Waste Management in a Hot Laboratory of Japan Atomic Energy Agency – 1: Overview and Activities in Chemical Processing Facility

Kazunori Nomura, Hiromichi Ogi, Masaumi Nakahara, Sou Watanabe, Atsuhiro Shibata

***Abstract*—**Chemical Processing Facility of Japan Atomic Energy Agency is a basic research field for advanced back-end technology developments with using actual high-level radioactive materials such as irradiated fuels from the fast reactor, high-level liquid waste from reprocessing plant. In the nature of a research facility, various kinds of chemical reagents have been offered for fundamental tests. Most of them were treated properly and stored in the liquid waste vessel equipped in the facility, but some were not treated and remained at the experimental space as a kind of legacy waste. It is required to treat the waste in safety. On the other hand, we formulated the Medium- and Long-Term Management Plan of Japan Atomic Energy Agency Facilities. This comprehensive plan considers Chemical Processing Facility as one of the facilities to be decommissioned. Even if the plan is executed, treatment of the "legacy" waste beforehand must be a necessary step for decommissioning operation. Under this circumstance, we launched a collaborative research project called the STRAD project, which stands for Systematic Treatment of Radioactive liquid waste for Decommissioning, in order to develop the treatment processes for wastes of the nuclear research facility. In this project, decomposition methods of chemicals causing a troublesome phenomenon such as corrosion and explosion have been developed and there is a prospect of their decomposition in the facility by simple method. And solidification of aqueous or organic liquid wastes after the decomposition has been studied by adding cement or coagulants. Furthermore, we treated experimental tools of various materials with making an effort to stabilize and to compact them before the package into the waste container. It is expected to decrease the number of transportation of the solid waste and widen the operation space. Some achievements of these studies will be shown in this paper. The project is expected to contribute beneficial waste management outcome that can be shared world widely.

***Keywords*—**Chemical Processing Facility, medium- and long-term management plan of JAEA Facilities, STRAD project, treatment of radioactive waste.

I. Introduction

JAPAN Atomic Energy Agency (JAEA) released the Medium- and Long-Term Management Plan of JAEA Facilities on April 30, 2017 (it was revised on April 30, 2018) [1]. In this plan, facilities were sorted in two categories; one is to be continued to use and the other is to be decommissioned near future. According to this plan, our research facility, Chemical Processing Facility (CPF, Fig. 1), is sorted into the latter. However, the decision on the concrete decommissioning schedule of CPF will be discussed anew in JFY 2021, because there are still some ongoing research and development plans to be continued for several years.

CPF is a basic research facility where hot experiments have been performed by using actual irradiated mixed oxide (MOX) fuels of fast reactor (FR) and high-level radioactive liquid wastes generated from the Tokai Reprocessing Plant (TRP) in Japan. Moreover, after the accident in Fukushima Daiichi Nuclear Power Stations, analytical support on a number of varied samples taken in the Fukushima site was added as one of the important purposes of CPF.

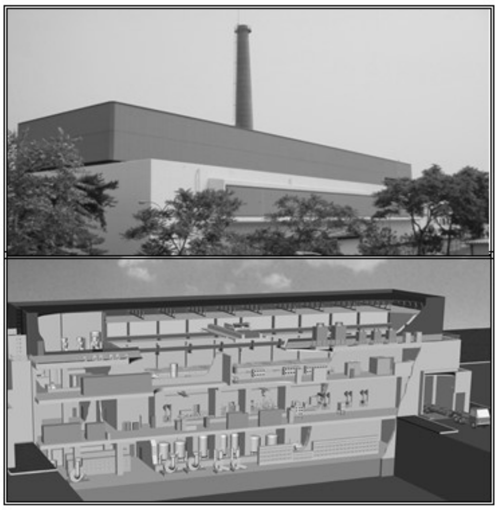

Fig. 1 Outlook of Chemical Processing Facility (CPF)

Through the experiments and analysis, a little but various kinds of liquid and solid waste have been produced in the hot equipment such as concrete shielded cells and glove boxes. Some of those wastes have remained in the equipment and their treatment issue has been questioned conventionally. Under this circumstance, we recently started research and development on treatment process of these wastes strenuously. It is required to keep the experimental field clean from the safety reason for the ongoing and future various experimental activities and some of the developed treatment processes will be serve the purpose of preparation for the decommissioning of research facilities.

