Secondary ion mass spectrometry combined with alpha track detection for isotope abundance ratio analysis of individual uranium-bearing particles

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ABSTRACT

Secondary ion mass spectrometry (SIMS) was used in combination with alpha track detection for the efficient analysis of uranium-bearing particles with higher ^{235}U abundances in environmental samples. A polycarbonate film containing particles was prepared and placed in contact with a CR-39 plastic detector. After exposure for 28 days, the detector was etched in a NaOH solution and each uranium-bearing particle was identified through observation of the alpha tracks recorded in the detector. A portion of the film containing each uranium-bearing particle was cut out and put onto a glassy carbon planchet. The films on the planchet were decomposed through plasma ashing for subsequent uranium abundance ratio analysis with SIMS. The alpha track-SIMS analysis of 10 uranium-bearing particles in a sample taken from a nuclear facility enabled $n(^{235}\text{U})/n(^{238}\text{U})$ abundance ratios in the range 0.0072–0.25 to be detected, which were significantly higher than those obtained by SIMS without alpha track detection. The duration of the whole analytical process for analysis of 10 particles was about 32 days. The detection efficiency was calculated to be $27.1 \pm 6.5\%$, based on the analysis of the particles in uranium reference materials. The detection limits, defined as the diameter of the particle which produces alpha tracks more than one for a 28-days exposure, were estimated to be 0.8, 0.9, 1.1, 2.1 and 3.0 μm for the particles having the same uranium abundance ratios with NBL CRM U850, U500, U350, U050 and U010 reference materials, respectively. The use of alpha track detection for subsequent SIMS analysis is an inexpensive and an efficient way to measure uranium-bearing particles with higher ^{235}U abundances.

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1. Introduction

Since the discovery by Young [1] and Silk et al. [2] in the late 1950s of nuclear tracks recorded in solids, solid-state nuclear track detectors (SSNTDs) have been studied and used in various research fields. In the environmental sciences, for instance, SSNTDs have been used to detect radionuclides in contaminated soil and aerosol samples [3,4], and to determine concentration levels of radon in the environment [5]. Recently, this technique has been applied successfully to the analysis of individual uranium-bearing particles in environmental samples for nuclear safeguards [6–10]. Here, environmental samples are taken from nuclear facilities by inspectors of the International Atomic Energy Agency, and the isotope abundance ratios of individual uranium-bearing particles are measured to detect undeclared nuclear materials and activities. For the identification of uranium-bearing particles, the samples placed in contact with fission track detectors such as Lexan and Makrofol are irradiated with thermal neutrons in a nuclear reactor. Uranium-bearing particles are then identified by observing

the fission tracks recorded in the detectors, and are measured by using thermal ionization mass spectrometry (TIMS) to reveal the uranium isotope abundance ratios. The number of fission tracks depends on the amount of ^{235}U atoms in the particle. Therefore, uranium-bearing particles with high ^{235}U abundances can be selectively identified, prior to the isotope abundance ratio analysis. This fact is significantly important for nuclear safeguards. Since fission tracks are induced by the irradiation of uranium with thermal neutrons, a neutron source such as a nuclear reactor is necessary, which limits the availability of this technique.

Secondary ion mass spectrometry (SIMS) is also a powerful tool to measure isotope abundance ratios of uranium-bearing particles [11,12]. The recent development of an automated particle measurement (APM) software has allowed us to perform rapid screening measurement over a sample [13,14]. Here, the exact locations and rough ^{235}U/^{238}U abundance ratios of individual particles can be determined by obtaining ^{235}U$^+$ and ^{238}U$^+$ secondary ion images. The detection capability of uranium-bearing particles with high ^{235}U abundances has been significantly improved, owing to the development of this software. However, this software can be used only for CAMECA IMS-series SIMS instruments.

