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Developing Combined Fission Track Analysis and Alpha Track Analysis to Study the Spatial Distribution of U and Pu Sorbed to Environmental Particles

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Fission track analysis (FTA) is a direct technique to detect trace amounts of nuclides with high fission cross sections. To improve the capability of finding the exact location of contaminated particles in a sample, we have investigated combining alpha track analysis (ATA) with FTA in a single track detector. Alpha tracks are used to record the image of a scanning electron microscopy (SEM) finder grid in a LEXAN® track detector, along with fission tracks from ^{235}U-coated particles. The SEM finder grid was coated with B, and alpha tracks were obtained by the ^{10}B(n, α)^{7}Li nuclear reaction. U-contaminated environmental particles were simulated by sorbing U to ion exchange beads. Images were obtained by neutron irradiation and chemical etching. In this paper, we describe our method and possible applications.

KEYWORDS: fission track analysis, alpha track analysis, actinides, imaging

I . Introduction

During the last century, nuclear technologies have become a cornerstone of advanced industrialized societies, with applications ranging from energy production to biotechnology and military usages. However, these activities have also created problems of radiological contamination in the environment. Those problems are caused by releases from reactors, disposal of radioactive wastes, and sometimes nuclear accidents[1]. In the United States, the Department of Energy (DOE) is faced with remediation of the contaminated environment[2]. However, safe and cost-effective remediation requires a basic understanding of contaminant behavior within the affected area.

Within soils and sediments, contaminants are usually not distributed homogeneously, and the chemical and physical forms of the contaminants can differ as well[3]. This is particularly true for contamination by the actinides. For example, Pu may be found in environmental matrices as discreet particles, or it may sorb to specific surfaces in a given soil or sediment[4]. In these cases, certain particles will have much higher concentrations of Pu than that determined for the bulk soil or sediment. However, sensitive methods to locate the particles to which the Pu is associated are limited. Nuclear track techniques allow detection of certain nuclides by recording their nuclear decay events as damage in the detector. Among these techniques, fission track analysis (FTA) allows recording of fission events, and can provide spatial information on the location of fissionable nuclides within solid samples. Similarly, alpha track analysis (ATA) records alpha decay, and can provide spatial information on the location of isotopes that decay by alpha mode. An advantage of track analysis is that the sample is not destroyed by the analysis, and therefore can be used for other studies.

The goal of our work is to develop track analysis methods that allow determination of the spatial distribution and location of environmental particles contaminated with either U or Pu. ^{235}U and ^{239}Pu both have high thermal neutron induced fission cross sections. Neutron irradiation for these two isotopes causes fission to occur, which can be recorded in a track detector. However, locating those particles in the sample from the images observed in the track detector can be problematic. Therefore, we have developed an ATA locating system that records the image of a locating grid used in the sample mount in the same detector used for FTA.

II. Experimental

1. Samples

In our development work, we used ion exchange beads (Analytical Grade Ion Exchange Resin AG® 1-x2 and AG® 1-x4, chloride form, Bio-Rad Laboratories, Hercules, CA) to which we sorbed natural uranium (0.072% ^{235}U). These beads served as analogs to soil or sediment particles.

2. Track Analysis System

FTA for ^{235}U was completed as described previously[4], except for the irradiation time. The fission tracks generated upon a 12 minute neutron irradiation (thermal neutron flux

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of 10^{12} $cm^{-2} \cdot s^{-1}$) of the sample were recorded on LEXAN® detectors. After cooling for a minimum of 1 hr, the detectors were etched in a solution of 6 M NaOH (70 °C) for 10 min.

Alpha tracks could also be observed in the same LEXAN® detector by this method. ATA was used to record the image of a scanning electron microscopy (SEM) grid in the LEXAN® detector. We used the (n,α) reaction with ^{10}B, which also produced ^{7}Li, as described below. After track development, detectors were rinsed with water and air-dried. Both fission and alpha tracks were observed with a microscope.

2. Electrodeposition

The SEM grids used were made of Cu (Ted Pella, Inc., Redding, CA), to which boron was deposited. A solution of 0.25 M boric acid (J.T. Baker, Philipsburg, NJ) solution was adjusted to pH around 10 with KOH (Fisher Scientific, Fair Lawn, NJ). The Cu SEM grid served as the anode, and the cathode was a platinum wire. The current was adjusted at 0.75 A and held for 1 hour. After electrodeposition, the grid was air-dried.

3. Sample preparation

The U-ladened ion exchange beads were placed on the LEXAN® detector, and covered with the B-coated SEM grid. This "sandwich" was then taped together to immobilize the ion exchange beads and grid during irradiation. The taped LEXAN® samples were then sealed in a polyethylene bags, and polyethylene bags were heat sealed in 4 dram polyethylene vials for irradiation. After neutron irradiation, samples were unpackaged, and the tape with the boronated SEM grid and ion exchange beads were removed from the LEXAN® detector, and saved for SEM analysis. LEXAN® detectors were etched and observed under a microscope to identify the location of the U-ladened ion exchange beads in the sample. The exact location is provided by the image of the SEM grid, which appears as alpha tracks.

4. SEM experiments

Scanning electron micrographs were obtained using a Hitachi S-570 scanning electron microscope. The tape to which the boronated grid and U-ladened ion exchange beads were adhered were sputter-coated with gold to obtain the SEM images shown in this paper.

