2–6 Determination of Plutonium-Isotope Ratios in Individual U/Pu Mixed Particles

— Novel Approach using Alpha and Mass Spectrometry for Nuclear Non-Proliferation —

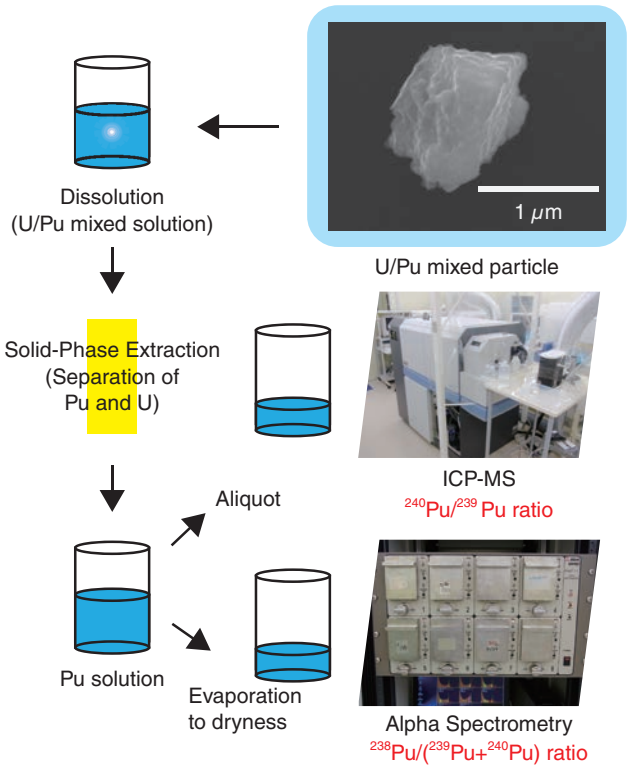

Fig.2-15 Analytical flow of Pu analysis for individual U/Pu mixed particles
A novel technique for the analysis of individual U/Pu mixed particles was developed by a combination of alpha and mass spectrometry.

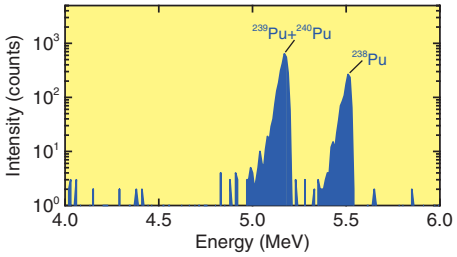

Fig.2-16 Alpha spectrum of a U/Pu mixed particle with a U/Pu ratio of 5 measured for 2000000 s
Well-separated peaks assigned to ^{238}Pu and ^{239}Pu+^{240}Pu were obtained in the spectrum.

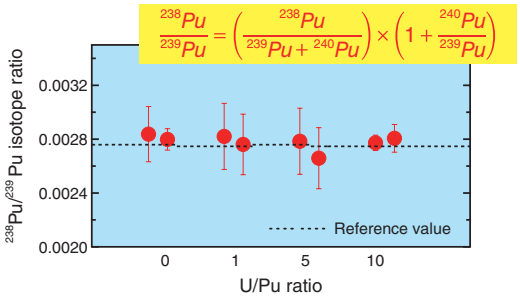

Fig.2-17 The ^{238}Pu/^{239}Pu isotope ratios measured for individual U/Pu mixed particles with U/Pu ratios of 0, 1, 5, and 10
The ^{238}Pu/^{239}Pu-isotope ratios determined with the proposed method were quite consistent with the reference values.

Plutonium (Pu) is a key element for nuclear-fuel cycles; it is produced by neutron capture by uranium (U) in nuclear reactors and is used as U/Pu mixed-oxide (MOX) fuel after purification. On the contrary, Pu with high ^{239}Pu abundances plays an important role in the production of nuclear weapons. Therefore, undeclared nuclear activities using Pu and U should be checked and prevented. We measure U- and Pu-isotope ratios in environmental samples taken at nuclear facilities from around the world to unveil undeclared nuclear activities in cooperation with the International Atomic Energy Agency (IAEA).

The main isotopes of Pu are ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. Although mass spectrometry is commonly used for isotope-ratio analysis, the determination of ^{238}Pu/^{239}Pu-isotope ratios in individual U/Pu mixed particles is impossible because the mass of ^{238}Pu is almost the same as that of ^{238}U. Therefore, there are no analytical techniques for measuring ^{238}Pu/^{239}Pu-isotope ratios in such particles. In the present study, we developed a novel technique using alpha spectrometry and inductively coupled plasma–mass spectrometry (ICP-MS) to determine Pu-isotope ratios including ^{238}Pu/^{239}Pu.

The analytical procedure developed in this study is shown in Fig.2-15. Each U/Pu particle was isolated using a glass needle under a scanning electron microscope and dissolved with acids. Pu in the sample solution was separated from U by solid-phase extraction. The ^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu, and ^{242}Pu/^{239}Pu-isotope ratios and the ^{238}Pu/(^{239}Pu+^{240}Pu) activity ratios in the Pu solution were determined with ICP-MS and alpha spectrometry, respectively. Finally, the ^{238}Pu/^{239}Pu-isotope ratios in each particle were calculated from the ^{240}Pu/^{239}Pu and ^{238}Pu/(^{239}Pu+^{240}Pu) ratios.

Fig.2-16 shows an alpha spectrum of Pu extracted from a U/Pu mixed particle with a U/Pu ratio of 5. Since well-separated peaks were obtained in this spectrum, the ^{238}Pu/(^{239}Pu+^{240}Pu)-activity ratio could be determined accurately. In addition, the ^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu, and ^{242}Pu/^{239}Pu isotope ratios were successfully determined with ICP-MS. Fig.2-17 shows the ^{238}Pu/^{239}Pu isotope ratios in individual U/Pu mixed particles with the U/Pu ratios of 0, 1, 5, and 10. These values calculated from the ^{240}Pu/^{239}Pu isotope ratios and the ^{238}Pu/(^{239}Pu+^{240}Pu) activity ratios were well consistent with reference values. These results indicate that the proposed method using alpha spectrometry and ICP-MS enables us to determine Pu-isotope ratios accurately for individual U/Pu mixed particles.

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Reference

Esaka, F. et al., Analysis of Plutonium Isotope Ratios including ^{238}Pu/^{239}Pu in Individual U-Pu Mixed Oxide Particles by Means of a Combination of Alpha Spectrometry and ICP-MS, Talanta, vol.165, 2017, p.122-127.