In a previous paper [15], we combined a fission track technique with SIMS, and demonstrated the effectiveness of this approach

for nuclear safeguards. The number of fission tracks being proportional to the number of fissionable ^{235}U atoms in each particle, uranium-bearing particles with the highest ^{235}U abundances can be detected selectively through observation of the fission tracks. This is particularly advantageous in the identification of undeclared nuclear materials and activities related to nuclear weapon programs. As mentioned above, fission tracks are induced by the irradiation of uranium with thermal neutrons. Therefore, a neutron source such as a nuclear reactor is necessary. In this work, we performed alpha track detection to identify uranium-bearing particles for subsequent isotope abundance ratio analysis with SIMS. Since alpha track detection is based on the decays of radionuclides through alpha emission, no neutron sources are necessary. In addition, the detectors are inexpensive, allowing us to perform the analysis more easily. Although Vlasova et al. [16] used SIMS for the analysis of a zircon particle containing natural uranium after alpha track detection, only $^{235}U^+$ and $^{238}U^+$ secondary ion signals were detected. We applied this method to the analysis of certified reference materials and environmental samples taken from nuclear facilities to confirm its effectiveness for nuclear safeguards.

2. Experimental procedure

2.1. Samples

Uranium-bearing particles in certified reference materials (CRM U050, U350 and U500, New Brunswick Laboratory (NBL), USA) were used for alpha track detection. The certified ^{235}U abundances of the U050, U350 and U500 materials were 5.010, 35.190 and 49.696%, respectively. In addition, three environmental samples taken from nuclear facilities by wiping particles with pieces of high-purity cotton cloth (10×10 cm^2, TX304, ITW Texwipe Co. Ltd., USA) were used for the identification and analysis of uranium-bearing particles. The NBL CRM U350 material was used for mass bias correction in mass spectrometry. For alpha track detection, CR-39 detectors (Baryotrak-P, Nagase-Landauer Co. Ltd., Japan) based on polyallyldiglycol carbonate ($C_{12}H_{18}O_7$) were used.

2.2. Sample preparation

Uranium-bearing particles with diameters of approximately 1, 3 and 5 µm from each standard reference material (U050, U350 or U500) were transferred onto Si wafers (5×5 mm^2, Semitec. Co. Ltd., Japan) by using a micro-manipulator under scanning electron microscope observation. The particles were then covered with a polycarbonate solution made through the dissolution of polycarbonate membrane filters (diameter: 25 µm; pore size: 0.2 µm; Toyo Roshi Kaisha Ltd., Japan) in a mixture of 1,2-dichloroethane and dichloromethane. These samples were placed in contact with CR-39 detectors and exposed for 7, 14, 21 and 28 days. After exposure the detectors were removed and etched for 3 h in a 7 M NaOH solution at 70 °C. A digital microscope (VHX-200, KEYENCE Co. Ltd., Japan) and a scanning electron microscope (JSM-7800F, JEOL Co. Ltd., Japan) were used for observation and counting of the alpha tracks.

Particles in the environmental samples taken from nuclear facilities were used for isotope abundance ratio analysis. These sample were recovered through filtration and collected on polycarbonate membrane filters by vacuum pumping [7]. The filter containing particles was placed in a 1-mL volumetric flask, and a mixture of 1,2-dichloroethane and dichloromethane was added. The mixture was then stirred until the filter was dissolved completely. The polycarbonate solution containing particles was poured onto a clean silica glass plate and dried to form a thin polycarbonate film containing particles. The films containing the particles in environmental samples were placed in contact with CR-39 detectors and exposed for 28 days. Here, four locations of the film and the detector were marked with a laser beam. After exposure the detectors were etched for 3 h in 7 M NaOH solution at 70 °C. The four marks of the film and the detector were carefully superimposed and uranium-bearing particles were then identified through observation of the alpha tracks with the microscope. A portion of the film containing each uranium-bearing particle (with dimensions of approximately 150×150 µm^2) was cut out with a Leica AS LMD laser-microdissection system (Leica Microsystems Ltd., Germany) and transferred onto a glassy carbon planchet (diameter: 25 mm; Hitachi Chemical Co. Ltd., Japan). Prior to SIMS analysis, the films on the planchet were ashed at a power of 300 W for 30 min with a plasma asher (PR-31, Yamato Scientific Co. Ltd., Japan).

