PRELIMINARY STUDY ON THE ALPHA TRACK ANALYSIS OF SPHERICAL URANIUM METAL PARTICLES

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Alpha track analysis for the determination of the trace amount of alpha emitting nuclides in a very small particle was performed as an efficient and powerful technique during safeguard inspection. Metal particles with well-defined spherical shape, size and isotopic compositions as a reference material were used to correlate the number of tracks or track diameter with an isotopic composition, eventually, to identify the uranium enrichment in the environmental swipe samples. Slopes in the number of tracks versus the exposure time curve provide a simple insight into the uranium enrichment of an unknown particle. Low enriched uranium metal particles result in slopes still steeper than the depleted or natural uranium metal particles. In addition, a linear relationship between track diameter and particle size is thought to be a useful first stage analytical tool as an efficient and convenient inspection guide. The significance of the simple linear model was also judged using the usual statistical tests.

KEYWORDS : Solid-state Nuclear Track Detector, Alpha Track Analysis, Swipe Sample, Uranium Micro-particles, Uranium Content, Enrichment, Number of Track, Track Diameter

1. INTRODUCTION

Micro-analytical techniques for analyzing samples from a nuclear facility or its surroundings containing radioactive particles have drawn a much interest from many researchers and international organizations, such as the International Atomic Energy Agency (IAEA) and the European Atomic Energy Community (EURATOM) [1,2]. The analysis of environmental samples engenders some difficulty, depending on the type of environmental samples, such as water, soil, sediment, airborne particles, vegetation and bio-organism. Since they are usually collected far away from the site where they have been released, all those samples will depend on a particular source, scenario, dispersion processes and deposition conditions (environment, matrix, etc.). Thus, the swipe sampling is useful to identify the origin of the sample released from the artificial processes, such as radioisotope enrichment or neutron irradiation since the swipe sample is collected on-site and the sampling material used is small and compact compared with the general environmental sampling methods.

Nuclear track analysis is usually used to localize the particles of interest. Then, a measuring technique is used simultaneously to analyze the particles using the same sample specimen during the full experimental procedures; *e.g.*, a fission track analysis followed by thermal ionization mass spectrometry (FTA/TIMS) [1] or fission track analysis followed by secondary ion mass spectrometry (FTA/SIMS) [3]. Recently, it has been reported that the combination of fission track analysis with scanning electron microscopy (FTA/SEM) could provide information on the enrichment of individual particles exploiting the relationship between the number of fission tracks registered per particle and the particle size [4].

The solid-state nuclear track detecting method (SSNTDM) was already well-known in many scientific areas since its initial discovery of the first tracks in a crystal of lithium fluoride by D.A. Young in 1958 [5,6]. At present, etched track detectors is the most widely used method of measuring radon concentration levels [7]. When an alpha particle strikes through a nuclear track material, it induces damages along its path. The most common etchable solid state nuclear detectors are CR-39™ based on polyallyldiglycol carbonate ($C_{12}H_{18}O_7$), LR-115™ based on cellulose nitrate, Lexan™ and Makrofol™ based on bisphenol-A polycarbonates ($C_{16}H_{14}O_3$), which are insensitive to light charged particles, X-rays and gamma rays. After etching by basic aqueous chemical solutions (e.g., NaOH or KOH), a track along the particle path can be created and observed by an optical microscope equipped with a micromanipulator.

Many track-etch studies have extensively concentrated on fission track analysis [8,9]. Alpha track method is known to be useful for detecting and mapping alpha-emitting particles such as U, Pu, Am and Th in the environmental samples [10]. The present research is to establish a feasibility study, by using the alpha track analysis method, on the technology discriminating environmental swipe samples with particles containing different concentrations of ^{235}U. The properties of the alpha track image, which is recorded on the LR-115 track detector, vary with the particle size as well as the ^{235}U enrichment of the particles from the track image. In this study, the correlations of various track properties with particle size and ^{235}U enrichment using a uranium metal particle were determined.

