m ³
	Zeolite	Approx. 10 ¹ at vessel surface	458 vessel
	Concentrated liquid waste	Approx. 10 ² beta ray	Approx. 195,000 m ³
As found / Dismantlement	Concrete / metal debris	Approx. from 10 ⁻³ to 10 ³	Approx. 54,000 m ³
Deforestation	Felled tree, Soil	Approx. from 10 ⁻³ to 10 ⁻¹	Approx. 68,000 m ³

This paper concentrates hereafter on concerns for management of secondary wastes, organic debris and rubble stored at and researches for characterization and safe storage, processing, and disposal of the waste.

ON-SITE WASTES AND MAJOR CONCERNS

Organic and inorganic debris and rubble from deforestation, explosion and dismantling are currently accumulated on-site as shown in Table 2. Rubble and other dismantled debris are sorted by dose rate and stored separately. Figure 1 and 2 show temporary storage design for rubble and organic debris. Both designs have radiation shielding reducing dose from waste storage areas to 1mSv/y at the site periphery. Samples are taken periodically from various points of storage area and dismantled locations for future analysis.

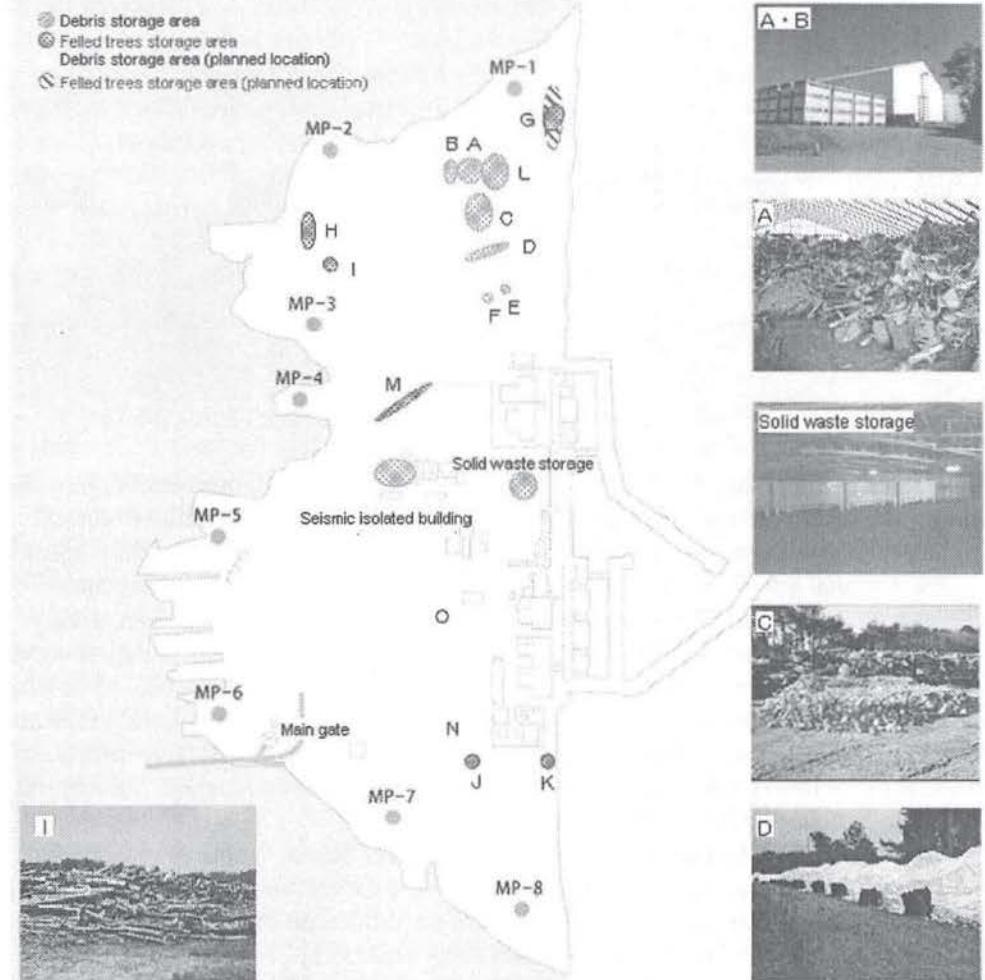
Table 2. Amount of debris and felled trees being temporarily stored (as of September 28, 2012)