2.3. Instruments

A secondary ion mass spectrometer (IMS-6 f, Cameca Co. Ltd., France) was used for isotope abundance ratio analysis. The pressure of the main chamber was less than 9×10^{-8} Pa. Each particle was irradiated with an O_2^+ beam of 15 keV. The current of the primary ion beam was between 6 and 18 nA. The mass to charge ratios of $^{234}U^+$, $^{235}U^+$, $^{236}U^+$, $^{238}U^+$ and $^{238}U^1H^+$ were counted sequentially in automatic peak jumping mode with acquisition times of 4, 2, 4, 2 and 4 s, respectively. Secondary ions were detected using an electron multiplier with a dead time of 30 ns. Other parameters were described in a previous paper [15].

The secondary ion signals vary with time in SIMS measurement. The signal drift was corrected by aligning the time series of isotopes through a linear interpolation approach [17]. Mass bias corrections were conducted by measuring isotope abundance ratio of a certified uranium reference material (NBL CRM U350) and the mass bias correction factor was calculated by using the true ratio divided by the measured ratio. The average value was calculated to be 0.45% per atomic mass unit. The hydride formation of uranium was determined by measuring the $^{238}UH/^{238}U$ ratio for each particle [18].

Uncertainties were estimated according to the principles described in the Guide to the Expression of Uncertainty in Measurements (GUM) [19,20]. Here, the contributions arising from the certified values of the reference material, the measured isotope ratios after time correction, and mass bias and hydride corrections were taken into account.

3. Results and discussion

3.1. Detection of alpha tracks created by particles in certified reference materials

Fig. 1 displays scanning electron images of NBL CRM U500, U350 and U050 particles with diameters of approximately 3 µm and their alpha tracks after exposure for 28 days. By careful observation of various images taken for these tracks using the SEM and the optical microscope, the numbers of alpha tracks for U500, U350 and U050 particles were determined to be 46, 20 and 2, respectively. Fig. 2 shows the numbers of alpha tracks of particles with diameters of approximately 1, 3, and 5 µm plotted against exposure time (7, 14, 21 and 28 days), together with their linear fit lines. Here, no alpha tracks were observed for the U050 particle with a diameter of 0.9 µm, which suggests that such a small particle with a ^{235}U abundance of less than 5% is difficult to
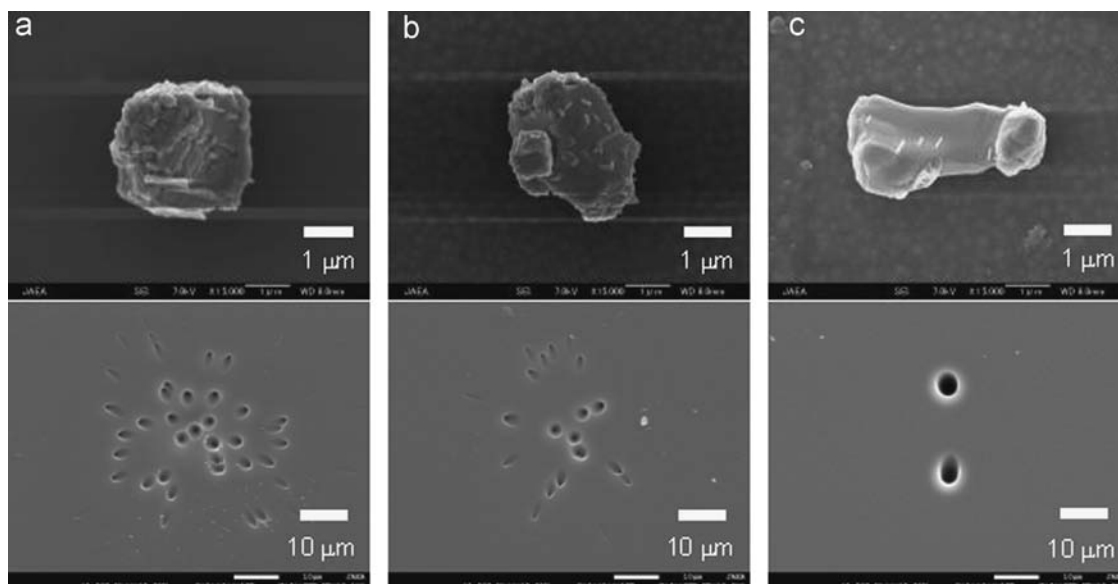
Fig. 1. Scanning electron images of uranium-bearing particles in NBL CRM: (a) U500, (b) U350 and (c) U050 samples and their alpha tracks after exposure for 28 days.
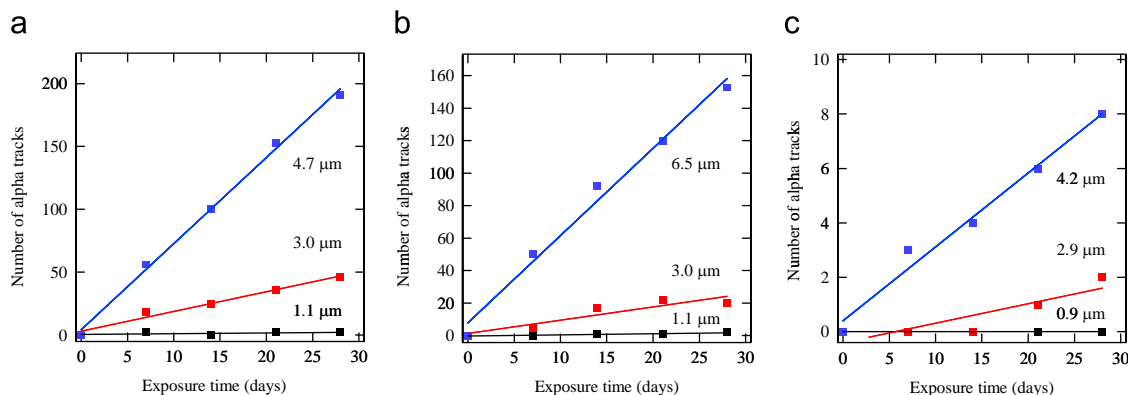