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II. Medium- and Long-Term Management Plan of JAEA Facilities

JAEA has been leading the research and development on nuclear science and technology in Japan for several decades. There are various nuclear facilities and many of them were constructed more than 40 years ago. In recent years, the measures against the aging of these rather old facilities are becoming one of the most serious concerns on this account. Namely, the increase of cost for the facilities aging management will be inevitable. In addition to this, there are some of the old facilities which have fulfilled their roles. It is possible to reduce their fundamental risk and maintenance cost by their decommissioning. Meanwhile, the back-end issue such as their decommissioning itself and related disposal of radioactive waste will require serious cost. After the Great East Japan Earthquake on March 11, 2011, the regulation was enforced from the viewpoint of earthquake- resistance while conventional regulation standard was reviewed. The situation in which a large amount of budget should be prepared was actualized gradually. We must response to the aging of nuclear facilities, the new regulations, and the back-end issue including decommissioning. However, it is obviously difficult to operate all facilities as before within the limited resources. Under these circumstances, JAEA formulated the Medium- and Long-Term Management Plan of JAEA Facilities. This comprehensive plan focuses on three objectives, namely, selection and consolidation of JAEA's nuclear facilities, measures to keep safety of facilities and management of back-end issue (Fig. 2). This plan identifies 45 facilities that will be survived and continue to be used and 44 facilities that will be decommissioned, narrowing down the former category of facilities through the selection and consolidation of currently existing facilities and measures to take with regard to both categories of facilities.

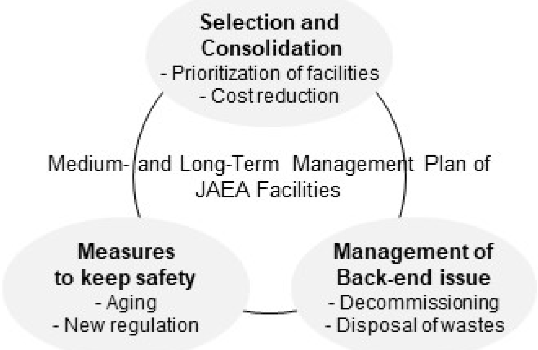

Fig. 2 Three objectives of Medium- and Long Term Management Plan of JAEA Facilities

III. Management Plan of CPF

A. Main Features and Experimental Wastes of CPF

Our research facility, CPF, was constructed in 1980 at Nuclear Fuel Engineering Laboratories of JAEA. Initial purposes of CPF were researches on reprocessing of spent FR fuels and on vitrification process of high-level radioactive liquid wastes and performance of glass. The facility has a research building with 3 floors above and 1 floor below the ground (lower sectional view in Fig. 1). In this building, there are two lines of the concrete shielded cells and 4 laboratories where glove boxes and hoods are installed. CPF is permitted to use irradiated and non-irradiated MOX fuels and high- level radioactive liquid wastes generated in the nuclear fuel reprocessing process. Maximum amount of nuclear material and radioactivity are regulated to each installation. Specifically, for example, up to 220 g of sum of Pu, ^{235}U and ^{233}U can be used in certain cell.

The first experiment with the use of radioactive material was performed in 1982, then applicability of PUREX process on FR MOX fuels with high burn-up and high Pu content has been examined through more than 20 series of active experiments with FR (Joyo, Phoenix and DFR) irradiated fuels from 1982 to 2005. Several fuel pins were treated in a series of the aqueous reprocessing experiments, i.e. shearing mechanically pin by pin, dissolving into nitric acid solution and solvent extraction with tributyl phosphate (TBP) in n- dodecane, which is generally applied for the nuclear fuel reprocessing process that is well known as the PUREX process. During this period, fundamental data on the vitrification of high-level active waste solution generated from TRP were collected systematically and that information was already reflected in design and operation of Tokai Vitrification Facility. The research activity in this field is now limited to basic research of disposal such as leaching test.