Amount of debris and felled trees being temporarily stored (as of September 28, 2012)

Storage location	Air dose rate at area border (mSv/h)	Type	Storage method	Storage quantity ^a	Area occupation rate
Solid waste storage	0.05	Concrete, metal	Container	2,000 m ³	35 %
A : north side of site	0.35	Concrete, metal	Temporary storage facility	11,000 m ³	98 %
B : north side of site	0.04	Concrete, metal	Container	4,000 m ³	98 %
C : north side of site	0.01	Concrete, metal	Stored outside	28,000 m ³	83 %
D : north side of site	0.01	Concrete, metal	Stored outside	2,000 m ³	86 %
E : north side of site	0.01	Concrete, metal	Stored outside	3,000 m ³	90 %
F : north side of site	0.01	Concrete, metal	Container	1,000 m ³	99 %
L : north side of site	under 0.01	Concrete, metal	Soil-covered temporary storage facility	2,000 m ³	26 %
Total (Concrete, metal)				64,000 m ³	76 %
G : north side of site	0.01	Felled tree	Stored outside	18,000 m ³	83 %
H : north side of site	0.01	Felled tree	Stored outside	16,000 m ³	93 %
I : north side of site	0.03	Felled tree	Stored outside	11,000 m ³	100 %
J : south side of site	0.05	Felled tree	Stored outside	12,000 m ³	77 %
K : south side of site	0.04	Felled tree	Stored outside	5,000 m ³	100 %
M : west side of site	0.01	Felled tree	Stored outside	6,000 m ³	29 %
Total (Felled tree)				68,000 m ³	74 %

*1: Totals may not add up since numbers have been rounded for under 100 m³ of volume. The total amount of felled trees is less than 1,000 m³ compared with last time so a value of "0" has been entered.

- Debris storage area
- Felled trees storage area
- Debris storage area (planned location)
- Felled trees storage area (planned location)



