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Waste Management in a Hot Laboratory of Japan Atomic Energy Agency – 3: Volume Reduction and Stabilization of Solid Waste

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***Abstract*—**In the Japan Atomic Energy Agency, three types of experimental research, advanced reactor fuel reprocessing, radioactive waste disposal, and nuclear fuel cycle technology, have been carried out at the Chemical Processing Facility. The facility has generated high level radioactive liquid and solid wastes in hot cells. The high level radioactive solid waste is divided into three main categories, a flammable waste, a non-flammable waste, and a solid reagent waste. A plastic product is categorized into the flammable waste and molten with a heating mantle. The non-flammable waste is cut with a band saw machine for reducing the volume. Among the solid reagent waste, a used adsorbent after the experiments is heated, and an extractant is decomposed for its stabilization. All high level radioactive solid wastes in the hot cells are packed in a high level radioactive solid waste can. The high level radioactive solid waste can is transported to the 2nd High Active Solid Waste Storage in the Tokai Reprocessing Plant in the Japan Atomic Energy Agency.

***Keywords*—**High level radioactive solid waste, advanced reactor fuel reprocessing, radioactive waste disposal, nuclear fuel cycle technology.

I. Introduction

THE Chemical Processing Facility (CPF) was built in Tokai-mura, Ibaraki. The facility has some hoods, glove boxes, and hot cells to handle radioisotopes and nuclear materials for beaker-scale experiments. The research and development for advanced reactor fuel reprocessing, radioactive waste disposal, and nuclear fuel cycle technology have been mainly carried out in the facility.

In the advanced reactor fuel reprocessing, aqueous and dry (non-aqueous) processing experiments are attempted in the CPF. The experiments for aqueous processing have been mainly conducted in the hot cells. Some irradiated nuclear fuel pins of fast reactor "Joyo" were transported from Oarai-machi, Ibaraki, to study the advanced reactor fuel reprocessing. The aqueous processing for a fast reactor fuel reprocessing is named the New Extraction System for Transuranic (TRU) Recovery (NEXT) [1]-[3]. The NEXT process basically consists of a mechanical disassembly, a short stroke shearing, a highly efficient dissolution, a U crystallization, a simplified solvent extraction for U, Pu, and Np co-recovery, and an extraction chromatograph for minor actinides (MAs) recovery [3]-[8]. The irradiated nuclear fuel pins are sheared short with a decladding machine to dissolve them efficiency. The short stroke sheared fuel pins are dissolved with HNO_3 solutions. A part of U crystalizes as uranyl nitrate hexahydrate by cooling the dissolver solution derived from irradiated fast reactor "Joyo" fuels. The U, Pu, and Np are co-recovered from the mother liquor of the U crystallization process by the solvent extraction process with centrifugal contactors. Among MAs, Am and Cm in a raffinate are separated from fission products (FPs) and lanthanides by the extraction chromatograph. On the other hand, the dry processing for the advanced reactor fuel reprocessing has been investigated in glove boxes which are managed with Ar atmosphere because metallic nuclear materials are handled. In the dry processing, metallic U and Pu are separated in a molten salt by a pyrochemical process, and the Japan Atomic Energy Agency (JAEA) conducts this process in cooperation with the Central Research Institute of Electric Power Industry (CRIEPI) with non-irradiated mixed oxide (MOX) fuel pellets. The MOX fuel is reduced, and metallic U and Pu are recovered in the molten salt by an electrorefining. The recovered metallic U and Pu are distilled to remove the attached molten salt on the surface of the metal.

To study the radioactive waste disposal, a vitrification was made from a high level radioactive liquid waste which was brought from the Tokai Reprocessing Plant (TRP) in the Nuclear Fuel Cycle Engineering Laboratories of the JAEA. The properties of the vitrification are evaluated in the hot cells. In addition, immersion experiments are carried out with the vitrified waste. The radionuclide profiles of FPs and MAs in a bentonite are obtained in diffusion experiments.

Regarding the nuclear fuel cycle technology, research on materials science has been carried out for a reprocessing equipment. The corrosion behavior of metals used for reprocessing equipment is evaluated in nuclear material nitrate solutions. An advanced analysis technology is also developed for the advanced reactor fuel reprocessing and a recovery of the Fukushima Daiichi Nuclear Power Station accident.

