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Determination of Plutonium Isotopes in Spent Nuclear Fuel Samples by Alpha Spectrometry Using ^{242}Pu as a Tracer

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1. Introduction

Determination of actinide elements and fission products in spent nuclear fuels is of importance for burnup determination and source term evaluation. Especially, the amounts of uranium and plutonium isotopes are used for the evaluation of burnup credit in spent nuclear fuels. Accordingly, determination of actinide elements such as U, Pu, Np, Am and Cm in spent nuclear fuel samples is essentially required for the purposes mentioned above. In this study, the plutonium isotopes such as ^{238}Pu, ^{239}Pu and ^{240}Pu, were determined by alpha spectrometry as a first attempt toward actinides determinations for the source term data for high burnup spent nuclear fuels.

Generally, mass spectrometry has been known the most powerful method for isotope determinations. However, in the case of ^{238}Pu in uranium matrix amples a contamination effect caused by ^{238}U could not be avoided eventhough the plutonium is separated from the uranium matrix. In this case, alpha spectrometry would be recommended instead rather than mass spectrometry. Consequently, in this study alpha spectrometry was used for the determination of the plutonium isotopes in spent nuclear fuel samples using ^{242}Pu as a tracer.

Determination of the transuranic elements in spent nuclear fuel samples is quite 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 and an optimum sample size depending on the nuclides and analytical method. In this study, an optimum sample size was estimated by calculating the amount of each nuclide according to burnup using Origen2 code.

2. Experimental

A synthetic sample solution containing uranium and fission products was prepared. An aliquot of synthetic sample solution containing ~ 100 μg U was taken and appropriate amounts of ^{239}Pu, ^{237}Np and ^{241}Am were added into the synthetic sample solution. The recovery yields of each nuclide were determined by separation and measurement according to the procedures (Fig1).
In spent nuclear fuel samples the contents of each nuclide were estimated based on the Origen2 code in order to decide an optimum sample size .

A spent fuel sample containing about 0.1μ g of U was taken. The sample solution in nitric acid medium was dried on a hot plate after 1.54 Bq of ^{242}Pu as a tracer was added into the sample. Finally, the sample matrix was transformed to 10 M HCl solution. The sample solution was loaded onto the anion exchange column. The plutonium was separated by reducing Pu^{6+} to Pu^{3+} using 10M HCl-0.1M HI as an eluent.

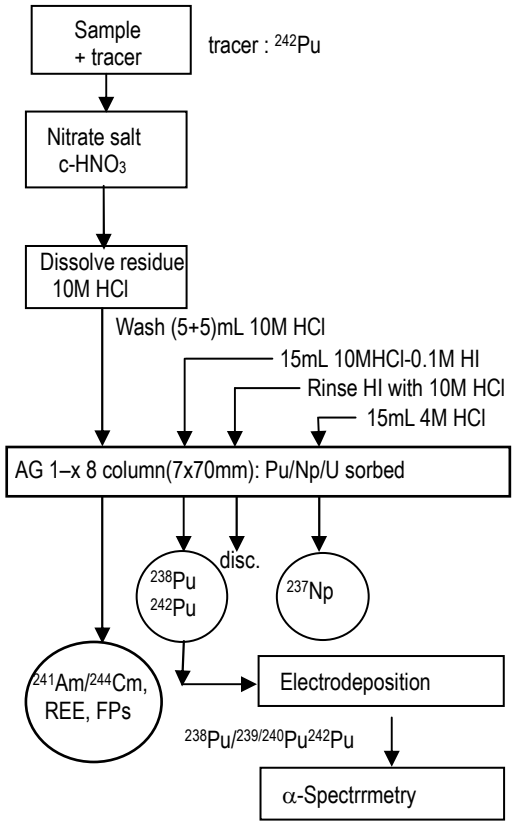

Fig 1. Sequential separation of transuranic elements in spent nuclear fuel sample solutions

The separated plutonium was electrodeposited in sodium sulfate electrolyte followed by alpha spectrometry. A total alpha activity was measured by a gross alpha counting and an individual activity of each nuclide was also measured by alpha spectrometry. The activity of the each nuclide was determined by applying the activity ratio of each nuclide to the total alpha activity. The alpha activities of plutonium isotopes were

measured at 5.5 MeV of ^{238}Pu, 5.16 MeV of ^{239,240}Pu and 4.9 MeV of ^{242}Pu, respectively. The recovery yield of plutonium isotopes was obtained by using ^{242}Pu as a tracer.

3. Results and discussion

Before going into a spent fuel sample an individual separation of the nuclides was checked with anion exchange column and HDEHP extraction column using ^{239}Pu, ^{237}Np and ^{241}Am. The ^{239}Pu were successfully separated from other nuclides. The recovery yield of ^{239}Pu was shown to be 82.3% in synthetic sample solution.

In Fig 2 the alpha spectrum of the plutonium isotopes in spent fuel sample was shown. The three peaks of ^{238}Pu, ^{239,240}Pu and ^{242}Pu were sharply resolved. A small peak of ^{244}Cm was also observed. This small peak of ^{244}Cm means that a large peak of ^{238}Pu(5.5 MeV) was not contaminated by the ^{241}Am(5.48 MeV) because the peak of ^{244}Cm is much higher than ^{241}Am in spent fuel sample. Total activity of ^{239}Pu and ^{240}Pu were measured because these nuclides were overlapped at 5.16 MeV. An individual activity of each nuclide was obtained by complementally using mass spectrometry and specific activities as well. The amounts of ^{238}Pu, ^{239}Pu and ^{240}Pu in the high burnup spent nuclear fuel samples were determined and compared with those by calculation using the Origen 2 code.

Determination of plutonium isotopes in spent nuclear fuel samples were successfully performed by alpha spectrometry using the ^{242}Pu as a tracer. In the future this approach will be extended to the determination of transuranic elements for a source term data for the high burnup spent nuclear fuels.

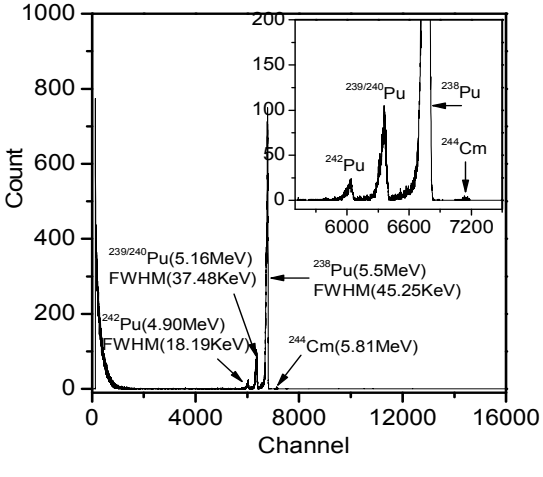

Fig 2 Alpha spectrum of plutonium isotopes in spent nuclear fuel sample

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