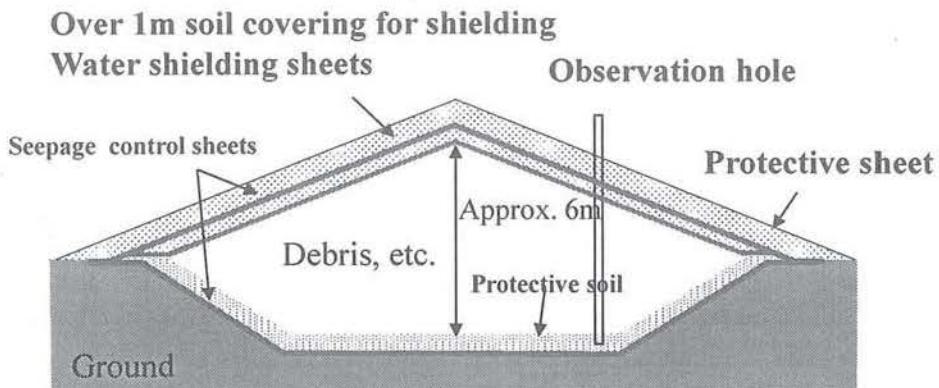


Figure 1. Overview of the temporary storage facility for rubble

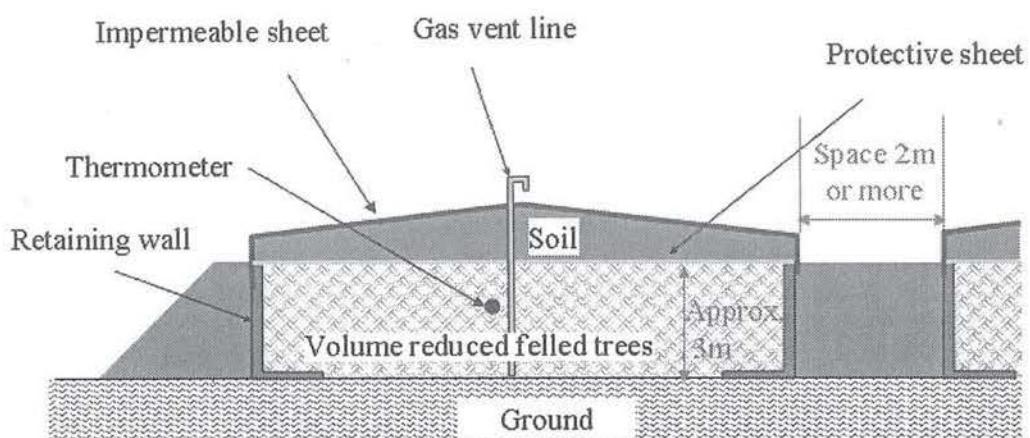


Figure 2. Overview of the temporary storage facility for organic debris (felled trees)

Dose rates at the peripheries of the station site, $\sim 10^2$ mSv/y [9], originate largely in cesium fallout. The summation of contributions of on-site temporally accumulated waste storages to the periphery doses are assessed as up to 10 mSv/y [10], that is physically minor but regarded by the national government cleanup policy as a kind of a performance indicator showing cleanup work progress. The policy requires that the radiations from the temporary storages to the peripheries should be evaluated below 1mSv/y, restricting placement of waste storages near the peripheries in an indirect manner. The restriction causes shortage of on-site waste storage area and/or imposes storages equipped with shielding. This symbolic performance indicator forces time-and-resource-consuming cleanup works and spoils effective utilization of limited on-site area. In this context, this symbolic indicator might not encourage but dispute cleanup works, and evoke anxiety of local governments.

On-site worker dose might become a serious issue for human resource management. Although dose levels of emergency workers are not the case for current cleanup workers, legal limits (50mSv/y and 100mSv/5y) have become critical conditions on the resource management for cleanup works expected to continue for the next forty years [11].

One of the major liquid waste issues is that groundwater sinks into the basement floors and increases the amount of accumulated water. Any agreements for ocean release of surplus

water, which is slightly contaminated but could be treated further to below release limit, have not been made due to current environmental policy issues. The surplus is about 500 tons per day and temporally stored on-site [4]. Storage tanks have been multiplying and reducing open on-site areas.

In addition to a series of serious boundary breaks from fuel degradation to building explosion, accident management activities such as seawater/boron injection and subsequent cooling water circulation operation might have influences on volumes, physical/chemical form and mobility of radionuclide by contacting and reacting with a variety of on-site materials. The contamination processes, which determine volumes, characteristics and radioactive inventories of accident wastes, are far from the usual plant contamination process originated in neutron activation of reactor materials. We believe that a brand-new waste management strategy should be considered to handle accumulation, identification, analysis, storage, processing and final disposal for the Fukushima Daiichi case.

Sludge, zeolite and concentrated effluent are produced through cooling water purification treatments. Sampling of these wastes from spent adsorbent vessels or a temporary storage is a never easy task due to their high dose rate and/or hard-to-hollow welded vessels designed, manufactured, and installed by a rush work after the accident.

Hydrogen produced inside spent adsorbent vessels due to radiolysis is a safety issue for long-term storage of the secondary wastes. Chlorides contained in these wastes as a result of tsunami and seawater injection are also a point of safety concern for future geologic disposal.

New decontamination system is under preparation to reduce concentrations of a variety of radionuclides in treated water furthermore for public acceptance of ocean release [12]. The system employs a variety of new and best suited adsorbents and co-precipitation agents. Waste management strategy should also cover these newly born secondary wastes from the system.

