Treatment of Spent Solvent in STRAD Project

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1. Introduction

Radioactive liquid wastes generated in nuclear facilities such as nuclear power plants, reprocessing facilities, research reactors and laboratories often contain not only radiotoxic isotopes but also hazardous chemicals. Japan Atomic Energy Agency has been conducting STRAD (Systematic Treatment of RAdioactive liquid waste for Decommissioning) project for treatment of legacy liquid wastes accumulating in hot laboratories [1]. In this project, fundamental studies to develop new technologies for the treatments are conducted utilizing stored liquids in Chemical Processing Facility (CPF) of Japan Atomic Energy Agency as reference. The facility has been operating mainly for development of reprocessing technologies of spent fast reactor fuel. More than 30 years operation with irradiated MOX fuel has generated many kinds of liquids, and those would be ideal targets of the studies. Several collaborative studies with universities, national organizations and private companies are launched to deal with each challenging task.

One of the challenging tasks is treatment of spent organic liquids involving spent solvent loading radioactive nuclides. Degradation products of the solvent sometimes obstruct efficient stripping of the radioactive elements. Removal of the radioactivity from the organic solvent is the principal task of the treatment for the safety reasons. Enhancement in chemical stability of the solvent for disposal or long-time storage must be the following subject. An appropriate treatment procedure has to be designed depending on compositions of the liquid. In this study, treatment procedure of spent organic solvent was experimentally tested on simulated spent solvent and genuine accumulated solvent in CPF of JAEA.

2. Treatment procedure

Elemental technologies for the liquid waste treatments could be roughly categorized into removal of radioactive elements, removal or destruction of chemicals such as ammonia, separation of phases, concentration, incineration, oxidation, acid digestion and solidification according to several reports on radioactive liquid waste management [2-6]. Treatment of the organic liquid waste is also expected to be designed by combining those elemental technologies. In this study, spent TRUEX solvent containing CMPO, TBP and normal dodecane was selected as a target liquid, and treatment procedure was considered and examined.

According to our previous study, almost all U and Pu extracted by CMPO, TBP and DBP, which is a typical degradation product of TBP, are possible to be recovered in alkaline solution by multiple stages lavage operation [7]. Small amount (about 0.01 g/L) of U and Pu still existed in the spent TRUEX solvent used for SETFICS process experiments carried out in 1996 [8]. Batch-wise lavage experiments with NaOH solution were carried out on the stored spent solvent inside the hot cell in order to recover them in aqueous solution. 100 mL of the spent solvent and 1 M NaOH solution was mixed in a plastic bottle and shaken for 30 min. After phase separation, the same lavage operation was repeated to enhance U and Pu recovery efficiency. In those operations, more than 98 % of U and Pu in the solvent was expected to be recovered as precipitation.

Another our study suggested that solidification of organic liquids with coagulants is possible although some improvements on the coagulants are required to enhance stabilities the solids. Geopolymer is one of promising material for solidification of organic liquids [9]. In this study, confinement of simulated solvent into geopolymer was experimentally tested. 5 wt% of PUREX or simulated spent TRUEX solvents were mixed with liquid glass and metakaolin after adjusting pH. The mixed pastes have been stored inside a temperature-controlled bath with 293 K for more than 1 month.

3. Results

Decontamination experiments in a hot cell

The TRUEX solvent stored in a hot cell of CPF and the operations inside the hot cell were shown in Fig. 1, where the muddy liquid and the transparent liquid were the TRUEX solvent and the NaOH solution, respectively. Since color of new TRUEX solvent is transparent and amount of the loaded Pu was not significant, the turbidity must be caused by degradation of organic compounds. Those liquids were mixed in a bottle and shaken by the device shown in the figure. The Organic and the aqueous phases were separated using a separating funnel. The solvent was still muddy even after the lavage operation, whereas the alkaline solution also got cloudy. The loaded cations were considered to be dispersed in the aqueous phase as Na2U2O7 and Pu(OH)4. Those precipitations were recovered by filtering, and amount of recovered U and Pu are currently analyzed to evaluate performance of the lavage operation.

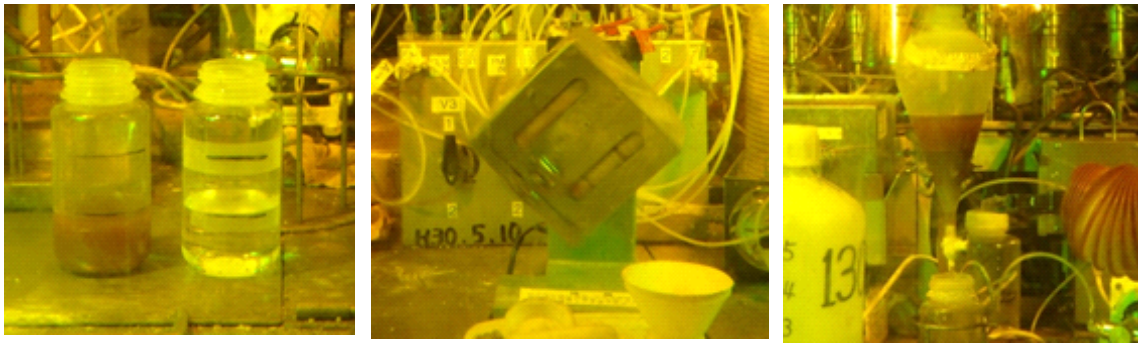

Figure 1. The lavage operation of the spent solvent in the hot cell of CPF

(Left) The spent TRUEX solvent and NaOH solution (Middle) Shaking device used in the hot cell (right) Phase separation operation

Inactive solidification experiments

Simulated PUREX solvent was successfully solidified with geopolymer as shown in Fig. 2. However, distinct cracks were found on surface of the solid, and the number of the cracks increased with time. The cracks were not found in a solid prepared without the organic solvent. Solidification of the geopolymer progresses by polymerization of silicate through dehydration condensation reaction, and droplets of organic liquids are considered to be dispersed inside the amorphous polymer. Size and/or concentration of the droplets might be too large for stable solidification. In order to evaluate chemical stability of the organic liquid inside the geopolymer, several analyses such as observation inside the solid, leaching test must be necessary. Improvements in the solidification conditions based on the analyses are expected to enhance stability of the solvent inside the geopolymer. Analyses on the solid and solidification experiment on TRUEX solvent are currently underway, and solidification of the genuine spent TRUEX solvent will also be examined in near future.

Figure 2. Solidified geopolymer with new PUREX solvent

4. Summary

Treatment of spent TRUEX solvent was experimentally tested as a part of STRAD project for liquid waste management generated in hot laboratories. Lavage of spent TRUEX solvent with NaOH solution was successfully done inside the hot cell, and U and Pu loaded in the solvent were expected to be recovered in the aqueous phase as precipitation. New PUREX solvent was solidified with geopolymer although several improvements in the solidification conditions should be required. Solidification experiments on the TRUEX solvent is progressing.

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