Concerning the research field for the FR fuel reprocessing technology development, mechanical system and composition of experimental equipment, utility pipes and vessels had been fixed just as mini reprocessing plant, because the initial design of these installments had been made for the demonstration of the essential applicability of the PUREX process to the FR spent fuel reprocessing. In order to contribute to the development of new technologies for the advanced FR cycle system, much more instruments, space and flexibility were required, and then the renovation of the CPF was carried out from 1996 to 2002 in accordance with new design [2].

New research projects of advanced aqueous reprocessing and of pyrochemical reprocessing were started after the renovation. They are for the innovation of reprocessing technology. Both aqueous process and pyroprocess have been studied aiming for establishing the future FR cycle.

Basic concept of innovative reprocessing is TRU recovery and their recycle use. Since TBP has poor extractability against trivalent actinides (Am and Cm), it is necessary to develop and to introduce new extractant. For example, addition of the extraction or extraction chromatography processes by new solvent such as bidentate organic ligand after U/Pu/Np recovery by the PUREX process have been proposed for trivalent minor actinides recovery [3]. Various new developed extractants other than TBP have been applied to high level raffinate generated from the PUREX process. So far, about 10 hot trials with actual high level raffinate for the development of trivalent minor actinides recovery process have been carried out in the concrete shielded cell. On the other hand, special glove boxes with Ar atmosphere, which equip electric furnace, are installed in one laboratory for the pyroprocess development under collaboration with Central Research Institute of Electric Power Industry. Several electrorefining experiments with a few grams of non-irradiated U and Pu have been carried out. The chloride compound such as lithium chloride and potassium chloride, which are medium of molten salt phase, adheres to surroundings of the metal product obtained as the result of experiment. Experimental studies about those new reprocessing technologies are still undergoing and will be producing some kinds of liquid waste, which had not been obtained before the renovation; their treatment method should be taken into account. In fact, organic solvent waste of new extractants and inorganic liquid waste of chloride solution are remained in the experimental equipment of CPF with or without containing nuclear materials. Moreover, after the accident in Fukushima Daiichi Nuclear Power Stations, analyses of various kinds of samples taken in the Fukushima site have been carried out in CPF [4]. The samples are mainly the contaminated water that circulates the failed fuel cooling system and the secondary wastes from the decontamination systems that are installed in order to reduce radioactivity concentration of the used cooling water. Since some samples contain certain portion of sea water and a large variety of reagents which are constituents of the secondary wastes from the decontamination system, these R&D and analyses also bring deferent types of liquid waste to treat safely and effectively. Apart from that, there was old liquid waste; so- called the “legacy waste”, which contained another chemical reagent than that generally used in the PUREX process. Some trials for the improvement of the PUREX process by using reagents, such as redox reagents, complexing reagents, had been carried out even before the renovation of the CPF and their treatments had been put off so far. Meanwhile, not only the liquid waste but also used experimental tools, such as polypropylene bottles of various size, counter current extractors (Fig. 3), plastic tubes and so on, were regularly produced after experiments and analysis.

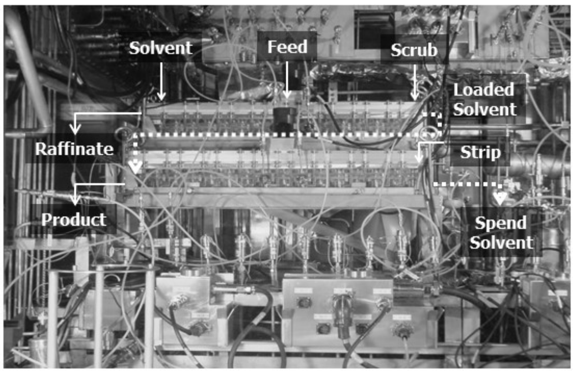

Fig. 3 Mini centrifugal contactor installed in shielded cell

B. Position of CPF in the Medium- and Long-Term Management Plan of JAEA Facilities

According to the Medium- and Long-Term Management Plan of JAEA Facilities, CPF is in the category of facilities to be decommissioned in near future. Nevertheless, some situation of CPF is different from other facilities to be decommissioned because of the feature of CPF. We are continuing research and development activities not only with certain amount of actinide elements obtained from actual irradiated FR fuel but also with various radioactive samples from Fukushima Daiichi Nuclear Power Stations. There are experimental study plans to be continued at least for more than several years. Many of these programs are implemented through competitive funding. Besides the aging guide of decommissioning is 40 years but CPF has been in operation for less than the guide. So there is a notice in the Medium- and Long term Management Plan of JAEA Facilities that the definite schedule and the facility to be handed down to will be discussed on another occasion in JFY 2021.

