Synergic effect for uranium extraction from soil by TBOD and co-solvent using Sc-CO_2

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1. Introduction

The extraction process of uranium is one of the most important fields from the uranium mining to the spent fuel reprocessing. However, conventional organic solvent extraction produces a lot of liquid waste. The SFE (Supercritical fluid extraction) is extraction by substances occurring at temperatures and pressures above critical. The agent most widely used for SFE is carbon dioxide, which is nontoxic, readily available, inexpensive and mild critical parameters (temperature above 31 ℃ and pressure above 73 bar). The Sc-CO_2 (Supercritical carbon dioxide) may be a good extraction solvent due to the ease with which it can be recycled, and the fact it leaves negligible amounts of secondary wastes. But Sc-CO_2 do not extract metallic contaminants because it has the non-polar property. So, in order to extract the radioactive metal from radioactive wastes, Sc-CO_2 needs chelating ligands or surfactants such as organophosphorus[1,2], crown ethers[3], β-diketone[4-6], or dialkyldithiocarbamates[7]. The TBOD has some advantages: (ⅰ) high irradiation stability (ⅱ) strong affinity to metallic ions (ⅲ) completely incinerated without secondary solid waste. At this work, we have to extract the uranium metal from contaminated soil which adsorbed non-radioactive uranium using TBOD in Sc-CO_2.

2. Methods and Results

2.1 Chemicals, mock-up soil specimen

Carbon dioxide was purchased from Air Tech Co. The uranium metal ion which has AA standard solution, dibutylamine, triethylmine, diglycolyl amine, tributyl phosphate (TBP), hexafluoroacetylacetone (HFA) and pyridine were purchased from Aldrich Co. Isopropyl alcohol (IPA) and buthanol were used reagent grade. According to the known method[8-10], the TBOD and proline were synthesized in a laboratory scale. The chemical structure of TBOD was shown as Fig. 1.

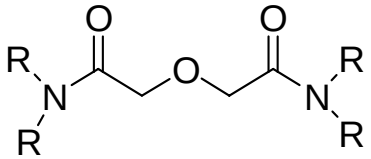


Fig. 1. Chemical structure of TBOD (R=$(CH_2)_3CH_3$)

The mock-up soil specimen which adsorbed non-radioactive uranium was prepared shown as Fig. 2. The soil was separated below 2 mm size by sieve. The organic compound which adsorbed to soil was removed from soil by 30 % hydrogen peroxide. The metallic compound which adsorbed to soil was removed from soil by 6 M nitric acid. The uranium metal was adsorbed to soil by rotary evaporator under 60 torr at 70 ℃.

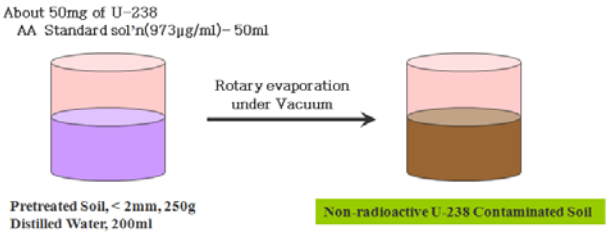


Fig. 2. Preparation of mock-up soil specimen

2.2 Equipments

The experimental equipments of supercritical fluid extraction was manufactured shown as Fig. 3. It composed of a high pressure syringe pump and a preheating coil, a premixing cell and an extraction cell in conventional oven with a thermo-controller and a separator. The ranges of pressure and temperature were up to 250 bar and 80 ℃. The extraction column of ID 10 mm × IL 200 mm and the premixing cell of 10 mL which has sapphire window were used. The carbon dioxide was pressurized by high pressure syringe pump.