In the experiments for the advanced reactor fuel reprocessing, radioactive waste disposal, and nuclear fuel cycle technology, our facility has generated the high level radioactive solid waste in the hot cells. It is divided into three main categories, a flammable waste, a non-flammable waste, and a solid reagent waste, in our facility. In this paper, we mainly report the treatment of high level radioactive solid waste derived from the aqueous processing for the advanced reactor fuel reprocessing

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and the transportation of high level radioactive solid waste in the hot cells from the CPF to a solid waste storage.

II. Treatment of High Level Radioactive Solid Waste

A. Volume Reduction of High Level Radioactive Solid Waste

A plastic product and a waste cloth are categorized into the flammable waste in the CPF. Among the plastic product, we have tried to reduce the volume of plastic bottles because there are many used plastic bottles in the hot cells. The large volume plastic bottles are cut with a band saw machine in the hot cells. We tried to melt the small volume plastic bottles with a heating mantle for their volume reduction because it is difficult to cut small volume plastic bottles by remote manipulation in the hot cells. The experiments for the small volume plastic bottles were carried out to evaluate the effect of the volume reduction before the heating mantle is installed in the hot cells. The heating mantle has a polytetrafluoroethylene beaker (ϕ150 mm × 200 mm) and a thermocouple.



Fig. 1 Molten polypropylene bottles after heating

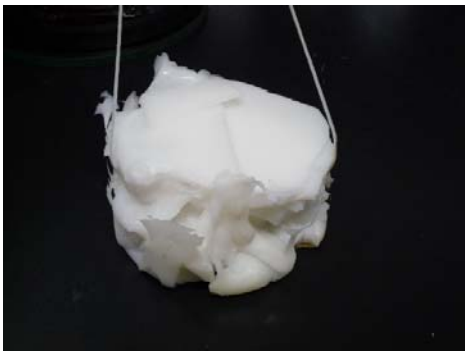


Fig. 2 Molten polyethylene container after heating

0.25 dm^3 polypropylene bottles were molten for the volume reduction. Temperature in the heater was controlled by a thermostat and set at 453 K. During the operation, temperatures in the beaker of the heating mantle and at the outer side of the heating mantle were 449 and 323 K, respectively. And then, a 0.25 dm^3 polypropylene bottle was put in the beaker and heated for 30 min, and another 0.25 dm^3 polypropylene bottle was put in the beaker every 10 min. Finally, three 0.25 dm^3 polypropylene bottles (two plastic bottles with caps and a plastic bottle without a cap) were molten. Fig. 1 shows the molten polypropylene bottles after heating. In the experiment, the volume was reduced by approximately 80%. In addition, 10 dm^3 polyethylene container was also molten for the volume reduction. To put in the beaker of the heating mantle, 10 dm^3 polyethylene container was cut. Approximately, two thirds of the cut 10 dm^3 polyethylene container was put in the beaker of the heating mantle. The temperature in the heater was set at 453 K and the heating mantle was operated for 60 min. The temperatures in the beaker of the heating mantle and at the outer side of the heating mantle were 449 and 323 K after heating for 60 min, respectively. Fig. 2 shows molten polyethylene container after heating. Approximately, two thirds of the cut 10 dm^3 polyethylene container was molten, and approximately 1 dm^3 of molten polyethylene was finally obtained.