2. EXPERIMENT

2.1 Materials

TX304 obtained from ITW Texwipe was used as swipe cotton, as is currently recommended by the IAEA for environmental sampling (Table 1). TX304 is made of 100% cotton and a strong, double-sided twill-pattern cotton wiper woven in a cross section of 118 × 60 threads per square inch. The product is manufactured with long staple cotton yarn to eliminate free-floating fibers on fabric surface. Swipe samples were taken by smearing pieces of cotton on the surface of worktables in a facility of the Korea Atomic Energy Research Institute (KAERI), Republic of Korea. The uranium distribution in the swipe samples was observed via alpha track mapping using LR-115 exposed for five days. The LR-115 detectors used in the present study were purchased from DOSIRAD, France (LR-115 film, Type 2, non-strippable). The detectors consist of an active layer of red cellulose nitrate on a 100μm clear polyester base substrate. The LR115 films are known to be unaffected by electrons or by radiations in the electromagnetic spectrum, such as gamma rays, X-rays or infrared radiations visible light, and are, therefore, to be handled without risk where such radiations are present. However, any form of abrasion, such as that arising from mechanical rubbing, folding, pressure, etc. should be avoided. LR-115 films were stored in a cool dry place below 5°C, relative humidity 50% or below in air tight plastic bags, and allowed them to reach the ambient temperature prior to use. Cotton gloves were used to handle the film samples.

The micrometer-sized spherical uranium metal particles prepared by KAERI were characterized by TIMS, as shown in Table 2.

2.2 Sample Preparations

Swipe samples were prepared by wiping the surface of the worktable with cotton cloth (10 × 10 cm^2, TX304, Texwipe). Various sizes of uranium metal particles in a range from 20 to 80μm and ^{235}U enrichments, namely, 0.2% (depleted uranium, DU), 0.7% (natural uranium, NU) and 19.95% (low-enriched uranium, LEU), were used in this study. The uranium metal particles were dispersed onto a poly carbonate film (Lexan) with a diluted Collodion solution which was used together with dried air to fix the particles. The prepared plates were then held in close contact with a piece of LR-115 detector cut to the size of the plate and fixed with tape. After the prepared samples were exposed for different lengths times, the Lexan film was removed from the plate and the LR-115 detector sheet was etched in a 2.5M NaOH solution for 160 minutes at 40°C in a shaking water bath to develop alpha tracks on the solid detector. The conditions of the bath were kept constant during the etching process to obtain a reproducible track number and shape. The effects of evaporation cannot be ignored, especially when being processed at 60°C. The dissolution of carbon dioxide

Table 1. Contamination Characteristics of TexWipe® Wipers*

Properties	Typical value
Particles 0.5~5.0μm	400 × 10^6 *particles/m^2*
Particles 5.0~100μm	15 × 10^6 *particles/m^2*
Fibers larger than 100 μm	80,000 *fibers/m^2*
Nonvolatile residue (IPA extractant)	0.35 *g/m^2*
Nonvolatile residue (DIW extractant)	0.20 *g/m^2*
Sodium	7 ppm
Potassium	5 ppm
Chloride	8 ppm

* Technical data sheet from the manufacturer

Table 2. Isotopic Composition of Uranium Metal Particles With Various ^{235}U Enrichments*

	Isotopic composition (atomic %)			
	^{234}U	^{235}U	^{236}U	^{238}U
DU	< 0.0005	0.2149	< 0.0005	99.7851
NU	0.0050	0.7004	< 0.0005	99.2946
LEU	0.1742	19.951	0.0019	79.8732

* LEU, NU and DU represent low-enriched uranium, natural uranium and depleted uranium, respectively

from the air into the sodium hydroxide solution results in a less-alkaline sodium carbonate solution. The temperature was controlled either by means of water jacket, or more satisfactorily, using a thermostatically-controlled environment, such as an air cabinet. Then, the film was washed in a diluted HCl solution adjusted up to pH 3 for at least 30 minutes, with agitation. The film was then rinsed for two minutes at 20°C in a 1:1 solution of distilled water and ethanol. Etched LR115 films are stable and can be stored more than ten years without any further treatment; therefore, they can be re-evaluated if needed.

2.3 Image Analyzing System for Track Analysis

Alpha tracks formed on the detector were measured using Leica Qwin software (Leica Microsystems GmbH, Germany) in MS-Windows operating system, while alpha tracks for the mapping of swipe samples were observed by the naked eye after etching. The program produced digitized images from a Leica DMLP microscope on which a Leica DC100 digital video camera (both from Leica Microsystems GmbH, Germany) was mounted. The threshold level was set on a bright field image until the entire area of the alpha track was detected. A further manual treatment was performed to exclude the background artifact in the track image.

