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Lab-scale implementation of a next-generation uranium and plutonium separation and purification process from spent nuclear fuel using monoamide solvent

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1. Introduction

In the back-end spent nuclear fuel cycle, the PUREX (Plutonium URanium EXtraction) process is industrially implemented in the AREVA NC plant at La Hague for UOX spent fuel reprocessing and recycling. Uranium and plutonium are purified by this process thanks to several liquid/liquid extraction steps. TBP (tri-n-butyl phosphate) is the extractant and uranium/plutonium separation is achieved by plutonium reduction which involves reductive reagents. In the framework of the development of Generation IV reactors and the treatment of fuels with increasing Pu content, applying the same chemical process would mean a quite large increase of reductive reagent consumption as well as more complex industrial separation workshops.

For these reasons, the French Alternative Energies and Atomic Energy Commission (CEA) developed new extraction systems such as monoamide solvent. They exhibit good potentialities to replace TBP thanks to their stability towards radiolysis and hydrolysis as well as their high selectivity with respect to fission products [1-2]. Moreover, the uranium/plutonium separation can be easily performed by a decreasing in nitric acid concentration without using a reductive reagent [2]. In this work, a mixture of N-N-di-(2-ethyl-hexyl)-butanamide (DEHBA) and N-N-di-(2-ethyl-hexyl)-isobutyramide (DEHiBA) was tested in order to demonstrate uranium and plutonium separation and purification from a genuine spent nuclear fuel [3-4].

2. Experimental, results and discussion

The experimental demonstration was carried out in the high-level shielded process line called CBP in the ATALANTE research facility of CEA Marcoule (France). In a first step, the genuine feed solution was obtained through the dissolution of spent UOX and MOX nuclear fuel in nitric acid and then clarification of this solution by cross-flow filtration. In a second step, thanks to data obtained with prior tests, a flowsheet was designed by the PAREX simulation code [5]. Several PMMA mixer-settlers were set up in the shielded experimentation facility. In addition to monitoring of process parameters such as flow rates, stirring speeds or temperatures, on-line UV-visible spectrophotometric measurements were implemented in relevant mixer settler to follow *in situ* uranium and plutonium concentrations. Samples were also analyzed by the shielded analysis line called CBA which provides real-time support and characterizations of the produced outflows. This experiment showed the quantitative recovery of uranium and plutonium without redox reactions at the back-extraction step and a good decontamination of these main end-product outflows (U, Pu) towards α and $\beta\gamma$ emitters (mainly ⁹⁹Tc, ¹⁰⁶Ru and ¹³⁷Cs).

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Key words: Uranium-plutonium separation, liquid-liquid extraction, monoamide, spent nuclear fuel