PROGRAM OF THE ANALYSIS AND RESEARCH LABORATORY FOR FUKUSHIMA-DAIICHI AND ADVANCED TECHNIQUES TO BE APPLIED IN THE LABORATORY

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ABSTRACT

Due to the Fukushima Daiichi Nuclear Power Plant accident in March 2011, the safe and secure implementations of the decommissioning have been positioned as the urgent tasks in Japan. Japan Atomic Energy Agency has a mission to analyse radioactive wastes having generated by the accident for the determination of long-term storage and disposal methods. This will be performed in two hot laboratories (Laboratory-1 and Laboratory-2) to be constructed as Okuma Analysis and Research Centre at Fukushima Daiichi Nuclear Power Plant site. In Laboratory-1, radioactive wastes such as rubbles and secondary wastes will be treated, whereas debris such as fuel debris and high dose structural materials will be handled in Laboratory-2. Currently, the detail considerations for advanced techniques and experimental apparatus to be installed are underway to start operations of each laboratory on schedule.

1. Introduction

In the Tokyo Electric Power Company (TEPCO) Fukushima Daiichi Nuclear Power Plant (1F-NPP) accident in March 2011, the reactor buildings for Unit 1-4 were damaged by hydrogen explosion due to loss of cooling system derived from tsunami following big earthquake. A lot of rubbles were generated by the explosion, and a part of them has been stored in temporary storage facilities at the 1F site but most of them have remained untouched in the destroyed reactor buildings. According to “Mid-and-long-Term Roadmap towards the Decommissioning of Fukushima Daiichi Nuclear Power Units 1-4”, which was devised by the Government of Japan and TEPCO et al [1], it is planned that the rubbles should be removed in order to permit one's access into the buildings for decommissioning operations. And they will be stored as radioactive waste in the future. In addition, the loss of cooling and feeding functions by the accident resulted in fuel melting and core melt down in reactor pressure vessel (RPV) at Unit 1-3. A part of melted core might be reached to prestressed concrete containment vessel (PCV). The nuclear fuels in these reactors are being cooled in a stable condition and radioactive material release from the reactor buildings is kept at a low level. However, much contaminated water has been generating because countermeasures for the invasion of groundwater by the building failure are insufficient. As a result, the secondary wastes such as cesium adsorption vessels and sludge have been also increasing due to the introduction of several radioactive removal systems such as Kurion, SARRY and ALPS.

Even though decommissioning operations such as removal of the rubbles and fuel debris and treatment of contaminated water will be progressed in the future based on the Roadmap, the waste treatment and disposal methods for the rubbles, secondary wastes and debris are not yet determined. In order to advance the decommissioning projects safely and timely, it is important to determine the methods soon.

Against such a background, it was determined that Japan Atomic Energy Agency (JAEA) should construct new advanced research facilities for promoting the decommissioning project with support of the Japanese Ministry of Economy, Trade and Industry (METI). The construction planning of these facilities has been considered since April 2013, and it was determined that two facilities would be constructed in Fukushima prefecture. One is a cold facility at Naraha Remote Technology Development Centre to develop remote-controlled equipment and devices for removal of fuel debris. The other has two hot laboratories (Laboratory-1 and Laboratory-2) at Okuma Analysis and Research Centre for analysing the characteristics of radioactive materials contained in the rubbles, secondary wastes and debris. In this report, the currently-considered design concepts of Laboratory-1 and Laboratory-2 were summarized. Not only the construction outline of Laboratory-1 and Laboratory-2 but also advanced techniques to be applied in these laboratories are presented.

2. Outline of facility

2.1 Purpose

Due to the 1F-NPP accident, a lot of radiated materials and contaminated materials by radioactive nuclides were generated and have been still generated. The materials are largely classified as three categories. The first one is “rubbles” including concrete segments, cut down trees, soils and so on. The second one is “secondary wastes” having been derived from decontamination equipment and multi nuclides removal equipment (ALPS). The third one is “debris” including fuel debris and high dose materials. Rubbles seem to be contaminated by I-129, Cs-137 and the other volatile nuclides released by fuel failure. On the other hand, it is assumed that the secondary wastes are contaminated by many radioactive nuclides such as Cs-137 and Sr-90. The radioactive levels of the rubbles and secondary wastes may range from background level to an order of Sv/h.

