

Application of Micro-coprecipitation Method to Alpha Source Preparation for Measuring Alpha Nuclides

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1. Introduction

An alpha source which has a remarkable energy resolution is required to accurately analyze such emitters of these actinide nuclides. Sources for alpha spectrometric measurements have usually been prepared by a vacuum sublimation, a direct evaporation, an electrodeposition, and a micro-coprecipitation [1,2]. Among the source preparations, an electrodeposition is a commonly used method for the preparation of sources for an alpha spectrometry, because this technique is simple and produces a very thin deposit, which is essential for a high resolution of the alpha peak. Recently, micro-coprecipitations with rare earths have been used to yield sources for α -spectrometry, as these methods are simple and more rapid than an electrodeposition[3,4].

In this work, the Pu, Am and Cm isotopes were purified from hindrance nuclides and elements with an a TRU resin in radioactive waste samples, and the activity concentrations of the Pu, Am and Cm isotopes were determined by radiation counting methods after alpha source preparation like micro coprecipitation.

2. Methods

2.1 Pretreatment of the samples

A liquid waste sample (10 mL) leached from the radioactive sample with HNO_3 was evaporated to a dryness on a hot plate and the residue was dissolved in a concentrated nitric acid solution. To compensate for a chemical recovery, ^{242}Pu (0.1 Bq) and ^{243}Am (0.1 - 0.5 Bq), as yield tracers, were added into a sample. The residue was dissolved with 30 mL of 8 M HNO_3 .

2.2 Separation of Pu and Am with an anion exchange resin and TRU resin

The sample solution with an 8 M HNO_3 medium was passed through a pre-conditioned anion exchange resin (Bio-Rad, 100 - 200 mesh) column (inner diameter; 10 mm, resin bead length; 80 mm)

with 8 M HNO_3 at a rate of 0.5 mL/minute. The column was then washed with 20 mL of 8 M HNO_3 to remove the Am and U isotopes. The effluent was evaporated to a dryness and reserved for a subsequent separation of the Am isotopes. Columns were washed with 20 mL of 9 M HCl to desorb the Th. Finally, Pu was eluted with 20 mL of 0.36 M HCl / 0.01 M HF, which does not form a chloride complex on the anionic exchange resin.

The residue that was separated from the Pu purification step was dissolved in 30 mL of 2 M HNO_3 . The content of Fe(III) was tested by adding one drop of 1 M NH_4SCN to a sample solution. If the test was positive (red color), about 200 mg of ascorbic acid was added to reduce the Fe(III) to Fe(II). The TRU Spec column (bed volume; 1.3 mL, length; 2.6 cm) was used to separate Am from U, and it was conditioned with 30 mL of 2 M HNO_3 . Samples were loaded onto the column followed by a washing with 20 mL of 2 M HNO_3 . The column was then washed with 4 mL of 9 M HCl and the Am fraction was eluted with 20 mL of 4 M HCl.

2.3 Comparison of an electrodeposition and a micro-coprecipitation

To investigate the electrodeposition and micro-coprecipitation yields, ^{242}Pu tracer and ^{243}Am tracer (about 0.5 Bq) were added into the solution containing the ^{239}Pu standard source and ^{243}Am standard source, respectively. The solution containing the ^{242}Pu tracer was evaporated to a dryness. The residue was dissolved in 1 mL of concentrated HCl and evaporated to a dryness. The Pu fraction was electroplated on to a stainless steel platelet or micro-coprecipitated on to a 0.1 m membrane filter. The solution containing the ^{243}Am tracer was carried out similar to case of ^{242}Pu . The samples prepared by an electrodeposition or a micro-coprecipitation were measured by an alpha spectrometer.

2.4 Measurement of the Pu, Am and Cm isotopes with an alpha spectrometer

The activity concentrations of $^{239,240}\text{Pu}$, ^{238}Pu , ^{241}Am and ^{244}Cm were measured by an alpha spectrometer. The measured activities for $^{239,240}\text{Pu}$, ^{238}Pu , and ^{241}Pu were corrected for their chemical yield by using the observed activities of ^{242}Pu . Also, the chemical yields for ^{241}Am and ^{244}Cm were obtained by measuring the alpha activity of ^{243}Am , because the chemical property of the Cm isotopes was similar to that of the Am isotopes. After the α -spectrometric measurement for $^{239,240}\text{Pu}$ and ^{238}Pu , the Pu sample coprecipitated with Nd on the membrane filter was dissolved with 10 mL of 8 M HNO_3 . The dissolved solution was divided into two fractions, where one was used for measuring ^{241}Pu by a LSC and the other was used for measuring ^{239}Pu , ^{240}Pu and ^{241}Pu by a TIMS.

3. Results and discussion

Among the source preparations, such as a direct evaporation, an electrodeposition and a micro-coprecipitation, the electrodeposition method and the micro-coprecipitation method were compared in this work. The recovery ranges of ^{242}Pu with an electrodeposition and a micro-coprecipitation were 65 - 96 % and 87 - 99 %, respectively. The mean value of the chemical yield of ^{242}Pu for an electrodeposition and a micro-coprecipitation was 86 ± 5 % and 96 ± 6 %, respectively. Also, the recovery of ^{243}Am for an electrodeposition was found to be in the range of 60 % to 95 % with a mean value of 80 ± 7 %. While the recovery of ^{243}Am with a micro-coprecipitation was found to be in the range of 91 % to 99 % with a mean value of 97 ± 6 %.

For the electrodeposition method, the recoveries for the Pu and the Am isotopes were lower and variable, compared to the coprecipitation method which resulted in higher and more stable recoveries, though the α -peak resolution (FWHM : 26.7 keV) for the electrodeposition was better than that for the micro-coprecipitation (FWHM : 43.2 keV). Electrodeposition requires rather elaborate equipment which is difficult to maintain and is plagued with problems, such as current fluctuations and pH changes during an electrodeposition, that can result in a low isotope recovery.

Also, for measuring ^{241}Pu by a LSC and ^{239}Pu and ^{240}Pu by a TIMS, it is necessary to dissolve

the Pu isotopes from a stainless steel disc or membrane filter after measuring the Pu isotopes by an alpha spectrometry. In this process, a lot of Fe or Ni ions were dissolved with the Pu isotopes from an electroplating plate and these ions have to be removed from the Pu isotopes with an anion exchange resin method, because the measurement of ^{241}Pu , ^{239}Pu and ^{240}Pu can be interfered by these ions. However, in the micro-coprecipitation method, the Pu isotopes are measured directly by a LSC or a TIMS after dissolving the Pu isotopes from the membrane filter. Therefore, in this work, the micro-coprecipitation method was used for the alpha source preparation because this method is simple and more rapid than an electrodeposition.

4. Conclusion

After the Pu isotopes in the radioactive waste samples were separated from the other nuclides with an anion exchange resin, the Am isotopes were purified with a TRU resin and an anion exchange resin or a TRU resin. Activity concentrations and chemical recoveries of ^{241}Am purified with the TRU resin were similar to those with the TRU resin and anion exchange resin. In this study, to save on the analytical time and cost, the Am isotopes were purified with the TRU resin without using an additional anion exchange resin.

After comparing the electrodeposition method with the micro-coprecipitation method, the micro-coprecipitation method was used for the alpha source preparation, because the micro-coprecipitation method is simple and more reliable for source preparation of the Pu, Am and Cm isotopes.

5. Acknowledgement

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6. References

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