2–6 Deducing the Origin of Nuclear Materials by Analyzing Uranium Particles in Environmental Samples

— Determination of Chemical Forms and Isotope Ratios in Individual Uranium Particles for Nuclear Safeguards —

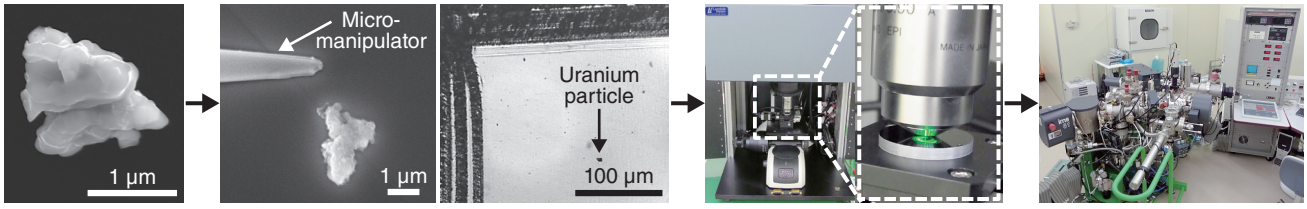

Fig.2-16 Analytical flow for individual uranium particles
A novel technique for the analysis of individual uranium particles of micrometer size was developed using a combination of micro-Raman spectroscopy and secondary ion mass spectrometry.

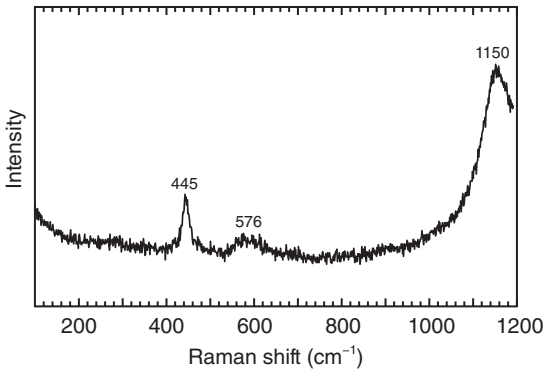

Fig.2-17 Raman spectrum of a uranium dioxide particle
Several Raman peaks assigned to the UO_2 structure were detected.

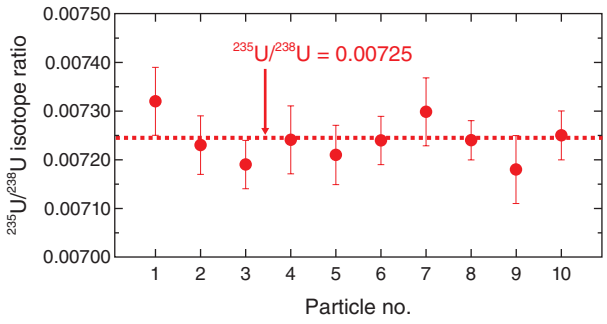

Fig.2-18 $^{235}U/^{238}U$ isotope ratios measured for individual uranium particles
The $^{235}U/^{238}U$ isotope ratios determined with the proposed method were consistent with that of natural uranium (indicated by a broken line).

Naturally occurring uranium consists of ^{234}U, ^{235}U, and ^{238}U isotopes. The uranium isotope ^{235}U is enriched to extract nuclear power effectively because it is the only fissile isotope of natural uranium. The abundance of ^{235}U is 0.72 % for natural uranium, 2 %–5 % for nuclear fuels for nuclear power plants, and over 90 % for the raw material for nuclear weapons. We routinely measure uranium isotope ratios in environmental samples, which are collected by International Atomic Energy Agency (IAEA) inspectors at nuclear facilities all over the world, to detect undeclared nuclear activities related to the production of nuclear weapons.

The nuclear fuel cycle is the progression of nuclear fuel through a series of processes. Various uranium compounds are used in the refining, conversion, enrichment, and fabrication processes, including triuranium octoxide in the refining process and uranium dioxide (UO_2) in the fabrication process. Investigation of the chemical states and isotope ratios of uranium particles may provide information on the origin of materials. However, there are few reports on the analytical techniques for obtaining both the chemical states and isotope ratios of individual uranium particles.

In this study, we developed a novel analytical procedure using micro-Raman spectroscopy (MRS) and secondary ion mass spectrometry (SIMS), as shown in Fig.2-16. Uranium particles on a substrate were identified by scanning electron microscopy and then transferred onto another substrate using a micro-manipulator. The chemical forms of uranium particles were analyzed by MRS, and the isotope ratios of the particles were determined by SIMS.

Fig.2-17 shows the Raman spectrum of a UO_2 particle in a natural uranium material. Characteristic Raman bands were observed at 445 cm^{-1}, 576 cm^{-1}, and 1150 cm^{-1}, which are the values assigned to the UO_2 structure. The uranium isotope ratios of individual UO_2 particles were determined by SIMS after the MRS measurement. Fig.2-18 shows the isotope ratios of ten individual UO_2 particles. The $^{235}U/^{238}U$ isotope ratios were highly consistent with that of natural uranium. We confirmed that the chemical states and isotope ratios of individual uranium particles could be analyzed by this proposed method.

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Reference

Yomogida, T. et al., Chemical State and Isotope Ratio Analysis of Individual Uranium Particles by a Combination of Micro-Raman Spectroscopy and Secondary Ion Mass Spectrometry, Analytical Methods, vol.9, issue 44, 2017, p.6261–6266.