3. RESULTS AND DISCUSSION

3.1 Alpha Track Analysis

An example of an environmental swipe sample is presented in Fig. 1. The corresponding alpha autoradiograph of the dust particles from the worktable is also shown for comparison. The light colored domain of the LR-115 plate after etching is seen as a mirror image of the swipe. The LR-115 image was magnified by the optical microscope, and could then be analyzed for the determination of the uranium enrichment. Before the investigation of the real environmental swipe samples, well-defined spherical uranium metal particles prepared by KAERI were used, as shown in Fig. 2. As expected, particles between $10\mu m$ and $100\mu m$ showed the typical alpha track patterns of the autoradiograph. In order to determine the uranium

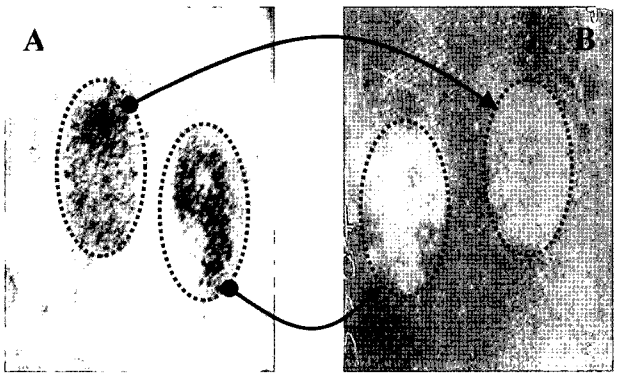

Fig. 1. Mirror Image of (A) a Swipe Sample and (B) Alpha-Autoradiograph on LR-115

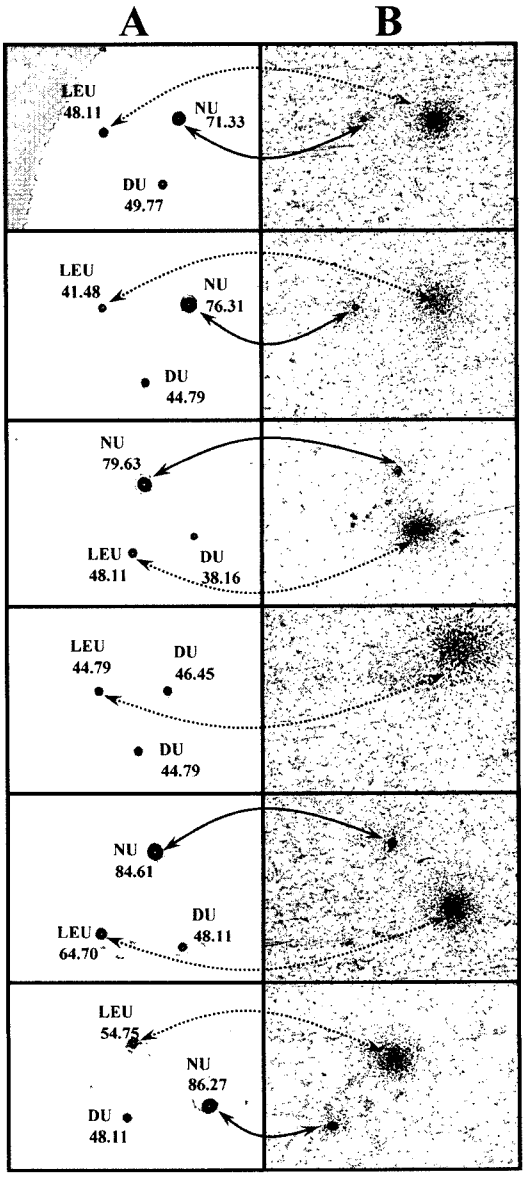

Fig. 2. Comparison of (A) Particles of Various Sizes and Enrichment Observed by Light Microscope (500 × Magnification). LEU, NU and DU Represent Low-Enriched Uranium, Natural Uranium and Depleted Uranium, Respectively. The Numbers Represent the Size of the Uranium Metal Particles in Micrometers and (B) Corresponding Alpha Autoradiograph

enrichment, the number of tracks per particle measured was compared with the values calculated using several assumptions (Section 3.2).