In Laboratory-1, the rubbles and secondary wastes with the surface dose up to about 1 Sv/h will be treated, and no fuel debris is included. On the other hand, higher radioactive level candidates, which are the rubbles and secondary wastes with the surface dose over about 1 Sv/h, and debris will be handled in Laboratory-2.

For the determination of the waste treatment and disposal methods of these radioactive wastes, it is necessary to evaluate the inventory, the integrity assessment during storage and so on. Furthermore, it is important to perform some examinations for clarifying the disposal configuration of the radioactive materials and development of the evaluation technology for the radioactive materials formed in the disposal configuration. Therefore, the main roles of each laboratory are determined as indicated below.

[Laboratory-1]

- Analysis of radioactive wastes such as rubbles and secondary wastes for its characterization
- Analysis for technology development of waste treatment and disposal
- Development of analytical technology

[Laboratory-2]

- Analysis of debris for its characterization
- Analysis for substantial removal of fuel debris
- Analysis for technology development of treatment and disposal of high dose wastes
- Development of analytical technology

2.2 Construction site

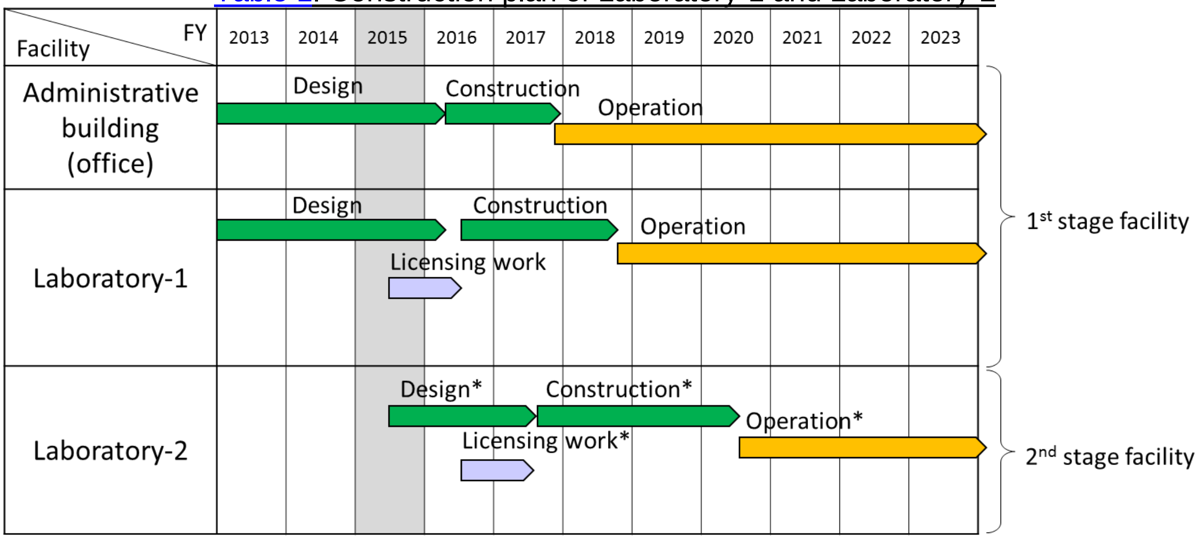
The suitable location of both laboratories to analyse these radioactive wastes is considered in terms of below requirements. Currently, the site has been tentatively selected in 1F site which is located at Okuma-machi in Fukushima prefecture.