Fig. 3 Sawing of the centrifugal contactor with the band saw machine

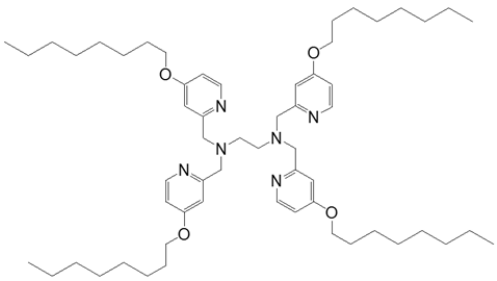


Fig. 4 Chemical structure of TOPEN

The non-flammable waste means mainly metal, and scientific glassware in our facility. The centrifugal contactors are used for the simplified solvent extraction process for U, Pu, and Np co-recovery. Two banks of the centrifugal contactor are installed in the hot cell. The centrifugal contactor mainly consists of a motor, a gearbox, rotors, and casing for the rotors. The gearbox and rotors of the centrifugal contactor are made of Al and stainless steel, respectively. The casing for the rotors is made of acrylic board. The centrifugal contactor is too large to dispose in a high level radioactive solid waste can. Therefore, the centrifugal contactors were taken apart as small as possible by remote manipulation in the hot cell. In addition, the gearbox and the casing for the rotors were sawed with a band saw machine. The photograph of sawing of the centrifugal contactor in the hot cell is shown in Fig. 3. A non-flammable sheet was laid because it is easy to recover the chip of the centrifugal

contactor. The centrifugal contactor was finally sawed into three pieces. It took approximately 7 h per a sawing for the main body (gearbox and casing for the rotors) of the centrifugal contactor.

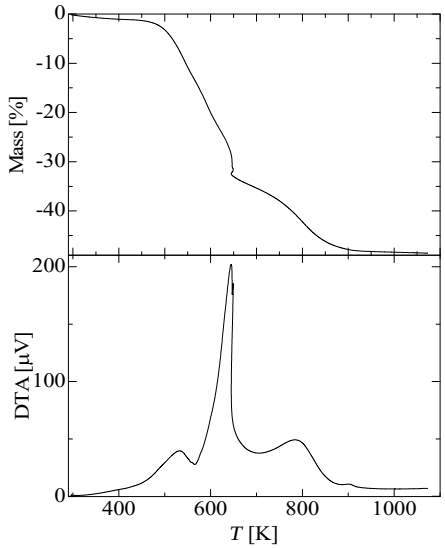


Fig. 5 TG/DTA curves of TOPEN impregnated adsorbent

B. Stabilization of High Level Radioactive Solid Waste

In the simplified solvent extraction process for U, Pu, and Np co-recovery, tri-*n*-butyl phosphate diluted *n*-dodecane is used as an extractant. The spent solvent is transferred to a tank for the organic solvent after it is analyzed. The adsorbent used in the extraction chromatography for MAs recovery consists of porous SiO_2 support, styrene-divinyl benzene co-polymer, and an extractant which interacts with MAs. A spent adsorbent should be treated as a solid waste, and chemical stability is one of the important factors in terms of longtime storage of the solid waste. It is concerned that the organic components in the adsorbent radiologically decompose and release explosive gas such as H_2 during the storage. The extractant and the polymer should be removed from the adsorbent in advance of the storage.

One of typical adsorbents which has been used in the hot cells of the CPF is *N*,*N*,*N'*,*N'*-tetrakis((5-octyloxypyridin-2-yl)methyl)ethylenediamine (TOPEN, Fig. 4 [9]) impregnated adsorbent, and TOPEN has relatively low ignition point (325 K). Therefore, the removal of TOPEN in the spent adsorbent was one of essential treatments.

Thermal decomposition of TOPEN was revealed in the consequence of systematic investigations on durability of TOPEN against acid and irradiation. Thermogravimetry (TG) / differential thermal analysis (DTA) curves of the TOPEN impregnated adsorbent are shown in Fig. 5. The predominant exothermic peaks were observed at about 600 and 800 K, and the weight reduction at the temperatures corresponded to decomposition of the polymer [10]. The organic compounds must be properly removed from the adsorbent at about 950 K regarding their mass fractions involved in the adsorbent (weight ratio, SiO_2: organic compounds = 55: 45). Therefore, exothermic peak at about 550 K would be caused by the TOPEN decomposition.