3.2 Comparison of the Measured Values of the Total Number of Tracks Per Particle With the Calculated Values

The radioactivity A (disintegration per day) of a spherical uranium metal particle with mass w (g), radius r (μm) and density d (g/cm^3) can be obtained as follows:

$$A = \lambda \times N = \frac{\ln 2}{T_{1/2}} \times N = \frac{\ln 2}{T_{1/2}} \times N_A \times \frac{w}{m_a} \quad (1)$$

$$\text{, where } w = \frac{4}{3} \times \pi \times r^3 \times d \times 10^{-12} \quad (2)$$

where λ is the decay constant (day^{-1}), $T_{1/2}$ is the half life (day), N is the number of radioactive atoms, m_a is the mass of the radioactive atoms (g/mol), and N_A is the Avogardo number. In the case of uranium, d is 18.95g/cm^3. Then, the number of tracks N_t as a function of exposure time t (day) and the uranium isotope concentration C (wt%) can be expressed by the following equation:

$$N_t = A \times C \times t = \frac{\ln 2}{T_{1/2}} \times N_A \times \left(\frac{\frac{4}{3} \times \pi \times r^3 \times d}{m_a} \right) \times C \times t \times 10^{-14} \quad (3)$$

If the particle contains only uranium isotopes, the total number of tracks per particle ${}^{T}N_t$ can be expressed by the following equations:

$${}^{T}N_t = {}^{235}N_t + {}^{238}N_t \quad (4)$$

$${}^{T}N_t = {}^{234}N_t + {}^{235}N_t + {}^{238}N_t \quad (5)$$

The number of alpha tracks observed is proportional to the ^{235}U enrichment. The calculated results do not agree well with the measured track numbers, as shown in Table 3. The calculated values of the number of tracks based on Eq. (3) are much higher in all cases than the measured values since Eq. (3) does not consider the geometrical efficiency of the alpha particles, registration efficiency of solid track detectors, or stopping power of alpha particles in the uranium matrix, etc. It is therefore assumed that simply 15% of the total number of tracks per particle can be finally seen in a LR115 plate after etching. This simplification might be helpful to understand the trend in the relative increment of the number of tracks. In the case of LEU, the relative increment of the number of tracks with the uranium enrichment is very high compared with that of DU and NU, but the relative increment of the number of tracks with the ratio of ^{235}U/ ^{238}U is quite reasonable in all cases considering that only the contribution of ^{234}U to the track count is taken into account in the calculation. The ^{234}U contribution becomes important with the enrichment. A steep increase in the number of tracks measured within an unknown particle indicates that this particle may be anthropogenic, but this is not sufficient to identify the enrichment in the environmental samples. On the other hand, the information on the number of tracks as a function of exposure time might be helpful for more reliable presumptive identification of the enrichment and for this reason it is discussed in the following section.

Table 3. Experimental and Calculated Number of Alpha Tracks From Uranium Metal Particle Assuming that r = 23.27μm is the Equivalent Size of 1μg of a Uranium Metal Particle

^{235}U enrichments (%)	# of alpha track in a sample (counts · day^{-1})			
	Experimental	Calculated*	Calculated**	Calculated***
DU	152±15	1,075	161	176
NU	165±17	1,103	165	318
LEU	2,232±115	2,217	332	5,651

* calculated by Eq. (4)
** considering the efficiency in Eq. (4): Eq. (4) × 0.15
*** including ^{234}U contribution and considering the efficiency: Eq. (5) × 0.15

3.3 Effect of Exposure Time on the Number of Tracks for the Identification of the Enrichment

In Fig. 3 the influence of exposure time between the uranium metal particles and the solid state track detector (LR-115) was investigated for one, two and three days of exposure time, respectively. According to Eq. (3), the activity increases linearly with exposure time. The total number of alpha tracks increased with the uranium

enrichment, as well as with the exposure time. LEU resulted in a slope much steeper than did either DU orNU. Consequently, the ^{235}U enrichments can be estimated from the abrupt slope changes.

