A Study on Chemical Separation of Pm-147 From RI Wastes

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1. Introduction

Promethium-147 is a rare-earth group nuclide which is not found in nature. And all types of its isotopes have radioactive characteristics. Pm-147 has a 2.62 y half-life. Pm-147 has a great power density, low hazard and possesses several attractive properties [1].

A lot of methods of the separation skills of Pm-147 have been investigated in the past. The first quantitative analysis of Pm-147 was conducted at ORNL (Oak Ridge National Laboratory).

Separation process of radioactive wastes have been made by ion-exchange with lactate, citrate and α-HIBA (ammonium α-hydroxyisobutyrate) as eluents. Another research reviewed large number of eluting agents for the separation of lanthanides from each other. However, above methods have to use large amount of eluents which cost are very expensive. Furthermore, the chemical separation process is quite complicated [2].

The object of this study is to develop a chemical process for the separation of Pm-147 radionuclide from RI wastes.

2. Experimental Section

2.1 Reagents

All the chemicals were purchased from commercial suppliers with analytical reagent grade. The Neodymium standard solution with 10,000 mg/L was purchased from Accustandard, USA. Nitric acid (65%), ammonia solution (25%), ethanol (99%) and acetone (99%) were purchased from MERCK (Germany). The oxalic acid was purchased from Aldrich Chem. Co, USA.

2.2 Equipment

A radiation shielded ICP-AES system (Thermo Jarrell Ash, USA) composed of an ICP-AES and a stainless steel glove box was employed for the analysis of the radioactive waste samples. Doubly deionized water was prepared by passing demineralized water through a Milli-Q Plus Ultra Pure Water System (Milipore).

2.3 Chemical separation

2.0 mL of Nd standard solution was pipetted to a 50 mL plastic beaker, the mixture solution was evaporated to incipient dryness on a hot plate with IR lamp. The resulting residue was dissolved in 4 mL of 9 M HCl with hand-shake and transferred to a 50 mL centrifuge tube. Cool to room temperature and 15 mL of saturated oxalic acid were added with stirring for 1 h. After adjusting its pH to 3.5-4.0 with 10% NH_4OH, it was centrifuged at 5,000 rpm for 5 min.[3] The supernatant was discarded and sequentially washing the precipitate with oxalic acid-based cleaning solutions which were used 9 mL in 3 mL increments. The washed precipitate was transferred to a 15 mL of alumina crucible and evaporated on a hot plate with 70°C for 2 h. To acquire the crystalline Nd_2O_3 phase, the dried precipitate was annealed at 900°C for 1 h.

3. Results and Discussion

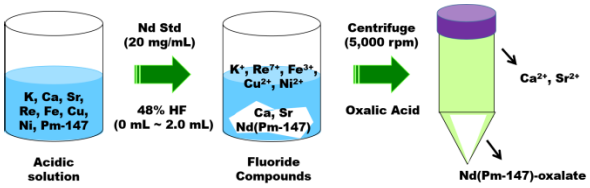

Fig. 1. Schematic illustration of Pm-147 chemical separation process.

A separation process of Pm-147 is shown in Fig. 1, and it is based on the wet chemistry. When the oxalic acid as a complexing reagent was added to the acidic solution, Nd-oxalate complex was spontaneously generated. Nd precipitates with oxalic acid at pH 3.5-4.0 in an oxalate medium and can be separated from some matrix elements that form insoluble metal-oxalate complex. After collecting the precipitates by using a centrifuge at 5,000 rpm for 5 min, thermal treatment was conducted for removal of moisture in the solidified products.

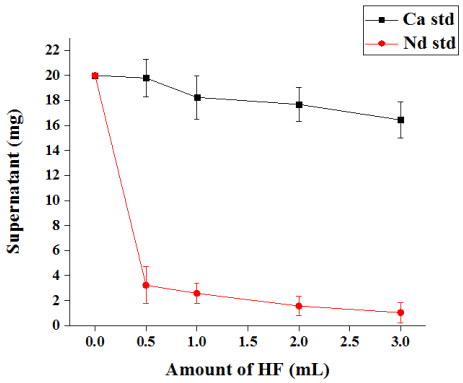

Fig. 2. Chemical behavior of Nd and Ca nuclides from radioactive waste sample solutions depending on the amount of HF (0 mL, 0.5 mL, 1.0 mL, 1.5 mL and 2.0 mL), which were measured with ICP-AES.

The relation between the precipitation behavior and the amount of HF is summarized in Fig. 2. When the HF was increased further from 0 mL to 2.0 mL, Nd nuclide was fully precipitated. On the other hand, Ca nuclide was not precipitated despite of its high Ksp values as well as high chemical affinity of Nd element with oxalic acid at pH 3.5-4.0. This result indicates that Ksp value of Nd was higher than Ca, which leads to precipitation of Nd as a fluoride compound.

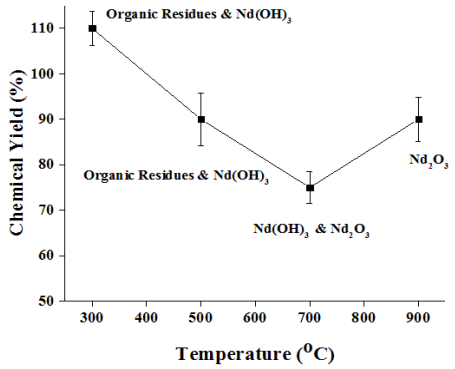

Fig. 3. Recovery rate for Pm-147 with an oxide compound (Nd_2O_3) at different temperatures, (a) at 300°C, (b) 500°C, (c) 700°C, (d) 900°C for 1h, respectively.

4. Conclusion

We developed the selective chemical separation process of Pm-147 from the radionuclides which include a variety of elements. As the chemical behaviors of Pm and Nd both are similar, Nd was used as a tracer of Pm. The formation of Pm-oxalate complex under acidic conditions by adding oxalic acid can be possible to separate unwanted radionuclides with low Ksp values. Based on the above results, the recovery yields of samples were measured with reliability.

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