14-3 New Reprocessing Method without Organic Reagent
－Development of Uranium-Plutonium Cocrystallization Process－

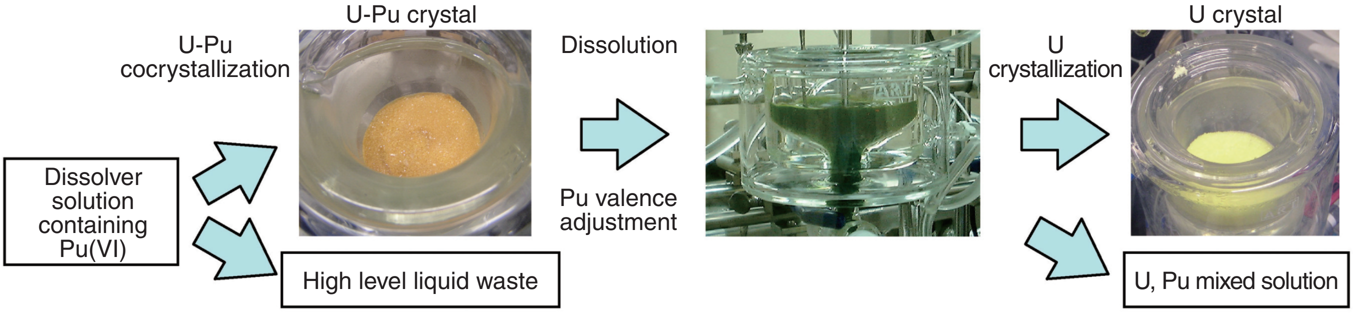

Fig.14-6　Steps in U-Pu cocrystallization reprocessing
The spent fuel dissolver solution is heated for Pu oxidation, and cooled for U-Pu cocrystallization. The U-Pu crystal (the color is orange) and mother liquor are separated by filtration. The crystal is dissolved in nitric acid solution, and Pu in the solution is reduced to Pu(IV). The solution is cooled again, resulting in U crystal (the color is yellow) and U-Pu mixed solution.

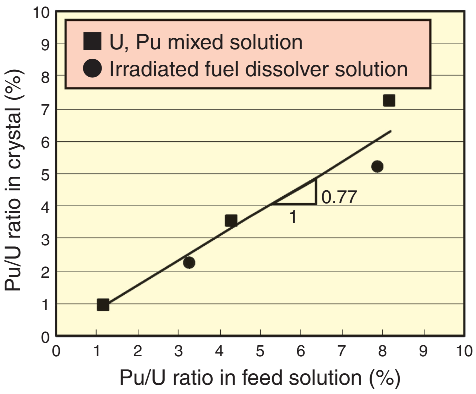

Fig.14-7　Comparison between Pu/U ratio in feed solution and Pu/U ratio in crystal
A good correlation with a linear coefficient of 0.77 is obtained. This means that the yields of Pu were lower than those of U.

Crystallization is considered to be one of the most promising methods for recovering uranium (U) and/or plutonium (Pu) in next-generation reprocessing. The principle of this method is based on the different temperature dependency of U and Pu solubility, which means no special reagent or extractant is required. Using U and Pu mixed solutions obtained by dissolution of irradiated fuel from the experimental fast reactor “JOYO” or unirradiated MOX (mixed oxide of U and Pu), fundamental experiments have been conducted in the hot laboratory named CPF (Chemical Processing Facility).

Through experiments, it has been revealed that hexavalent Pu (Pu(VI)) was co-crystallized with U even though Pu concentration was below its solubility limit. Taking advantage of this phenomenon, an innovative reprocessing process relying on crystallization alone was created (Fig.14-6), which features are;

- Pure Pu cannot be recovered.
- U/Pu ratio in the U, Pu mixed product is controllable by means of only U recovery posterior to the co-crystallization.
- Organic reagents are not used

The mechanism of U/Pu co-crystallization is considered to be similar to co-precipitation where the target element reacts with a carrier compound and a reagent additive. Usually, the carrier has a crystalline structure similar to that of the target element. The nitrate hexahydrates of U and Pu have the same type of crystal structure.

In the U-Pu co-crystallization, Pu is the target element and U acts as the carrier. U crystal is produced by temperature dropping instead of reagent addition, and Pu precipitates together with it. Conducting experiments using solutions with several kinds of U/Pu ratio, it has been confirmed that the U and Pu were co-crystallized under any conditions. It has also become clear that the U/Pu ratio in the crystal is lower than that of the feed solution (Fig.14-7).

In order to commercialize this system, methods for improvement of U/Pu co-crystallization yield and purification of resulting U and Pu must be developed.

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Reference

Shibata, A. et al., Experimental Study on U-Pu Cocrystallization Reprocessing Process, Journal of Nuclear Science and Technology, vol.46, no.2, 2009, p.204-209.