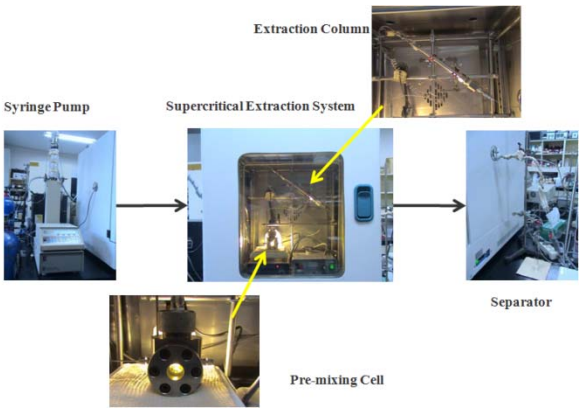


Fig. 3. The Sc-CO_2 extraction equipments

2.3 Experiments

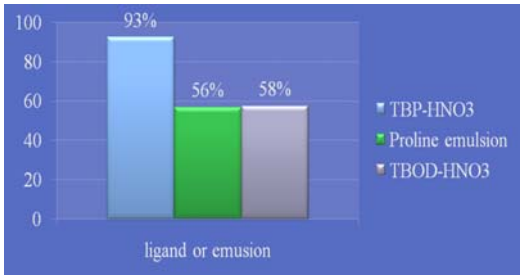
The mock-up soil specimen, 10 g, which adsorbed non-radioactive uranium metal (200 ppm) was replaced in extraction column. At the first experiments, we

extracted uranium by only chelating ligand under Sc-CO_2. The chelating ligands (TBOD, TBP, HFA, Pyridine) and/or nitric acid were replaced in premixing cell with magnetic stirrer. At the second experiments, we extracted uranium by TBOD and co-solvent under Sc-CO_2. The chelating ligand (TBOD-nitrate complex) and co-solvent (IPA or buthanol) were replaced in premixing cell with magnetic stirrer. And then, the carbon dioxide was introduced to experimental pressure as 200 bar after increased to experimental temperature as 40 ℃. After 20 min, the carbon dioxide was introduced continuously as 200 bar at 40 ℃ for 40 min. The uranium metal concentration in soil was analyzed by ICP-AES.

2.4 Results and discussion

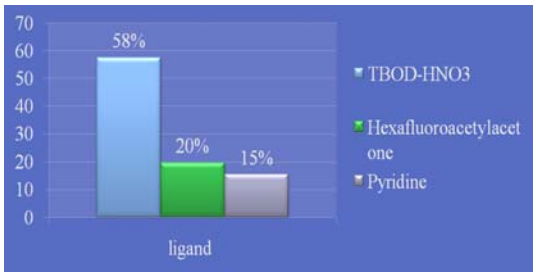
The experimental results of TBP, TBOD, Proline using Sc-CO_2 were summarized at Fig. 4. The extraction efficiencies using TBOD in Sc-CO_2 were lower than that using TBP in Sc-CO_2.

Fig. 4. Extraction efficiency of uranium from mock-up soil by TBP, Proline, TBOD in Sc-CO_2



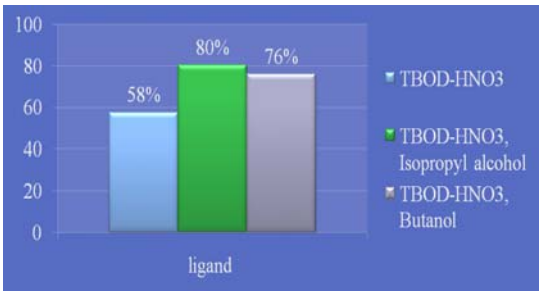
The experimental results of TBOD, HFA, Pyridine using Sc-CO_2 were summarized at Fig. 5. The extraction efficiencies using TBOD in Sc-CO_2 were higher than that using TBP in Sc-CO_2.

Fig. 5. Extraction efficiency of uranium from mock-up soil by TBOD, HFA, Pyridine in Sc-CO_2



The experimental results of TBOD-nitrate complex with IPA or buthanol as co-solvent using Sc-CO_2 were summarized at Fig. 6. The extraction efficiencies using TBOD and co-solvent in Sc-CO_2 were higher than that using only TBOD in Sc-CO_2.

Fig. 6. Extraction efficiency of uranium from mock-up soil by TBOD-nitrate complex with co-solvent in Sc-CO_2



3. Conclusions

The extraction efficiencies using TBOD in Sc-CO_2 were lower than that using TBP in Sc-CO_2. But the TBOD is more environmental ligand than TBP. The extraction efficiencies using TBOD and co-solvent in Sc-CO_2 were higher than that using only TBOD in Sc-CO_2 .

Acknowledgment

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