Fig. 2. Changes in the number of alpha tracks with exposure time measured for uranium-bearing particles in NBL CRM: (a) U500, (b) U350 and (c) U050 samples. The sizes represent diameters of the particles.

detect in a 28-day exposure time. The theoretical number of alpha tracks created by ^{234}U atoms can be calculated by the following equation.

$$n(\alpha T, 2\pi, {}^{234}U) = \frac{1}{2} \cdot \frac{4}{3} \cdot \pi \cdot \left(\frac{\varphi}{2} \cdot 10^{-4}\right)^3 \cdot d(U_3O_8)$$
$$\cdot \frac{M(U_3)}{M(U_3O_8)} \cdot A({}^{234}U) \cdot X \cdot T \quad (1)$$

where $n(\alpha T, 2\pi, {}^{234}U)$ is the number of alpha tracks created by ^{234}U atoms, φ is the particle diameter (μm), d is the density of the uranium compound, M is the molar mass, $A({}^{234}U)$ is the abundance of ^{234}U atoms, X is the ^{234}U specific radioactivity (Bq/g) and T is the exposure time.

The theoretical total numbers of alpha tracks created by ^{234}U, ^{235}U, ^{236}U and ^{238}U atoms were calculated and compared with the fitted data for the U500, U350 and U050 particles with diameters of approximately 5 μm in Fig. 2. Consequently, the detection efficiency, defined as the ratio of the observed number to the theoretical number of alpha tracks, was estimated to be 27.1 ± 6.5%. Pyo et al. [21] used LR-115 detectors to detect alpha tracks created by particles with depleted, natural, and low-enriched uranium, and estimated the detection efficiencies to be between 13 and 6%. These values are lower than the one of the CR-39 detectors in the present study.