We roughly estimate that decommissioning of CPF will take about 30 years if it is going very well. Firstly, irradiated fuels stored in CPF are treated in order to recover U and Pu. Secondly, recovered nuclear materials are transported from CPF to other facilities. Thirdly, in order to reduce the dose rate in the concreate shielded cells and glove boxes, solid and liquid wastes remained in these installations are also transported to appropriate facilities after the waste treatment for stabilizing them in safety form. Then existing vessels, pipes, and some devices in the cells are removed and inside the cells are decontaminated remotely up to enough level for direct decontamination operation. Certain volume of secondary liquid waste is generated in each step as a matter of cause and it must be treated for effective disposal.

IV. R&D for Treatment of Waste in CPF

In the case that CPF is going to continue the research and development activities, the remaining liquid waste in the equipment must be treated in order to keep enough area for experimental operation in safety. On the other hand, if the decommissioning of CPF is decided, the treatment of wastes for cleaning up the concreate shielded cells and glove boxes is required firstly, because the treatment and disposal of these kinds of liquid wastes reduces radiation doze rate of environment in the concrete shielded cells and glove boxes.

TABLE I
Typical Chemicals in Liquid Waste

Place of generation	Phase	Representative chemicals
Experiments	Aqueous	Nitric acid, Lactic acid, Hydrazine, Hydroxyl amine nitrate
	Organic	TBP, Dodecane, CMPO, and their degradation products (with leaving to load actinide elements in some items)
Analysis	Aqueous	Nitric acid, Ammonium peroxodisulfate, Hydrazine, Hydroxyl amine nitrate, Sulfamic acid, oxidants, reductants,
	Organic	Pyridine, TOPO, Ethyl acetate, xylene, TTA, TBP, DBP

Various kinds of radioactive liquid wastes (Table I) are taking up so much space in the concrete shielded cells and glove boxes. JAEA started fundamental studies to develop new technologies for processing these accumulated liquid wastes. The study is conducted as a collaborative research project with universities, institutes and private companies, who have a lot of awareness of this issue and scholarly interest in common. The project was named as the STRAD, which stands for Systematic Treatment of Radioactive liquid waste for Decommissioning [5]. For example, concentration of radioactive liquid waste containing various chemical compounds using the reverse osmosis membrane has been examined for efficient and safety treatment of the liquid wastes accumulated in the hot laboratories. It makes possible to reduce volume of the liquid to treat without concentration by heating, which requires heating system and significant time in the small laboratory. Decomposition methods of chemicals, such as ammonium salt [6], organic acid, etc., have been investigated, because such chemicals may cause a troublesome phenomenon. Solidification of aqueous or organic liquid wastes has been also studied by adding cement or coagulants. Although these studies have been progressed to maintain our hot laboratory for the moment, treatment of the various kind of accumulated liquid wastes in the step of decommissioning or dismantling of the facilities would be also one of the most difficult challenges and those information and knowledge are expected to apply the step of decommissioning.

In the equipment of the facility, there also remain the high level radioactive solid wastes; flammable and non-flammable waste, solid reagent waste and so on. In the case of CPF, these high level radioactive wastes are packed in a specialized drum and then the drum is conveyed in a storage facility in TRP. Since the number of drum to be conveyed at one time is limited, volume reduction of the solid waste before packing in the drum is important for the efficient maintenance and management of the facility. For example, being a bulky waste, a plastic bottle, which is frequently used in hot experiments, occupies unnecessary space in the drum when it is packed as it is. However, its low melting point brought rather easy method of reducing waste volume. A simple apparatus of waste volume reduction was designed and it has been working in an adequate performance.

V. Conclusion

Whether CPF is going to survive or to be decommissioned, the treatment of wastes for cleaning up the concreate shielded cells and glove boxes is required for subsequent events. Although this kind of attempt for waste treatment tends to become routine work, scientific challenge to apply an up-to-date idea will make it attractive when its effect highlights the effect and need for the facility waste management [7], [8].

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