WASTE MANAGEMENT STRATEGY AND CLEANUP ROADMAP

Fukushima Daiichi cleanup roadmap is shown in Figure 3. All activities are still in a primary stage of the roadmap that should be further modified and substantiated to clarify the endpoint, boundary conditions and a right way for the entire decommissioning of the station site.

Proper management of wastes from cleanup work is an essential matter for comprehensive decommissioning of the station site. It is, however, facing the above-mentioned social/political issues and highly complicated technical difficulties. United workforce of members from the government, TEPCO, JAEA and other Japanese top authorities are now seriously planning, preparing and performing national level activities for cleanup.

Waste management strategy should contain plans and schedule of waste management, such as characterization, storage, processing, and disposal, for cleanup of the site. The plans approaching toward a suitable endpoint shared among all cleanup authorities and stakeholders should be accomplished. The strategy should also make clear boundary conditions, ensure accessibility to various waste management knowledge, and define resources necessary for acquiring new data and technologies if needed. Waste data and knowledge necessary for reaching the endpoint should be systematized and databased. Radionuclide inventories and other factors with significant impacts on safety of geologic disposal should be evaluated and optimized through system design for waste processing and final disposal using the database. Plans with priorities and schedules for these cleanup steps applied for various areas and materials should be implemented and reviewed periodically till the endpoint is reached.

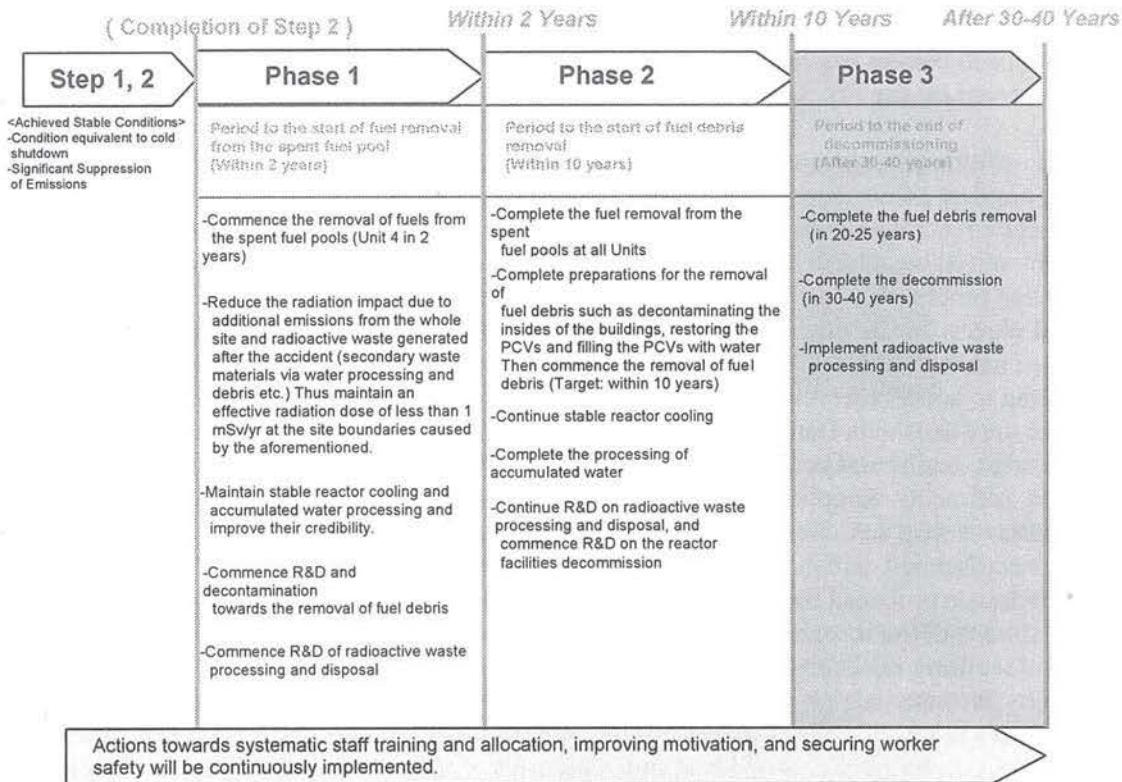


Figure 3. Mid-and-long Term Roadmap Summary

R&D for Resolving Technical Issues

Scope

Japan Atomic Energy Agency (JAEA) has extensively conducted R&D on management of the accident waste based on the Fukushima Daiichi research and development roadmap [9]. The scope of the R&D includes the following major areas.