- It should be located in 1F site for easy and safe transfer of samples.
- Air dose rate in facilities has to be low enough for low exposure of employees and accurate analysis.
- Infrastructure of utility, water and so on should be ready or to be easily prepared. And there should be little issues of logistics, such as enough width of approach roads.
- Enough area, rather flat, and no big site preparation

2.3 Construction planning

The rubbles have been stored in some temporary storage areas or left inside 1F site. For these rubbles, the determination of the waste treatment and disposal method in reactor buildings should be done as soon as possible for the acceleration of decommissioning. Especially, Laboratory-1 can begin to be constructed faster than Laboratory-2 because fuel debris is not treated in Laboratory-1 and the licensing procedure would be relatively relaxed. Furthermore, fuel debris is planned to be removed from 2021 according to the Roadmap, so that Laboratory-2 should be constructed until the start of fuel debris removal. Therefore, it is considered that Laboratory-1 should be constructed as 1st stage facility with an administrative building for office work, entrance control, and emergency response. On the other hand, Laboratory-2 will be constructed as 2nd stage facility. The construction plan of each laboratory is shown in Table 1. Now detail design of the Laboratory-1 is under way, and the operation will be started from approximately 2018. The operation of the laboratory-2 will be started in approximately 2020 by taking account for the beginning period of fuel debris removal.

Table 1. Construction plan of Laboratory-1 and Laboratory-2



*The operational start time of Laboratory-2 is under investigation including the period of design and licensing works.

2.4 Materials to be treated in facility

The detail objects of radioactive wastes to be treated in Laboratory-1 and Laboratory-2 are shown in Table 2. As mentioned in section 2.1, the rubbles and secondary wastes with the surface dose up to about 1 Sv/h will be treated in Laboratory-1. Rubbles includes concrete rubbles, metal rubbles, cut-down plants for land preparation, surface soil in 1F site, burned ash and so on. Secondary wastes derived from mainly ALPS will be candidates.

On the other hand, the rubbles and secondary wastes with the surface dose over about 1 Sv/h will be treated in Laboratory-2. In particular, the secondary wastes generated from decontamination equipment, Kurion and SARRY will be mainly handled. In addition, debris such as fuel debris, high dose reactor core/structural materials and demolition wastes will be treated.

The acceptance guidelines of the radioactive wastes are tentatively decided in Table 3 and Table 4. In Laboratory-1, 1600 samples of the radioactive wastes will be received per year. Assuming the receiving of heavy rubbles with low dose rate (<1 mSv/h), it is planned that the weight is about 100g/batch and the maximum weight is up to about 300kg/batch. In contrast, the weight of high dose rubbles (<1 Sv/h) will be limited to be under about 2kg/batch because of reducing radiation exposure and the difficulty of handling a heavy sample. On the other hand, in Laboratory-2, the detail acceptance guideline is hardly determined. Currently, it is supposed that the surface dose rate of high dose rubbles or secondary wastes is over 1 Sv/h and surface dose rate in shielding container is less than 2 mSv/h. At present, the receiving of fuel debris is expected to be 12 samples per year.

Table 2. The candidates to be used in Laboratory-1 and Laboratory-2

Items	Candidate for analysis		Lab-1	Lab-2
Rubble etc.	Rubble	Concrete, metal	○	○
	cut-down plants	Cut-down plants for land preparation	○	—
	soil	Surface soil in 1F site	○	—
	burned ash	Burned ash such as cut-down plants and hazard suit	○	○
Secondary wastes	Sludge	Decontamination equipment ・sediment of Barium sulfate, etc	(○)	○
		Advanced Liquid Processing System (Multi nuclides removal equipment: called "ALPS") ・Iron hydroxide sludge, etc	○	(○)
	Absorption material	ALPS ・Activated carbon, Titanate, chelating resin etc.	○	(○)
		Cesium absorption equipment ・Zeolite (Kurion (USA))	—	○
		Cesium absorption equipment ・Zeolite (SARRY)	—	○
Fuel debris etc.	Fuel debris	Oxide: $(U,Zr)O_2$-X, $(U,Pu,Zr)O_2$-X, etc Alloy: U-Zr-Fe, U-Pu-Zr-Fe, etc Molten corium	—	○
	Reactor core/ structural material	Reactor pressure vessel(RPV), Primary containment vessel(PCV), Constructional material in RPV	—	○
	Demolition waste	Concrete, Apparatus, etc (After fuel debris removal)	—	○