In order to estimate detection limits in alpha track detection, the number of alpha tracks was plotted against the particle diameter. Here, uranium abundance ratios of NBL CRM U850, U500, U350, U200, U100, U050 and U010 were used for calculation. The detection limit was defined as the diameter of the particle which produces alpha tracks more than one for a 28-days exposure. Here, we assumed that the particles were in the form of U_3O_8 and spherical shape and the detection efficiency was 27.1%. As shown in Fig. 3, the detection limits were calculated to be 0.8, 0.9, 1.1, 1.4, 1.6, 2.1 and 3.0 μm for U850, U500, U350, U200, U100, U050 and U010, respectively. This means that it is possible to detect high enriched uranium particles with the diameter of approximately 1 μm by alpha track detection.

Regarding the U500 particle with a diameter of 4.7 μm, the contributions of the alpha decays of ^{234}U, ^{235}U, ^{236}U and ^{238}U atoms to the number of alpha tracks were calculated to be 94.9, 4.2, 0.2, and 0.7%, respectively. This means that most of the alpha tracks are created by the alpha decay of ^{234}U atoms. In fission track detection, the tracks are mainly created by fissionable ^{235}U atoms. The number of fission tracks is therefore strongly correlated to the ^{235}U abundance in the particle. On the other hand, the number of alpha tracks is not directly correlated to the number of ^{235}U atoms in the particle. However, not only ^{235}U but also ^{234}U is enriched in the uranium enrichment process. This implies that uranium-

bearing particles with higher [235]U abundances can be identified selectively by using alpha track detection.

3.2. Analysis of particles in environmental samples taken from nuclear facilities

Alpha track-SIMS was applied to the analysis of three environmental samples (A, B and C) taken from nuclear facilities. Fig. 4 (a) shows an alpha track image of a uranium-bearing particle in sample A, which produced 25 alpha tracks in the CR-39 detector. After plasma ashing an SEM image and an X-ray spectrum of the particle were recorded, as shown in Fig. 4(b) and (c). These confirmed that the particle had a diameter of 3.3 μm and contained uranium and oxygen. The peak assigned to carbon in the spectrum is due to the planchet material. In this study, we selected 10 particles having more than two alpha tracks for subsequent SIMS analysis. The uranium isotope abundance ratios were then measured for individual particles in sample A, and are shown in Fig. 5(a). Here, the $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ abundance ratios are plotted against the $n(^{235}U)/n(^{238}U)$ abundance ratios. The results obtained through stand-alone SIMS are also shown in Fig. 5(a). In this case, particles in the sample were recovered onto a planchet by using a vacuum impactor [22]. Each uranium-bearing particle was then identified by SEM-EDX, and transferred onto another planchet with a micro-manipulator for subsequent SIMS analysis. The micro-manipulation was performed to avoid molecular ion interferences by
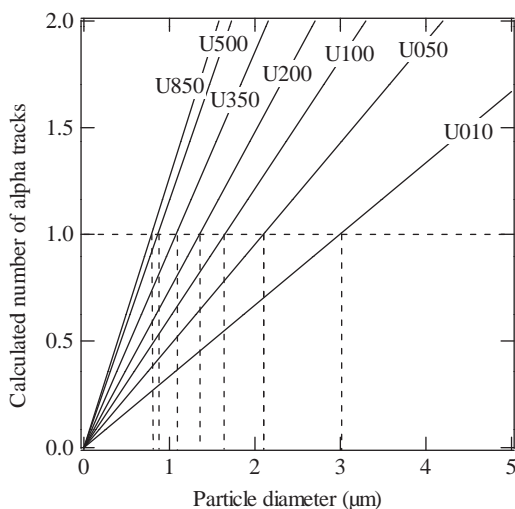
other elements in other particles and mixing of several uranium-bearing particles. The detailed procedure was described in a previous paper [12]. The $n(^{235}U)/n(^{238}U)$ abundance ratios obtained through SIMS without alpha track detection were between 0.0061 and 0.017. On the other hand, three particles with higher $n(^{235}U)/n(^{238}U)$ abundance ratios of approximately 0.25 were detected through SIMS with alpha track detection. The $n(^{235}U)/n(^{238}U)$ abundance ratios measured for all particles were between 0.0072 and 0.25. These results indicate that the use of alpha track-SIMS is effective for the measurement of uranium-bearing particles with higher [235]U abundances than the ones measured with the stand-alone SIMS. The isotope abundances of the uranium-bearing particle shown in Fig. 4 were determined to be 0.238% for [234]U, 19.9% for [235]U, 0.004% for [236]U, and 79.8% for [238]U. By taking into account these abundances and the particle diameter (3.3 μm), the detection efficiency was estimated to be 26.5%. Here, the particle was assumed to be in the form of U_3O_8. The efficiency corresponds with that determined using uranium certified reference materials (27.1 ± 6.5%). In the analysis of sample B with alpha track-SIMS, a particle with a $n(^{235}U)/n(^{238}U)$ abundance ratio of approximately 0.25 was identified, as shown in Fig. 5(b), while SIMS without alpha track detection afforded abundance ratios of up to 0.038. Moreover, in the analysis of sample C (Fig. 5(c)), three uranium-bearing particles with higher $n(^{235}U)/n(^{238}U)$ abundance ratios than the ones obtained with the stand-alone SIMS were detected by alpha track-SIMS.

