

Filtration Characteristics of Re-TPPCL and Cs-NaTPB Precipitates Solids Generated in a Uranium Recovery Process from a Simulated Spent Nuclear Fuel by Using an Alkaline Solution

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1. Introduction

Most of the processes for a back-end fuel cycle being currently studied or developed as advanced nuclear fuel cycles should have a high proliferation-resistance and environmental friendliness in order for nuclear energy to be a viable energy source in the future [1]. Recently, new concepts of inputting a precipitation or crystallization unit process into the advanced aqueous fuel cycle systems or dissolving spent fuel in an alkaline carbonate solution instead of a nitric acid solution have begun to be studied to enhance the safety, economic competitiveness, and to minimize the production of a secondary waste stream (Fig. 1). In order to develop a uranium recovery process from the spent nuclear fuel by using a high alkaline carbonate solution, it is very important to clarify various kinds of solids formation in a uranium recovery process.

In this study, the filtration characteristics of Re-TPPCL and Cs-NaTPB precipitates were experimentally examined and discussed regarding the filterability of the precipitates.

2. Experimental

2.1 Chemicals and chemical composition of simulated experimental solution

Chemicals of an analytical grade were used without further purification. The compositions selected for simulated solutions were based on the information from spent fuel (light water reactor fuel irradiated to 33,000 MWd/MTU and cooled for 5 years) and the ORIGEN computer code. Rhenium was substituted for technetium, because their chemical behavior was very close in solution and non-radio isotope of technetium does not exist.

2.1 Preparation of precipitates of Re and Cs

The procedure for the precipitation of Rhenium (Re) with TPPCL (tetraphenyl phosphonium chloride) and Cesium (Cs) with NaTPB (sodium tetraphenyl borate) were the same. For Re-TPPCL precipitation, 0.0017M Re_2O_7 in 0.5M Na_2CO_3 - 0.5M H_2O_2 solution was mixed with the same volume of 0.0017M TPPCL precipitant in a 0.5M Na_2CO_3 solution and under a constant stirring for 30 minutes at room temperature (20-25 °C). For Cs-NaTPB precipitation, the procedure for the precipitation was the same that of Re-TPPCL precipitation, only different thing was the concentration of Cs_2CO_3 and NaTPB as 0.007M were used. All the experiments of the precipitation and filtration were

performed in a batch experiment. The precipitates were separated by filtration and dried in oven for 72 hours at 50 °C

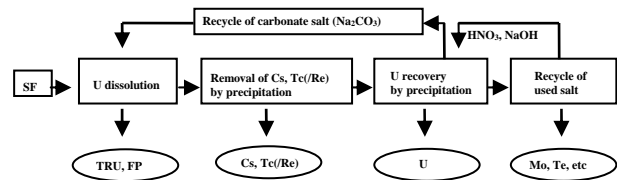


Fig. 1. Schematic flow of a conceptual process using a high pH carbonate media diagram to recover U from spent nuclear fuel.

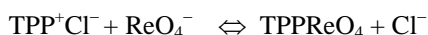
2.2 Experimental and Analytical Method

The filtration experiments were conducted in a laboratory filtration apparatus, equipped with lubricated rotary vane-type vacuum pump (Gast Co.), a 47mm all-glass vacuum filter holder and nitrocellulose membrane filter (Millipore Co., pore size = 1.2 μm). Particle size distribution and microscope image of the precipitates were determined by using a particle size analyzer (Malvern Ins., model, mastersizer/E) and an optical microscope (Leica dmlp), respectively. The precipitates were dried in a drying oven.

3. Results

3.1 Filtration characteristics of Re-TPPCL precipitate

In the process of Fig. 1, after leaching uranium from the spent fuel with the impurity elements of Cs, Tc, Te, and Mo, the Cs and Tc are to be removed from the solution before the dissolved uranium is recovered by a precipitation. Cs^+ and TeO_4^+ are well known to be precipitated with STPB and TPPCL, respectively. Fig. 2 shows the precipitation characteristics of the elements dissolved together with uranium in a carbonate solution of 0.5M Na_2CO_3 with 0.5M H_2O_2 by STPB and TPPCL. The STPB and TPPCL had only a selective precipitation for Cs and Re(Tc), respectively, with no selectivity to Mo and Te. Such precipitants were experimentally confirmed to have no selectivity to uranium in the solution as well. From these results, Cs and Tc, which are detrimental fission product elements to the environment, could be simply removed from the carbonate solution containing the peroxy-carbonato uranyl complex ions before the following step of the uranium precipitation by a pH-control of the solution. The precipitation reaction of Re-TPPCL in an aqueous solution is expressed as the following reaction.



The shape of TPPReO₄ was a needle-shaped and the mean particle size was 30 μm.