○:Treated　(○):Having a possibility of treatment　—:Not treated

Table 3. Acceptance guideline in Laboratory-1 (Not determined yet)

Items	Acceptance guideline
Rubbles	【low dose materials】 ・Surface dose rate : Lower than 1mSv/h ・Weight : About 100g/batch ・Maximum size : Needs to pass a pair of swinging doors ・Maximum weight : Up to about 300kg/batch 【high dose materials】 ・Surface dose rate : Up to about 1Sv/h (not absolute limitation) ・Weight : About 100g/batch ・Maximum size : Needs to be handled in the iron cells ・Maximum weight : Up to about 2kg/batch
Secondary wastes	

Table 4. Acceptance guideline in Laboratory-2 (Not determined yet)

Items	Acceptance guideline
Rubble etc.	・Surface dose rate in shielding container : less than 2mSv/h ・Weight : About 100g/batch
Secondary wastes	
Fuel debris etc.	・Not determined

3. Analysis plan

In Laboratory-1, it is needed to carry out analyses indicated below at least. Especially, it should be considered that there is a possibility of characteristic changes due to hydrogen explosion for the rubbles. Additionally, it is necessary to investigate whether some chlorides derived from seawater by tsunami, which can play a bad role of keeping the disposal configuration, are included or not. In Laboratory-1, most of analyses will be devoted to radioactive analysis for rubbles and secondary wastes.

- Radioactivity analysis of key nuclides (38 nuclides) which can be considered for evaluation of waste disposal method [2-3]
- Evaluation for chemical-bonding state of radioactive nuclides, which affects the diffusion coefficient under the soil
- Chemical analysis of harmful materials which influence the waste configuration and environment
- Mechanical property and physical property measurement for integrity assessment of 1F buildings and the waste configuration
- Other analysis, for example hydrogen generation rate in long-term storage.

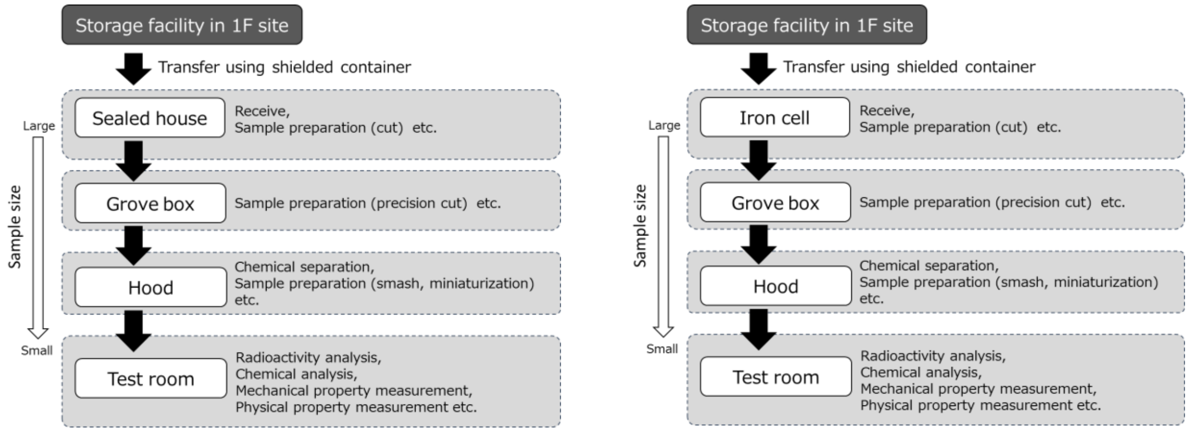
In Laboratory-2, because the characteristic of fuel debris is uncertain, the detail analysis plan is not determined yet. However, the evaluations and analyses indicated below will be carried out based on the experience of TMI-2 accident.