- Characterization of the waste
- Investigation for safe storage of the waste
- Investigation for processing and disposal of the waste

(1) Characterization of the waste

Although characterization of the waste is essential to plan processing and disposal of the waste, analysis for the characterization is very time-consuming process because of enormous volume and complex composition of the waste caused by the core melt and the subsequent hydrogen explosions. JAEA has analyzed properties of relatively low activity wastes such as contaminated water after treatment, building debris, and felled tree stored at the Fukushima Daiichi NPS since January 2012. Among radioactive nuclides, long-lived alpha and beta nuclides

are important for safety assessment of waste disposal. Chemicals in the waste which influence its processing and disposal will be analyzed in FY2013. JAEA is also developing analytical techniques and methods to save time and to improve detection limits.

(2) Investigation for safe storage of the waste

The major wastes stored at the Fukushima Daiichi NPS are, building debris, felled trees, contaminated water, and secondary wastes generated by treatment of the contaminated water. The first three wastes are technically easy to store except a problem of their enormous volumes. The secondary wastes such as spent zeolites and coagulation-sedimentation sludge have some issues for storage. Radioactive cesium concentrated in wastes and saline remained in wastes might cause radiolysis hydrogen fire and corrosion of storage vessels.

JAEA has investigated hydrogen gas generation/diffusion and corrosion of vessels for safe storage of the secondary waste of contaminated water treatment. Figure 4 shows the approach for safe storage of the wastes. The major subjects of the study are characterization of the wastes, hydrogen gas diffusion simulation in storage vessel, corrosion test of storage vessels, and chemical stability test of the waste. JAEA will prepare the measures for safe storage of the spent zeolites and the coagulation-sedimentation sludge in FY 2013.

