Determination of Neptunium, Americium and Curium in Spent Nuclear Fuel Samples by Alpha Spectrometry Using ^{239}Np and ^{243}Am as a Spike and a Tracer

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1. Introduction

Determination of actinide elements and fission products in spent nuclear fuels is of importance for a burnup determination and source term evaluation. Especially, the amounts of uranium and plutonium isotopes are used for the evaluation of a burnup credit in spent nuclear fuels. Additionally, other actinides such as Np, Am and Cm in spent nuclear fuel samples is also required for the purposes mentioned above. In this study, ^{237}Np, ^{241}Am and ^{244}Cm were determined by an alpha spectrometry for the source term data for high burnup spent nuclear fuels ranging from 37 to 62.9 GWD/MtU as a burnup.

Generally, mass spectrometry has been known as the most powerful method for isotope determinations such as high concentrations of uranium and plutonium. However, in the case of minor actinides such as Np, Am and Cm, alpha spectrometry would be recommended instead. Determination of the transuranic elements in spent nuclear fuel samples is different from that for environmental samples because the amount of each nuclide in the spent fuel samples is higher and the relative ratios between each nuclide are also different from those for environmental samples. So, it is important to select an appropriate tracer[1,2] and an optimum sample size depending on the nuclides and analytical method.

In this study ^{237}Np was determined by an isotope dilution alpha(gamma) spectrometry using ^{239}Np as a spike, and ^{241}Am and curium isotopes were determined by alpha spectrometry using ^{243}Am as a tracer[3]. The content of each nuclide was compared with that by the Origen-2 code[4].

2. Experimental

Spent nuclear fuel samples were dissolved with (1+1) HNO_3 in a hot cell and the solutions were diluted. An optimum sample size was estimated by calculating the amount of each nuclide according to burnup using Origen-2 code. An appropriate amount of sample was taken and loaded onto anion exchange column. The elements were separated by anion exchange[5] and/or HDEHP extraction chromatography[3]. A large sample size containing about 100 μg U was taken for ^{237}Np and a small sample size containing about 0.1 μg of U for Am and Cm. The Np was separated using 4 M HCl as an eluent after separation of Pu(Fig 1). Am and Cm were separated by HDEHP extraction chromatography after a group separation onto anion exchange column. The separated elements were determined by an alpha spectrometry after an electredeposition.

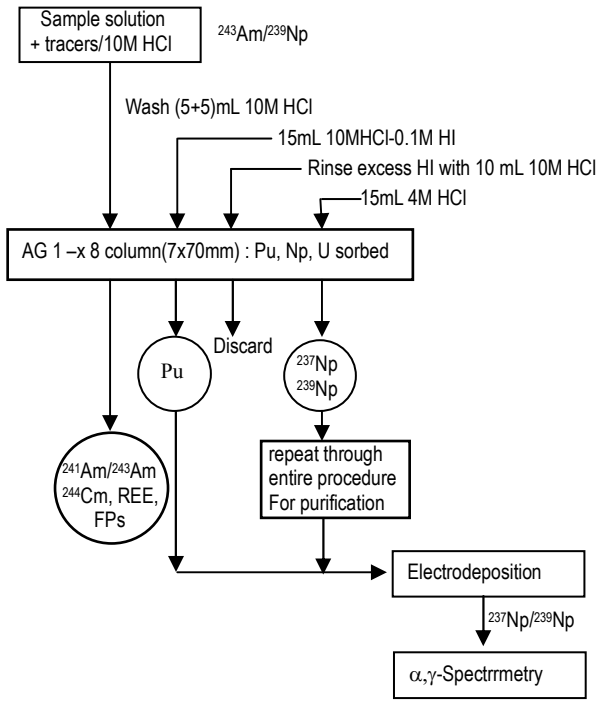

Fig 1. Sequential separation of transuranic elements in spent nuclear fuel sample solutions