- Chemical and physical property measurement of fuel debris for its characterization
- Radioactivity analysis of key nuclides (38 nuclides) for high dose rubbles and secondary wastes
- Evaluation for chemical reaction such as corrosion between fuel debris and the storage flask
- Evaluation for generation of burnable gas which can cause fire and explosion in long-term storage
- Analysis for estimation of the extension behavior of the 1F-NPP accident

4. Treatment method of materials

In Laboratory-1, preparations and measurements for analyses should be rapidly and simply performed because the amount of analysis samples is estimated to be so large, which may be 1600 samples per year. Thus, the treatment of low dose rubbles and secondary wastes (<1mSv/h) should be easily handled. Therefore, as shown in Fig. 1(a), low dose materials will be transferred from a storage facility in 1F site to a sealed house of Laboratory-1. At there, heavy materials (< 300kg/batch) can be received and sample preparation will be roughly performed by hand work. In grove box and hood, precise sample preparation, chemical separation and so on will be carried out. Basically, test equipment to measure radioactive, physical and chemical properties will be applied into the test room. For high dose materials, the receiving area will be an iron cell in terms of reduction of radiation exposure, and the weight will be smaller than that of low dose ones. This is assuming that the size (or weight) to install into iron cells should be small. The material flow is indicated in Fig. 1(b). In iron cells, sample preparation will be done by remote-handling. Operations in grove box, hood and test room, will be carried out by hand work with wearing protective clothing.

In Laboratory-2, the amount of analysis samples is estimated to be so smaller than that treated in Laboratory-1. This is because the treatment of fuel debris will be carried out by remote handling using mainly concrete and iron cells. In addition, it is important that precise and careful analyses should be performed because there are many uncertain characteristics in fuel debris. The transfer method is under consideration, but it is tentatively assumed that fuel debris will be transferred into a concrete cell by using a cask. As shown in Fig. 2, sample preparation will be mainly performed in concrete cells. Precise sample preparation, chemical separation and so on will be carried out in iron cells. Then, radioactivity analysis, chemical analysis and so on will be carried out in grove box and hood. To apply techniques for non-destructive tests or systematic characterization of various fuel debris with different compositions is necessary and under consideration.



(a) Low dose material (b) High dose material

Fig. 1. Material flow in Laboratory-1

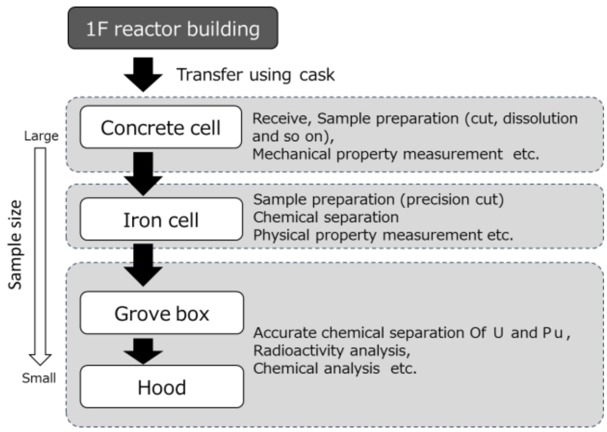


Fig. 2. Material flow in Laboratory-2

5. Advanced technology to be applied

5.1 Radioactivity measurement

For both laboratories, the characteristic and inventory evaluations of radioactive wastes will be performed by mainly radioactivity measurements and it is needed to carry out the accurate measurement. Of course, the radioactivity measuring apparatus with high sensitivity will be installed, but there is a possibility of including high-level background to the experimental data because air dose rate in 1F site is relatively high due to the released radioactive materials by fuel failure. Thus, the usual method of air ventilation, which air flow from outside toward a controlled area, should not be applied in both laboratories because the radioactive nuclides in outside air may come into inside the buildings. In particular, when the door placed in a sample receiving area is open, it must be prevented from the coming of dirty air. Therefore, intake air will be purified through high performance filters and the whole buildings will have a positive pressure against the outside. This allows to keep the high sensitivity of the radioactivity measuring apparatus to be installed.