The TIMS analysis also may be used for isotope abundance ratio measurement after alpha track detection. Here, a portion of the film containing a uranium-bearing particle is placed directly on a filament without plasma ashing for subsequent isotope abundance ratio analysis [6–10]. This procedure is simpler than that of alpha track-SIMS. However, there is a possibility of measuring several uranium-bearing particles simultaneously. Fig. 6 shows an example of an ashed film containing two uranium-bearing particles. By observation of the alpha tracks, it was difficult to identify the presence of two uranium-bearing particles in this film. If the film which contains these particles were measured with TIMS, average uranium isotope abundance ratios would be obtained. The presence of these two particles can be checked by SEM observation and they can be isolated with a micro-manipulator for subsequent SIMS analysis if required [12,23].

4. Conclusions

Alpha track detection of uranium-bearing particles was performed prior to isotope abundance ratio analysis with SIMS. The alpha track detection of uranium-bearing particles in certified reference materials (NBL CRM U050, U350 and U500) allowed a detection efficiency of 27.1 ± 6.5%. This method was then applied to the analysis of three environmental samples taken from nuclear
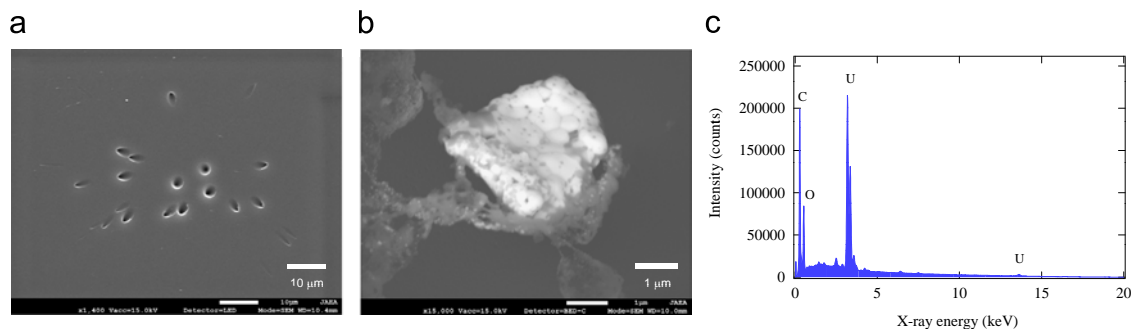
Fig. 3. The calculated numbers of alpha tracks plotted against the particle diameter. These were calculated based on the uranium isotopic compositions of each certified reference material (NBL CRM U850, U500, U350, U200, U100, U050 or U010).

Fig. 4. Scanning electron images of (a) alpha tracks and (b) a uranium-bearing particle in sample A and (c) its X-ray spectrum.