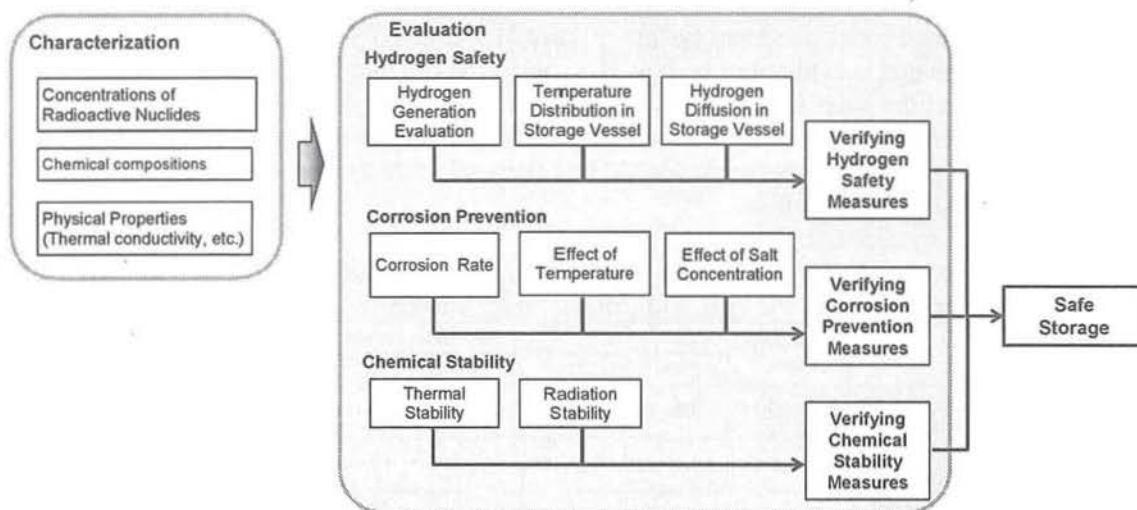


Figure 4. Procedures for safe storage of contaminated water treatment wastes

(3) Investigation for processing and disposal of the waste

There are many issues for processing and disposal of the accident waste because of huge volume, complex radiological and chemical composition, etc. The detailed R&D plan for processing and disposal of the waste is being prepared and will be established by the end of FY2012. The R&D should be conducted by the following steps. First, processing and disposal of the waste using existing technologies will be evaluated. If there are issues for waste processing or disposal using conventional technologies, then new technologies should be investigated to resolve the issues and finally safety of processing and disposal should be verified.

Progress Status

JAEA has conducted R&D on the accident waste management for a year and a half. The investigation is only the beginning of the long way for cleanup of the Fukushima Daiichi NPS and extensive R&D should be needed for the future. This section describes the results of JAEA's study and R&D subjects required for proper management of the accident waste.

(1) Characterization of the waste

Radioactive nuclides contained in the stored waste have been analyzed since January 2012. About 30 nuclides which are important for assessment of waste processing and disposal were selected as objective. At first, nine contaminated water samples from the contaminated water treatment system was analyzed in order to evaluate inventory of the secondary wastes generated from the contaminated water treatment, because it is difficult to sample the secondary wastes due to their extremely high activity. Then, three new contaminated water samples and 12 building debris and five felled trees are being analyzed. The analysis will be conducted to understand overview of distribution of radioactive nuclides in the waste for the coming a few years and then will be conducted to evaluate inventory of the waste.

Table 3 shows concentrations of radioactive nuclides in the contaminated water samples from the contaminated water treatment system (Figure 5). Cesium-137 and Sr-90 were major radioactive nuclides and beta nuclides such as H-3, Ni-63, Se-79, and I-129 were detected. Although alpha nuclides such as U, Np, Pu, Am, Cm were under the detection limit, the detection limits were relatively high because of small sample volume on each radiochemical analysis (100 mm^3). The detection limits should be improved in the present analysis by increasing the volume of the sample.

Table 3. Activity concentrations of gamma and beta nuclides in contaminated water samples
(Alpha nuclides such as U, Np, Pu, Am, and Cm are under the detection limits.)

No.	Sample	Sampling Date	Activity Concentration (Bq/cm^3)						
			^{60}Co	^{137}Cs	^3H	^{63}Ni	^{75}Se	^{90}Sr	^{129}I
1	Concentrated RW basement highly contaminated water	Nov/1/2011	4.9×10^0	7.4×10^5	3.3×10^3	6.3×10^{-1}	8.3×10^0	2.9×10^5	2.5×10^{-1}
2	Water after cesium adsorption device processing (Tandem)	Aug/9/2011	1.7×10^1	1.1×10^4	6.0×10^3	1.5×10^0	2.7×10^0	1.2×10^5	8.3×10^{-2}
3	Water after cesium adsorption device processing (Single)	Nov/8/2011	7.4×10^0	7.7×10^0	4.0×10^3	7.4×10^{-1}	2.5×10^0	2.0×10^5	2.7×10^{-1}
4	Water after decontamination device processing	Aug/9/2011	9.9×10^0	5.3×10^{-1}	6.3×10^3	4.4×10^{-1}	3.1×10^0	1.2×10^4	8.5×10^{-2}
5	Water after 2 nd cesium adsorption device processing	Nov/8/2011	4.6×10^{-1}	$<2.7 \times 10^{-1}$	3.3×10^3	$<3.8 \times 10^{-1}$	1.6×10^1	1.0×10^5	1.3×10^{-1}
6	Desalination device outlet water	Nov/1/2011	$<6.0 \times 10^{-2}$	$<1.3 \times 10^{-1}$	3.9×10^3	$<3.1 \times 10^{-1}$	8.1×10^1	4.0×10^1	$<2.1 \times 10^{-2}$
7	Evaporative concentration device inlet water	Nov/1/2011	1.4×10^1	6.6×10^0	6.1×10^3	1.1×10^0	3.0×10^0	2.3×10^4	1.8×10^{-1}
8	Evaporative concentration device outlet water	Nov/1/2011	$<6.1 \times 10^{-2}$	$<1.3 \times 10^{-1}$	5.4×10^3	$<3.2 \times 10^{-1}$	7.8×10^1	3.5×10^1	$<2.1 \times 10^{-2}$
9	Evaporative concentration device concentrated waste water	Nov/3/2011	2.7×10^0	5.3×10^1	6.2×10^3	$<3.1 \times 10^{-1}$	9.4×10^1	3.2×10^3	1.3×10^0

