# Treatments of Radioactive Waste Solutions Generated In A Hot Laboratory of Japan Atomic Energy Agency

Sou Watanabe, Hiromichi Ogi, Yoichi Arai, Haruka Aihara, Atsuhiro Shibata and Kazunori Nomura

*Japan Atomic Energy Agency, Ibaraki, Japan*
*Corresponding author: Sou Watanabe <watanabe.sou@jaea.go.jp>*

## History and current status of CPF

Chemical Processing Facility (CPF) was constructed in 1980 in Nuclear Fuel Engineering Laboratories of Japan Atomic Energy Agency (JAEA) for researches on reprocessing of spent fast reactor (FR) fuels and on vitrification process of high level radioactive liquid wastes. The facility equips hot cells, glove boxes and hoods, and irradiated and non-irradiated MOX fuels can be handled. The first experiment on radioactive samples 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 21 times active experiments on fast reactor (Joyo, Phoenix and DFR) irradiated fuels from 1982 to 2005. Few fuel pins were treated in a series of the aqueous reprocessing experiments i.e. shearing, dissolving and solvent extraction processes. During this periods, fundamental data of vitrification on high level active waste solution generated from Tokai Reprocessing Plant was systematically collected, and that information contributed to design of Tokai Vitrification Facility (TVF).

In 1996-2002, renovation of the facility was carried out to start new research projects of advanced aqueous reprocessing and of pyrochemical reprocessing (Aose et al., 2007). In the new aqueous reprocessing, separation procedure in the PUREX was replaced by crystallization of U, U/Pu/Np co-recovery by modified PUREX flow-sheet and trivalent minor actinides (Am and Cm) recovery by solvent extraction or extraction chromatography (Funasaka & Itoh, 2007). About 10 times experiments for the new process have been carried out in the hot cell so far. Special glove boxes with Ar atmosphere which equip electric furnace were installed in a laboratory for pyroprocess under collaboration with Central Research Institute of Electric Power Industry (CRIEPI), and electrorefining experiments on few grams of U and Pu has been carried out (Kitawaki et al., 2011). Experimental studies about those new reprocessing technologies are still undergoing.

After the accident in Fukushima Daiichi Nuclear Power Stations, not only R&D on the reprocessing but also various analyses on samples taken in the Fukushima site are one of the most important tasks of CPF. Currently future operation and maintenance plans of CPF are under discussions in JAEA.

## Waste solution stored in CPF

More than 30 years experimental and analytical activities inside the hot cells and glove boxes have produced plenty amount of radioactive liquid wastes. The facility has waste solution tanks for storage of active solutions however it does not equip functions of processing the solutions for disposal. Nuclear fuel materials in the waste solutions were recovered using conventional reprocessing procedures and then they were converted to oxide form for storage. The waste solutions with relatively simple compositions such as nitic acid solution or spent PUREX solvent have been

transferred into the tanks after the U and Pu recovery. The solutions containing reactive chemical compounds have not been mixed with each other in the tanks in order to avoid unexpected hazardous chemical reactions during the storage, and those have been temporarily stored inside the hot cells or glove boxes separately in small bottles. Those solutions will obviously be one of the most troublesome wastes at the time of decommissioning of this facility which is already under discussion,

Those chemical compounds are necessary to be removed or decomposed before mixing with other solutions inside the shielded environment with specific restrictions. Appropriate treatment procedures of those solutions have to be individually developed after careful risk assessment. JAEA has started systematically investigation on safety treatments of the radioactive waste solution accumulated in the CPF from 2015, and several kinds of the solutions generated by the experiments have been successfully processed inside the hot cell and residual solutions were transferred into the tanks. However, treatment procedures for large part of solutions generated by the analyses and organic solutions are still uncertain due to their complicated compositions. In order to develop appropriate processes for those liquids, collaborative research programs with several universities and national research organizations were started from 2017.

**Treatments of waste solutions in CPF**

Treatments on most of the aqueous experimental wastes and on a part of the aqueous analytical wastes have already been finished at the end of 2017. Examples of the treatment procedures are followings;

- Phosphoric acid solution
  The solution was used as a electrolyte of electrolytic decontamination experiments inside the hot cells, and it contained 3 mol/L of phosphoric acid, $7\times10^5$ Bq/mL of $^{137}Cs$, $4\times10^5$ Bq/mL of $^{239}Pu$ + $^{240}Pu$ and $8\times10^5$ Bq/mL of $^{239}Pu$ + $^{241}Am$. Those solution was solidified by adding $Al^{3+}$, and then disposed as high level solid wastes.
- Lactic acid
  This solution was used as reductant of Pu in a modified PUREX process experiment conducted in a hot cell, and it contained 2 mol/L lactic acid, $1\times10^3$ Bq/mL of $^{137}Cs$, $2\times10^4$ Bq/mL of $^{239}Pu$ + $^{240}Pu$ and $1\times10^5$ Bq/mL of $^{239}Pu$ + $^{241}Am$. All lactic acid was oxidatively decomposed using Fenton reaction to be $CO_2$, acetic acid and formic acid without releasing hydrogen gas. The residual solution was sent to one of the liquid waste tank.
- Solution containing Chloride ions (Tada et al., 2017)
  The solution was generated by series of experiments on pyrochemical process and by analysis on contaminated water containing sea water samples at the Fukushima site. So far, chloride ions have not been aggressively treated except in the glove boxes for pyroprocess due to their corrosiveness. The waste solution of the pyrochemical experiments contained totally 70 g of U and 12 g of Pu. Those elements were recovered through solvent extraction with PUREX solvent after exchanging anion from chloride to nitrate by adding $Ag(NO_3)$ for AgCl precipitation formation. The precipitation was disposed as the solid wastes, and U and Pu are stored as oxide form. Chloride ions in the analytical samples of the Fukushima were treated with the same manner with the previous one.

Appropriate treatment procedures of those solutions have been experimentally examined by inactive experiments in advance with operations on the genuine waste solutions. The residual solutions will be treated based on small scale inactive and active demonstrations whose procedures will be developed through the collaborative researches. In our current schedule, treatments of all those solutions in CPF will be completely finished by the end of FY2020.

**Current activities of the collaborative studies; STRAD project**

The collaborative studies were combined to be a one project at the beginning of 2018 in order to share experiences and knowledges between collaborators. The purpose of this project is contributions not only to the developments for waste solution treatment in CPF but also to waste management or decommissioning of other nuclear facilities. The project was named as Systematical Treatment of Radioactive waste solution for Decommissioning (STRAD), and targets of study and collaborators are currently increasing to address forthcoming decommissioning of various facilities treating radioactive nuclides. Current representative studies in this project are decomposition of reactive chemicals in analytical waste liquids, recovery of nuclear materials from organic solvents and solidification of waste liquids to confine reactive compounds inside the solid. Some achievements of these studies will be shown in the presentation. The project is expected to produce beneficial waste management database which can be referred world widely.

## References

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