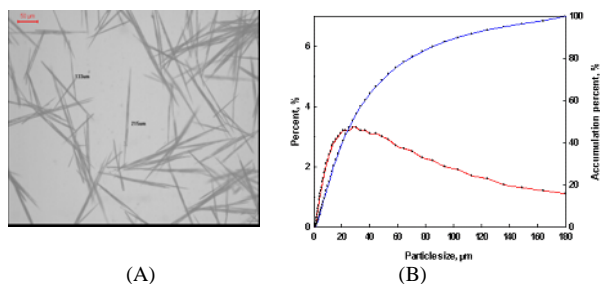
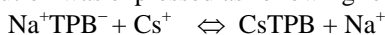


Fig. 2. (A) Microscope image of TPPReO₄, and (B) Particle size distribution of TPPReO₄

3.2 Filtration characteristics of Cs-NaTPB precipitate

The precipitation reaction of Cs-NaTPB in aqueous solution was expressed as following reaction.



The shape of Cs-TPB was a globular-shaped and the mean particle size was 15 μm (Fig. 3).

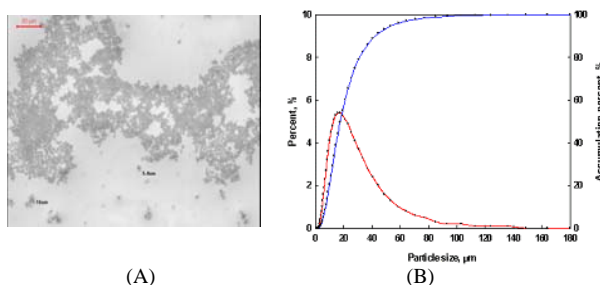


Fig. 3. (A) Microscope image of CsTPB and (B) Particle size distribution of CsTPB

3.3 Filterability of of Re-TPPCL and Cs-NaTPB precipitate

The filterability of the TPPReO₄ and CsTPB precipitates formed during the removal of Tc(Re) and Cs ions by the precipitation techniques using the TPPCl and NaTPB, precipitants were experimentally examined. The particle size distribution of the precipitate greatly affected the filtering characteristics of the precipitates. The mean particle size of the CsTPB precipitate is smaller than TPPReO₄ precipitate and the filterability of the TPPReO₄ precipitate was better than that of the CsTPB precipitate. Fortunately, both the filterability of the TPPReO₄ and CsTPB precipitate could be smoothly filtered in our experimental conditions. The specific resistance coefficient (α) of the TPPReO₄ and CsTPB precipitates in a constant pressure filtration condition were 1.27x10⁸ m/Kg and 4.42x10⁸ m/Kg, respectively (Fig. 4). When the specific resistance coefficient (α) of a precipitate was below 10¹¹ m/Kg, the filterability of the precipitate could be stated generally well (Fig. 5). We used the correlating filtration theory with industrial practice. The filtration rate a of precipitate could be expressed as the Ruth equation [2-3].

$$\frac{1}{A} \cdot \frac{dV}{dt} = \frac{P}{\mu(Rc + Rm)} = \frac{P}{\mu\{\alpha(W/A) + Rm\}}$$

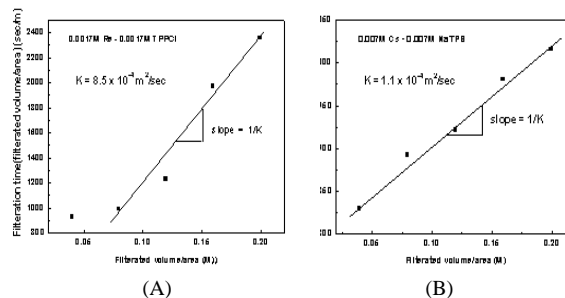


Fig. 4. The filtration coefficient of precipitates, (A) TPPReO₄ precipitates and (B) CsTPB precipitate

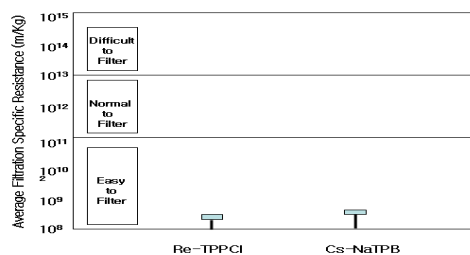


Fig. 5. Filterability of Re-TPPCL and Cs-NaTPB precipitate

4. Conclusions

Filtration characteristics of TPPReO₄ and CsTPB precipitates solids generated in a uranium recovery process from a simulated spent nuclear fuel by using an alkaline solution were examined in constant pressure filtration conditions. The shape of TPPReO₄ was a needle-shaped and the mean particle size was 30 μm. The shape of CsTPB was a globular-shaped and the mean particle size was 15 μm. The filterability of the TPPReO₄ and CsTPB precipitate could be smoothly filtered in our experimental conditions. The resistance coefficient (α) of the TPPReO₄ and CsTPB precipitates at a constant pressure filtration were 1.27x10⁸ m/Kg and 4.42x10⁸ m/Kg, respectively. Based on the results of this study, the filterability of the TPPReO₄ and CsTPB precipitates could be stated generally well, because the specific resistance coefficient (α) of a precipitate was below 10¹¹ m/Kg [3].

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