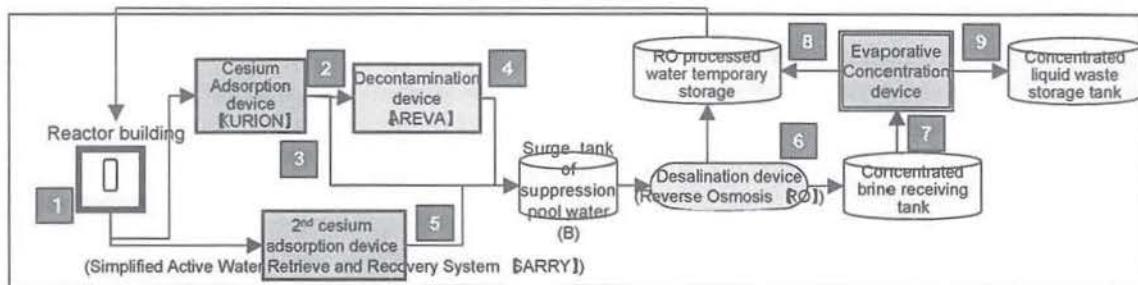


Figure 5. Contaminated water sampling points

In parallel with the radioactive nuclide analysis, new analysis techniques have been developed to deal with high-activity waste samples rapidly and remotely. Figure 6 shows the schematic diagram of laser ablation assisted resonance-enhanced multi photon ionization mass spectrometry. This technique has been developed for analysis of long-lived beta nuclides such as Mo-93. Sample is atomized by laser ablation and aimed nuclide is selectively ionized by resonance-enhanced multi photon ionization and analyzed using the subsequent time of flight mass spectrometer. Concerning alpha nuclides, capillary electrophoresis has been applied for separation of actinides. This technique is suitable for operation in glove box and cell because it can automatically separate actinides and reduce secondary waste significantly.

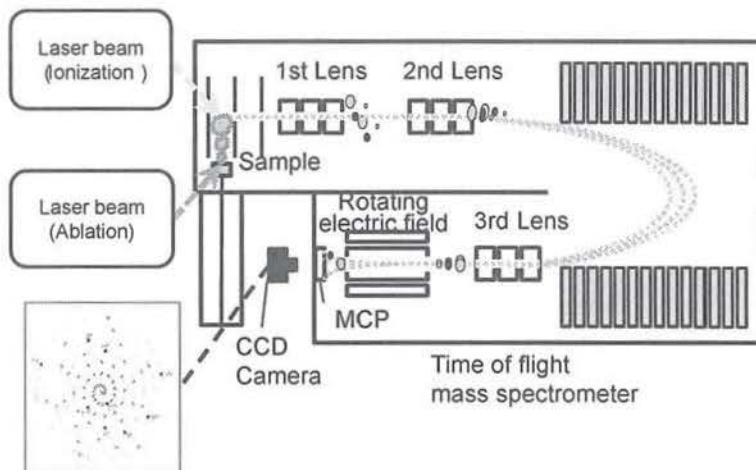


Figure 6. Schematic diagram of laser ablation assisted resonance-enhanced multi photon ionization mass spectrometry

(2) Safe storage of the waste

The secondary waste from the contaminated water treatment system has issues such as containing saline and high concentrations of radioactive nuclides. JAEA has studied hydrogen generation and diffusion in a spent zeolite vessel, corrosion behavior of storage vessels, etc. for safe storage of the waste.