5.2 Identification of received sample

In general hot laboratories, analyses and testing as basic research, post-irradiation examination and waste management research are basically carried out. These laboratories were constructed after prior determination of test purpose, examination contents and the characteristics of received samples in its laboratories. However, the accurate determination of especially examination contents and the characteristics of received samples are impossible for the Laboratory-1 and Laboratory-2 to be constructed in 1F site because much information of the inventory in radioactive wastes, the contamination distribution in radioactive wastes, the fuel amount of fuel debris and so on is not clear. In other words, Laboratory-1 and Laboratory-2 will be constructed in order to clarify the information. For both laboratories, radioactive wastes, which the size and radioactivity distribution is unclear, will be transferred. Thus, the analysis position should be determined from the unidentified materials as a first step. Otherwise, it seems to be difficult to go to the next steps such as chemical separation and analysis. Therefore, the application of below techniques toward each laboratory is under consideration.

[Laboratory-1]

Even though low dose radioactive wastes will be treated by hand work in a sealed house, it is too difficult to identify the analysis position from especially the large size wastes due to uncertain contamination distribution on the materials. Currently, gamma camera device which can allow non-destructive test and visualization of radioactive nuclides by simple gamma-ray measurement has been already developed, but the resolution is not good and the application of this device seems to be difficult. If the resolution is improved, the identification of analysis position would be simply determined and it is need not to carry out ineffective operations such as frequent sample preparations and pre-examinations for the determination of analysis position. In addition, the gamma camera device with high-performance may contribute to simply classify the radioactive wastes stored in temporary storage facilities at 1F site by the accurate visualization of radioactive nuclides. Therefore, the development and improvement will be positively performed toward the application of high-performance gamma camera device.

[Laboratory-2]

Debris such as fuel debris and high dose core/structural materials will be transferred using a cask. The debris is assumed to be enclosed in a metal container. It is considered that especially fuel debris will be formed in an inhomogeneous mixture with concrete and core/structural material, therefore it seems to be difficult to identify the analysis position by looks. As one of the PIE techniques, X-ray CT scan technique has been applied to non-destructive test of fuel assembly of fast breeder reactor [4]. This technique has a high resolution and can permit the visualization of thick samples. The application of the X-ray CT scan technique to fuel debris is expected to be effective for understanding the structure of fuel debris. This technique will be installed by taking account of not only penetrative power of

X-rays to fuel debris and container but also the removal method of fuel debris from metal container after the X-ray CT scan test.

6. Summary

- As one of decommissioning projects for Fukushima Daiichi Nuclear Power Plant, it is planned that Japan Atomic Energy Agency will construct new facilities for analysis and research of radioactive wastes such as rubbles, secondary wastes and debris.
- In Laboratory-1, radioactive wastes such as rubbles and secondary wastes will be mainly treated. Detail design is under way, and the operation will be started from 2018.
- In Laboratory-2, radioactive wastes such as high dose rubbles and secondary wastes will be treated. Debris such as fuel debris and high dose reactor core/structural materials will be also handled. Detail design will be started soon and the operation will be started until the beginning period of fuel debris removal.
- The introduction of advance technology to each laboratory and the improvement of non-destructive devices to both laboratories is positively under consideration, which will contribute to the acceleration of decommissioning projects.

Reference

[1] Tokyo Electric Power Company web page (accessed on Sep. 16, 2015), http://www.tepco.co.jp/decommision/planaction/roadmap/index-j.html

[2] Nuclear Safety Commission of Japan, Report on the upper bounds of radioactivity concentration for radionuclides in TRU wastes, May 2007.

[3] Japan Atomic Energy Agency, Second progress report on research and development for TRU waste disposal in Japan. Repository design, safety assessment and means of implementation in the generic phase, JAEA-REVIEW--2007-010

[4] K. Katsuyama et al, High energy X-ray CT study on the central void formations and the fuel pin deformations of FBR fuel assemblies, Nuclear Instruments and Methods in Physics Research B 255 (2007) 365–372