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Uranium and plutonium extraction by *N,N*-dialkylamides using multistage mixer-settler extractors

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Abstract

N,N-Dialkylamides (monoamides) are known as extractants for U and Pu, and many studies have been carried out mainly by single-stage batch method. We have focused on two monoamides: *N,N*-di(2-ethylhexyl)-2,2-dimethylpropanamide (DEHDMPA) and *N,N*-di(2-ethylhexyl)butanamide (DEHBA), and proposed a multistage extraction process for recovering U and Pu by these monoamides. A continuous counter-current experiment was carried out to demonstrate the validity of this process. This process consisted of two cycles, and the 1st cycle and the 2nd cycle employed DEHDMPA and DEHBA as extractants, respectively. The feed solution for the 1st cycle was 5.1 mol/dm^3 (M) nitric acid containing 0.92 M U, 1.6 mM Pu, and 0.6 mM Np. The raffinate collected in the 1st cycle was used as the feed for the 2nd cycle. The ratios of U recovered in the U fraction and U-Pu fraction were 99.1% and 0.8%, respectively, and the ratios of U in the used solvents were <0.04%. The ratio of Pu recovered in the U-Pu fraction was 99.7%, and the ratio of Pu in the used solvents was in the order of 10^{-3}–10^{-4}%. The concentration ratio of U with respect to Pu in the U-Pu fraction was 9, and this indicated that Pu was not isolated. The decontamination factor of U with respect to Pu in the U fraction was obtained as 4.5×10^5. These results supported the validity of the proposed process.

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Keywords: Monoamide; *N,N*-Dialkylamide; Uranium; Plutonium; Reprocessing; Extraction; Mixer-settler

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1. Introduction

The recovery of U and Pu from spent nuclear fuel is one of important tasks for recycle use of nuclear materials, and many methods have been investigated. Among these methods, commercially operating reprocessing plants employed a hydrometallurgical method known as PUREX process. PUREX process uses tri-*n*-butyl phosphate (TBP) as the extractant for U and Pu. TBP is an excellent extractant in that it has high distribution ratios for U(VI) and Pu(IV), and has a high loading capacity for U(VI). However, TBP has a few drawbacks. For example, the degradation products of TBP, such as dibutyl phosphate and monobutyl phosphate, cause the formation of a third phase and/or precipitation. Moreover, TBP cannot be decomposed into gases by incineration in that TBP contains phosphorus in its molecular structure, and this leads to increase the volume of secondary waste.

We have focused on *N,N*-dialkylamides (monoamides) to overcome the drawbacks mentioned above. Monoamides are known as extractants for U and Pu from nitric acid media, and as stable as TBP against radiolysis. Since monoamides consist of C, H, O, and N elements, they can be decomposed into gases by incineration. Furthermore, monoamides can be diluted using hydrocarbons, such as *n*-dodecane, which is used as a diluent for TBP in reprocessing plants, and thus monoamides would be applied to the existing extraction and separation devices. The specific features of monoamides mentioned above make them promising candidates as extractants for reprocessing by hydrometallurgical method, and many investigations on monoamides have been carried out. The use of monoamides has already been proposed in a few processes such as ARTIST (Amide-based Radio-resources Treatment with Interim Storage Transmutation) developed in Japan[1] and GANEX (Group ActiNide EXtraction) developed in France.[2] Moreover, U extraction by a monoamide using centrifugal contractors has already been carried out.[3]

Most of the former studies on monoamides have been performed by single-stage batch method, and they reported distribution ratios of metal elements such as U, Pu, and fission products. The data obtained by the single-stage experiments have contributed to fundamental understanding of monoamides. Although studies on the extraction properties of metal elements by monoamides using multistage systems have also been performed, limited number of literatures are available.[2-9] Since commercially operating reprocessing plants employ multistage devices such as pulsed columns and mixer-setter extractors, multistage experiments are necessary to evaluate practical application of monoamides. In the present study, therefore, we carried out a continuous counter-current experiment using monoamides, and the applicability of the monoamides was evaluated.

2. Experimental

2.1. Continuous counter-current experiment

A continuous counter-current experiment comprising two cycles was performed at room temperature (ca. 293 K) using mixer-settler extractors. Each mixer-settler has 16 stages, and the volumes of the mixing and the settling parts in each stage were 6 cm^3 and 17 cm^3, respectively. Two types of monoamides, *i.e. N,N*-di(2-ethylhexyl)-2,2-dimethylpropanamide (DEHDMPA, Fig. 1. (a)) and *N,N*-di(2-ethylhexyl)butanamide (DEHBA, Fig. 1. (b)) were used as extractants.

Figure 2. shows the experimental conditions for the 1st cycle. The 1st cycle employed two mixer-settlers labeled Bank 1 and Bank 2. Bank 1 consisted of 4-stage U extraction step and 12-stage Scrub-1 step, and Bank 2 consisted of 10-stage U back-extraction step. The feed solution fed to the 4th stage was 5.1 M nitric acid containing U, Pu,

(a) (b)

Fig. 1. (a) Molecular structure of *N,N*-di(2-ethylhexyl)-2,2-dimethylpropanamide (DEHDMPA); (b) Molecular structure of *N,N*-di(2-ethylhexyl)butanamide (DEHBA).