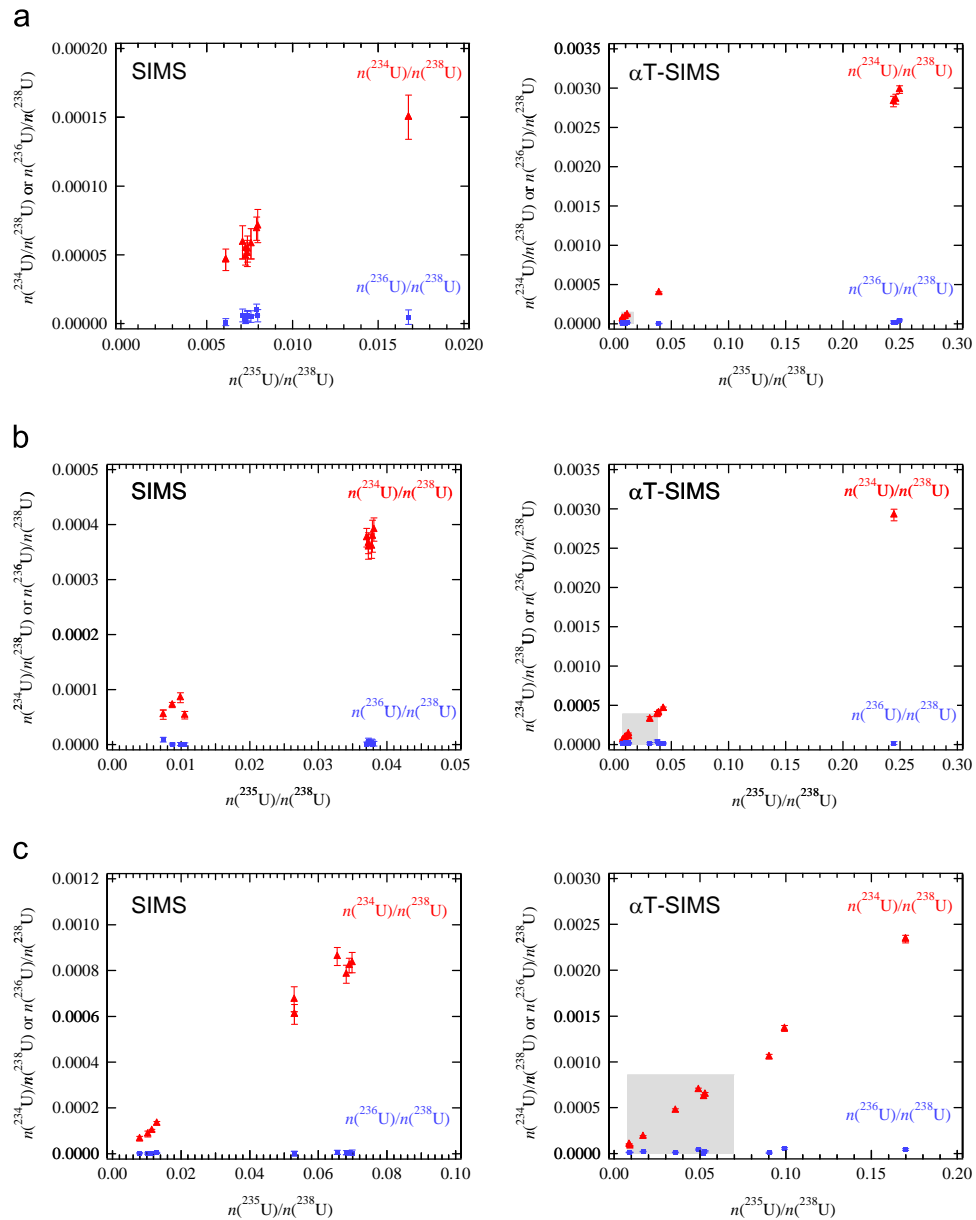**Fig. 5.** The $n(^{234}U)/n(^{238}U)$ or $n(^{236}U)/n(^{238}U)$ abundance ratios plotted against the $n(^{235}U)/n(^{238}U)$ abundance ratios measured for samples A, B and C by SIMS without and with alpha track detection. The shaded areas indicate the ranges of uranium abundance ratios obtained by SIMS without alpha track detection. The error bars represent expanded uncertainties ($k=2$). (a) Sample A, (b) Sample B and (c) Sample C.
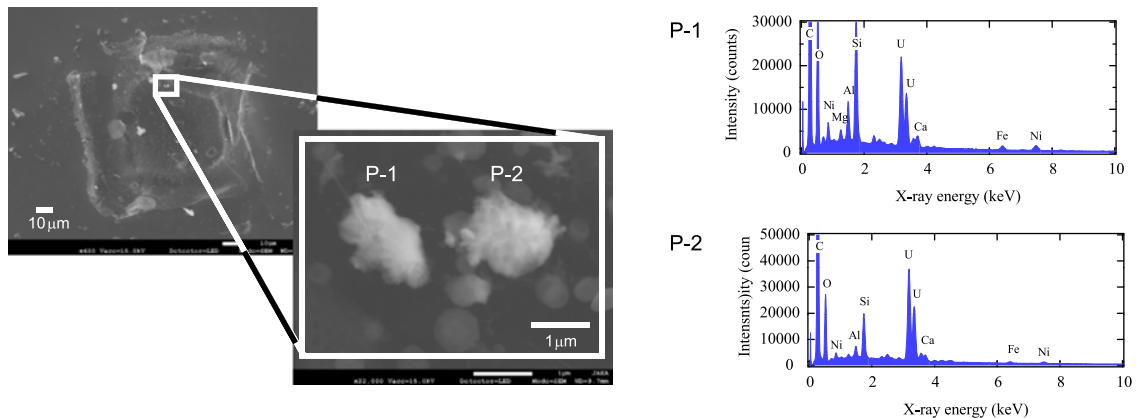