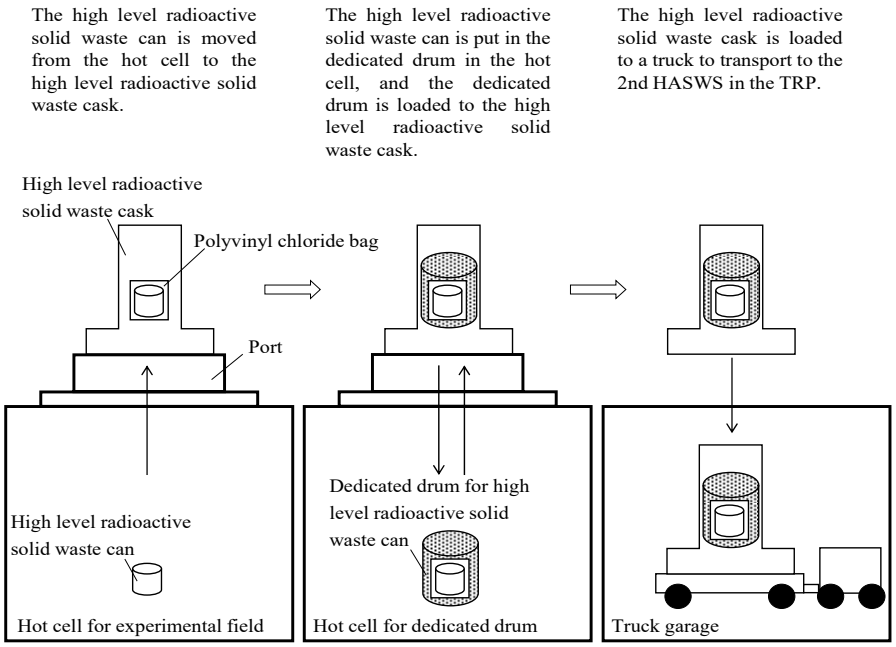


Fig. 6 Flow of transportation of high level radioactive solid waste can from the CPF to the 2nd HASWS in the TRP

Based on the above evaluation, a thermal treatment for 1 g of TOPEN impregnated adsorbent using an electric furnace was carried out to check the removal behavior of the organic compounds under non-radioactive environment. The adsorbent

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was heated at 973 K for 90 min, and white smoke was observed during heating. The yellow TOPEN was completely disappeared after heating, and the weight of the remaining particles was 55% of the initial value. The organic compounds were successfully removed. Given the fact, the spent adsorbent was also heated with the electric furnace installed in the hot cell, and similar thermal behavior was confirmed. In the previous study, it is known that most of gas generated from adsorbents is CO_2 when similar adsorbents are heated [11]. Therefore, it would treat safely a large amount of the adsorbent in the hot cells.

III. Transportation of High Level Radioactive Solid Waste

Fig. 6 shows flow of transportation of high level radioactive solid waste can. The molten plastic bottles, the disconnected experimental equipment, for example centrifugal contactor, and stabilized adsorbents etc. are packed in the high level radioactive solid waste can. The solid waste is taken a picture, measured its size and weighted before they are packed in the high level radioactive solid waste can. And then, a lid is put on the high level radioactive solid waste can. The can is moved from the hot cell for experimental field to a high level radioactive solid waste cask above the hot cell. The high level radioactive solid waste can is moved to another hot cell to put it in a dedicated drum for high level radioactive solid waste can. The dedicated drum is placed in the hot cell which is not contaminated with radioactive materials. The dedicated drum is loaded to the high level radioactive solid waste cask, and then weighted and measured radiation dose on its surface. The cask is loaded to a truck to transport from the CPF to the 2nd High Active Solid Waste Storage (2nd HASWS) in the TRP in the Nuclear Fuel Cycle Engineering Laboratories of the JAEA. The high level radioactive solid waste cask is returned from the 2nd HASWS to the CPF after being unloaded the dedicated drum containing the high level radioactive solid waste can.

IV. Conclusion

Some experiments for the advanced reactor fuel reprocessing, the radioactive waste disposal, and the nuclear fuel cycle technology have carried out with radioisotopes and nuclear materials in the hot cells in the CPF. The facility has generated the high level radioactive solid waste in some experiments in the hot cells. To reduce their volume, the plastic product was heated with the heating mantle, and the non-flammable waste is sawed with the band saw machine. The extractant in the used adsorbent for the extraction chromatograph process is decomposed by heating to disposal stably. All the high level radioactive solid wastes are put in the high level radioactive solid waste can and transported to the 2nd HASWS in the Nuclear Fuel Cycle Engineering Laboratories. It would be necessary to try further to reduce the volume of the high level radioactive solid waste, and we should address simpler stabilization method of the high level radioactive solid waste for safe disposal.

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