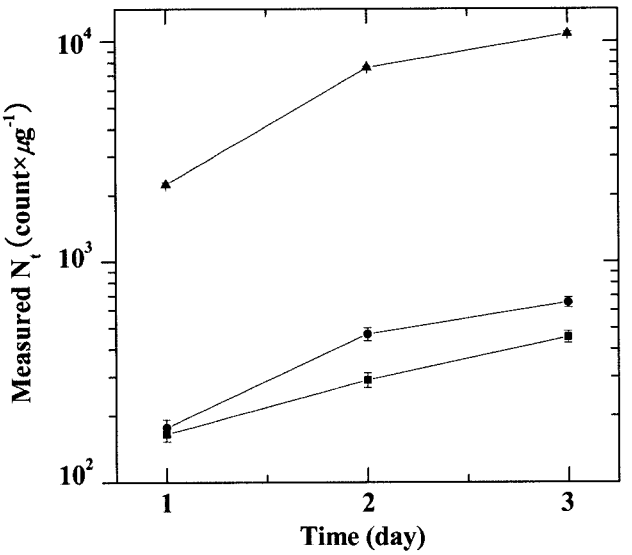

Fig. 3. Correlation Between Exposure Time of Uranium Metal Particles and the Number of Alpha Tracks Produced Using a Solid State Track Detector (LR-115) for (■) DU, (●) NU and (▲) LEU, Respectively

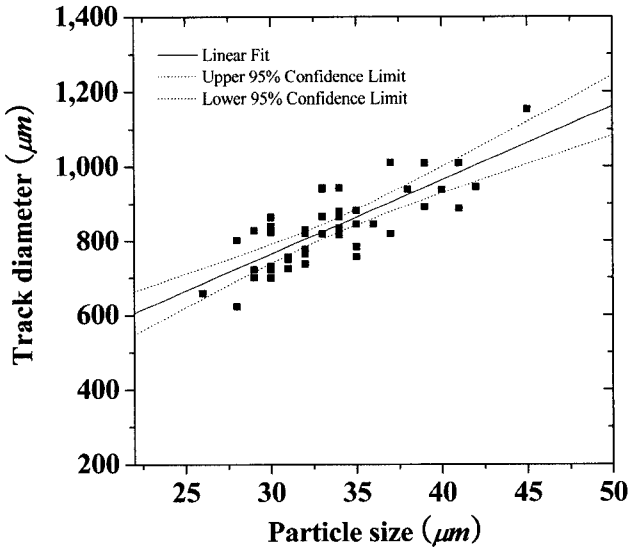

Fig. 4. Correlation Between Low Enriched Uranium(LEU) Metal Particle Sizes and Alpha Track Diameters

3.4 Determination of Particle Size of Uranium Metal Particles From the Track Size Measurements

It is important to obtain a database of the size and shape of the specific tracks for the well-defined fissile particles in terms of their size and shape. For a specific enrichment, the particle size can be inferred from the relationship with the track size, as shown in Fig. 4. The data (n=44) were successfully correlated with Eq. (6) via simple linear regression analysis.

$$D_{track} = 167 + 20 \times D_{particle} \quad (6)$$

An F test was used to assess the significance of simple linear models. The F value is the ratio of the regression mean square to error mean square, and a value of 72.599 was found in this study. A simple linear model was significant with a P-value of F-statistics less than 0.0001 and a coefficient of determination (R^2) value of 0.634. Consequently, the uranium metal particle size can be estimated from the Eq. (6).

4. CONCLUSIONS

The alpha track technique was used in this study to estimate the ^{235}U enrichment using reference uranium metal particles with various ^{235}U contents and sizes. This work has shown that alpha track analysis can be useful for the mapping of the particles containing only uranium in environmental swipe samples as well as for estimating the ^{235}U enrichments of those particles. The alpha track technique can be proposed as a screening tool to identify the enrichment of the particles of interest. In this study, it is proposed that the slope changes in the number of tracks vs. exposure time curve indicate uranium enrichment. Further studies on the effect of the exposure time are required to identify the enrichment more precisely. In addition, the overall efficiency for the total activity detectable on the LR-115 plate was only about 13%, 8%, 6% for DU, NU, LEU, respectively (Table 3). The measured values are too much lower than expected, so further studies on theoretical calculations of the number of tracks are also needed to more precisely understand the relationship between the number of tracks and the uranium enrichment as well as to analyze more complex systems containing various actinide elements from the real environmental samples.

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