Fig. 6. Secondary electron images and X-ray spectra of uranium-bearing particles (P-1 and P-2) in an ashed polycarbonate film.

facilities. As a consequence, alpha track-SIMS enabled the measurement of uranium-bearing particles with higher $n(^{235}U)/n(^{238}U)$ abundance ratios than the ones obtained by SIMS without alpha track detection. This indicates that alpha track-SIMS is an effective tool for the measurement of uranium-bearing particles with the highest ^{235}U abundances in environmental samples.

As mentioned in the Introduction, the SIMS method using a newly developed APM software is an excellent technique for isotope abundance ratio analysis of uranium-bearing particles. The screening of $^{235}U/^{238}U$ isotope abundance ratios over a sample enables us to identify particles with high enriched uranium efficiently. The combination of fission track with thermal ionization mass spectrometry is also a powerful tool for this purpose. The fission track technique has higher sensitivity for the detection of uranium-bearing particles, compared to the alpha track technique. The analysis of sub-μm particles with depleted or natural uranium is possible in both APM-SIMS and FT-TIMS. The present study indicated that at least 3 μm in diameter is necessary to analyze such depleted or natural uranium particles in alpha track-SIMS. Therefore, the analytical performance of these APM-SIMS and FT-TIMS is superior to that of the proposed alpha track-SIMS method. However, the fission track method needs a nuclear reactor as a neutron irradiation source. In addition, there is a problem regarding mixing of several uranium-bearing particles. For TIMS analysis, a film containing a uranium-bearing particle is placed onto a filament. If the film contains several uranium-bearing particles, the obtained data would be an average value of these particles. In the case of APM-SIMS, the APM software can be used only for CAMECA IMS-series SIMS instruments. The mixing of several uranium-bearing particles is also a problem in this method.

On the other hand, the alpha track technique needs only inexpensive alpha track detectors for detection of uranium-bearing particles and can be combined with various mass spectrometers such as SIMS including other manufacturer's instrument, TIMS and ICP-MS. The mixing problem can be avoided as mentioned in Sections 3.2.

In this work, we used this method for the analysis of uranium-bearing particles. This method can also be applied to the analysis of plutonium-bearing particles. Since ^{239}Pu has a shorter half-life than ^{234}U, it is expected that smaller particles can be identified in alpha track detection. However, the analysis of plutonium-bearing particles with SIMS is considerably difficult due to a spectral interference of ^{241}Am to ^{241}Pu. Here, the ^{241}Am is a decay product of ^{241}Pu. The combination of alpha track detection with other techniques such as TIMS and ICP-MS would be able to overcome this problem.

Acknowledgments

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