3. Results and discussion

3.1 Determination of ^{237}Np

A spike addition method was adopted for the determination of ^{237}Np using ^{239}Np as a spike. The tracers such as ^{235}Np($t_{1/2}$, 396.1d) and ^{236}Np($t_{1/2}$, 5000y) were not available in our laboratory. So, ^{239}Np was used. The standard solution of ^{243}Am contains an equal amount of ^{239}Np because ^{243}Am decays to ^{239}Np by an alpha emission and reaches a secular equilibrium state after an equilibrium time of about 40 days, so an equal amount of ^{239}Np is produced. Two measurements for one sample were conducted by taking samples (“sample” and ”spiked sample”) equivalent to ~ 100 μ g U each. About 30 ~ 60 Bq of ^{239}Np(^{243}Am standard solution) was added into one sample(“a spiked sample”). The two samples were applied separately to each anion exchange column. The ^{239}Np and ^{237}Np in the “sample”

and "spiked sample" were measured by a gamma spectrometry and an alpha spectrometry(4.79 MeV), respectively, after a separation followed by an electrodeposition[6]. The alpha spectrum is shown in Fig 2. The content of ^{237}Np was measured by an isotope dilution alpha(gamma) spectrometry(IDAS) by using the ratios of ^{237}Np/^{239}Np in the "sample" and "spiked sample", respectively, and compared with that by calculation..

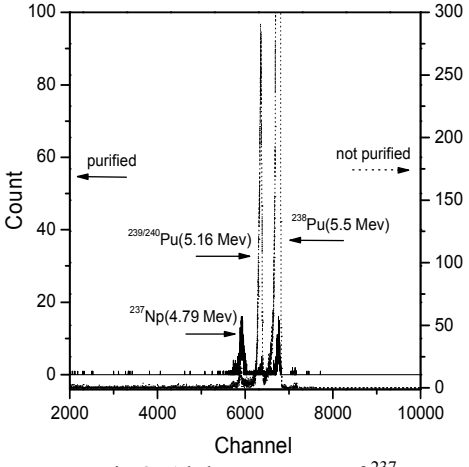

Fig 2. Alpha spectrum of ^{237}Np

3.2 Determination of Am and Cm

For the determination of Am and Cm, a small sample size equivalent to about 0.1 μg of U was taken after a considerable dilution of the sample solution. About 15 Bq of ^{243}Am as a tracer was added into the sample solution. ^{243}Am standard solution was directly used as a tracer without milking ^{239}Np from it.

An extraction column filled with the HDEHP adsorbant was preconditioned with 20 mL of 0.1 M HNO_3. The effluent containing Am and Cm not adsorbed onto the anion exchange column in the previous step of the anion exchange separation was dried on a hot plate and treated with nitric acid. The sample medium was transformed to nitrate salt. The sample solution was quantitatively transferred to the HDEHP column. The Am and Cm were eluted together with 6 mL of 0.05 M DTPA-0.5 M LA with successive additions of 3 mL each. The lanthanides as well as Mo and Zr are then eluted with 5 mL of 6 M HNO_3. But, this step was omitted in this work.

The separated nuclides were determined by an alpha spectrometry after an electrodeposition. The alpha activities of ^{241}Am, ^{244}Cm and ^{242}Cm were measured at 5.48 MeV, 5.81 Mev and 6.11 Mev, respectively(Fig 3). The measurements were also compared with those by calculations.

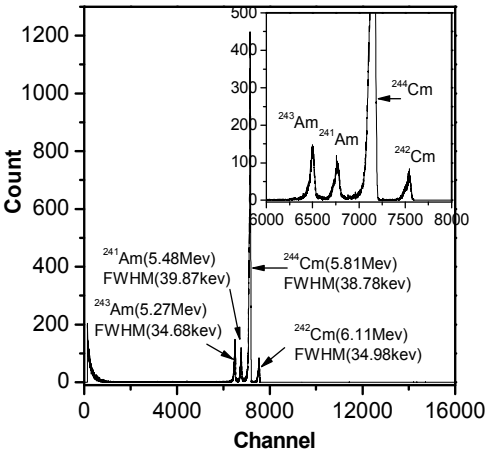

Fig 3. Alpha spectrum of ^{241}Am, ^{244}Cm and ^{242}Cm

3. Conclusion

Determination of ^{237}Np, ^{241}Am, ^{244}Cm and ^{242}Cm as source term data for spent nuclear fuels was successfully performed. A comparison between the measurement and the calculation showed that the measurement was lower than the calculation by 15% for ^{237}Np, while the measurements were higher than the calculations from about 40% to 90% for ^{241}Am, ^{244}Cm and ^{242}Cm. More data will be required for a code verification related to the spent nuclear fuels.

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