Figure 7 shows analytical results on hydrogen diffusion in the spent zeolite vessel [13]. Analytical conditions were uniform hydrogen production rate of 18.3L/day, 237W of decay heat,

and no steam generation. Analytical results show that air is slowly introduced into the vessel bottom through the outlet water pipe. The mixed gas of air and hydrogen is released to outside through the inlet water pipe and the vent tube. This thermal-hydraulic behavior keeps the hydrogen concentration under 4% of the hydrogen explosive limit.

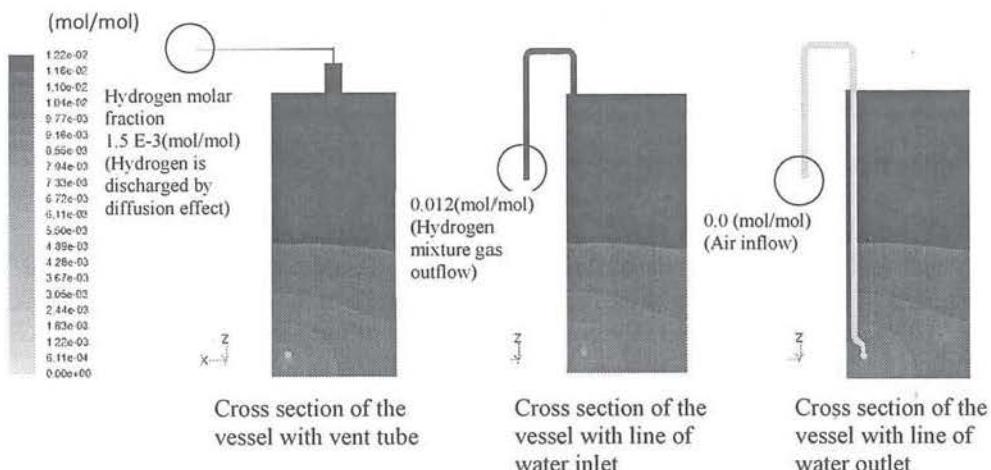


Figure 7. Analytical results on hydrogen diffusion in a spent zeolite vessel

Salt and radiolysis products such as hydrogen peroxide might accelerate corrosion of storage vessels. The effect of salt concentration on corrosion has been investigated by corrosion test with test pieces and computer simulation. No corrosion was observed in a 500-hour corrosion test using test pieces and artificial seawater (2,000 ppm of Cl^-). Corrosion test for evaluating effect of gamma ray radiation is being prepared. The basic results on storage vessel corrosion will be summarized by the end of FY 2013.

(3) Investigation for processing and disposal of the waste

The detailed R&D plan for processing and disposal of the waste is being discussed and it will be established by the end of FY2012 (March 2013). The plan will aim the following goals.

- 2017: Evaluating applicability of existing methods in accordance with waste characteristics
- 2021: Confirming safety predictions of waste processing and disposal

The investigation plan should contain wide range of topics such as characterization, inventory estimation, storage, processing, transportation, disposal concept, disposal safety assessment, etc.

CONCLUSIONS

- The Fukushima Daiichi nuclear power station accident produced significant volume of radioactive waste.
- The waste has very different characteristics from usual radioactive waste produced in nuclear power stations due to core melt, hydrogen explosions, seawater/boron injection, etc.

- At present, the biggest issue is shortage of storage area for the waste by government policy.
- The unusual waste provides a lot of issues on storage, processing, final disposal, etc. and it should require a brand new management strategy and extensive R&D.
- R&D works such as radionuclide analysis of the waste, hydrogen generation/diffusion analysis in the storage vessel, etc. have been performed in JAEA for characterization and safe storage of the waste.
- The detailed R&D plan for processing and disposal of the waste will be established by the end of FY2012.

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