III. Results and discussion

1. FTA and ATA

Figure 1 shows a typical image of fission tracks obtained when U-ladened ion exchange beads are irradiated with thermal neutrons. As can be seen, the fission tracks are not distributed randomly within the detector, but rather are arranged as discreet star burst patterns. The patterns in this figure indicate that two ion exchange beads were located in this field of view. Thus, FTA provides spatial information on the distribution of ^{235}U particles in this sample. We have obtained similar results for sediment particles contami-

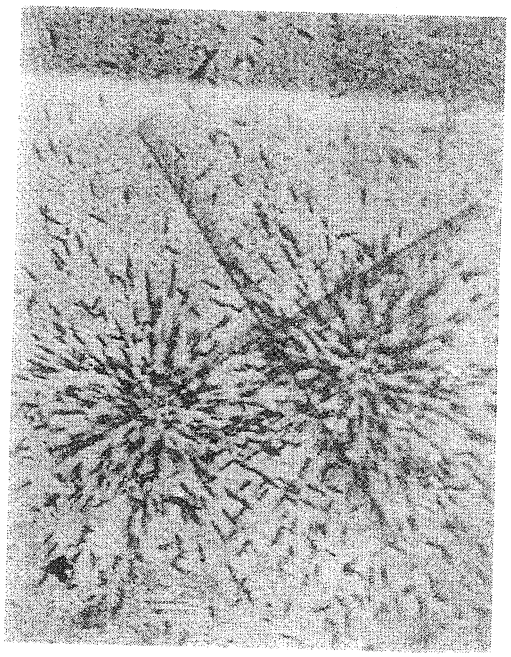

Figure 1 Fission track image of two soil particles contaminated with either ^{235}U or ^{239}Pu. Note that the fission tracks are not randomly distributed within the detector, but rather arranged in a "star-burst" shape indicative of a discrete particle. Magnified 400 times.

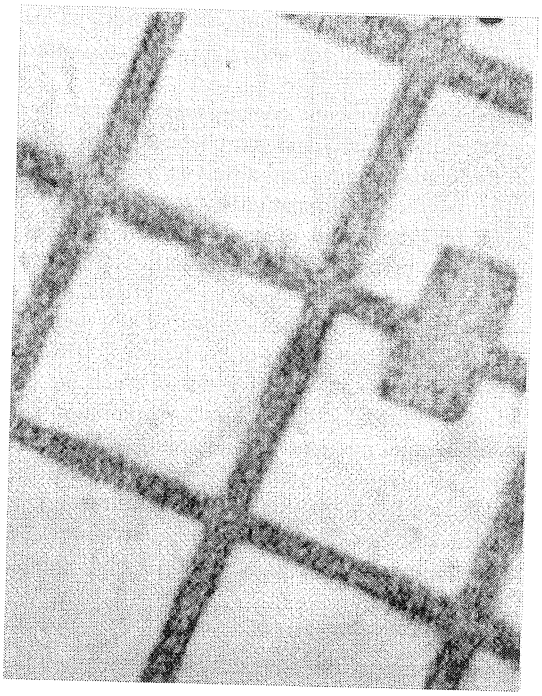

Figure 2 Alpha track image of an SEM finder grid that was coated with B and irradiated with neutrons as described in the text. Magnified 100 times.

nated with ^{239}Pu[4]. It is important to note however, that without the image of a locator grid in the track detector, relocating those two ion exchange beads in a bulk sample would be very difficult.

Figure 2 is the image of alpha tracks obtained after B was electrodeposited on the SEM grid and irradiated with neutrons. This image was obtained using the same irradiation and etching procedure used for FTA of the ion exchange beads shown in **Fig. 1**. Thus, if the tracks from both fission and alpha events observed independently in **Figs. 1** and **2** can be recorded in the same LEXAN® detector, the location of the contaminated particle(s) can be found in the original sample mounted in contact with the finder grid. It is important to note that the tracks observed in the LEXAN® detector provide a mirror image of the sample mounted on the finder grid.

This is demonstrated in **Fig. 3** (**A** and **B**). Fig. 3A is the image recorded in the LEXAN detector; it represents both FTA and ATA recorded in the same detector. The fission tracks are the circled star bursts, whereas the alpha tracks make up the outline of the SEM grid. **Fig. 3B** is an SEM image of the sample from which the combined FTA/ATA was obtained. In both images, the locating system can be clearly seen. The locations of the star burst patterns from the FTA for ^{235}U correlate exactly with the location of the actual ion exchange bead observed in the SEM as shown, except that the two images are mirror reflections of each other.

IV. Conclusions

We have demonstrated that we can record both fission tracks and alpha tracks in LEXAN® detectors. This allows us to record the images of U-ladened ion exchange beads by FTA and the image of an SEM locator grid by ATA in the same detector. The outline of the locator grid in the LEXAN® detector provides an absolute locating system for contaminated particles in the actual sample. We have verified this by matching the track detector images with SEM images. We are now working to apply this method to actual environmental particles, and our method should allow location of ^{235}U and ^{239}Pu contaminated soil or sediment particles. Once these particles can be located, they can be further studied by other microscale techniques.

V. Acknowledgements

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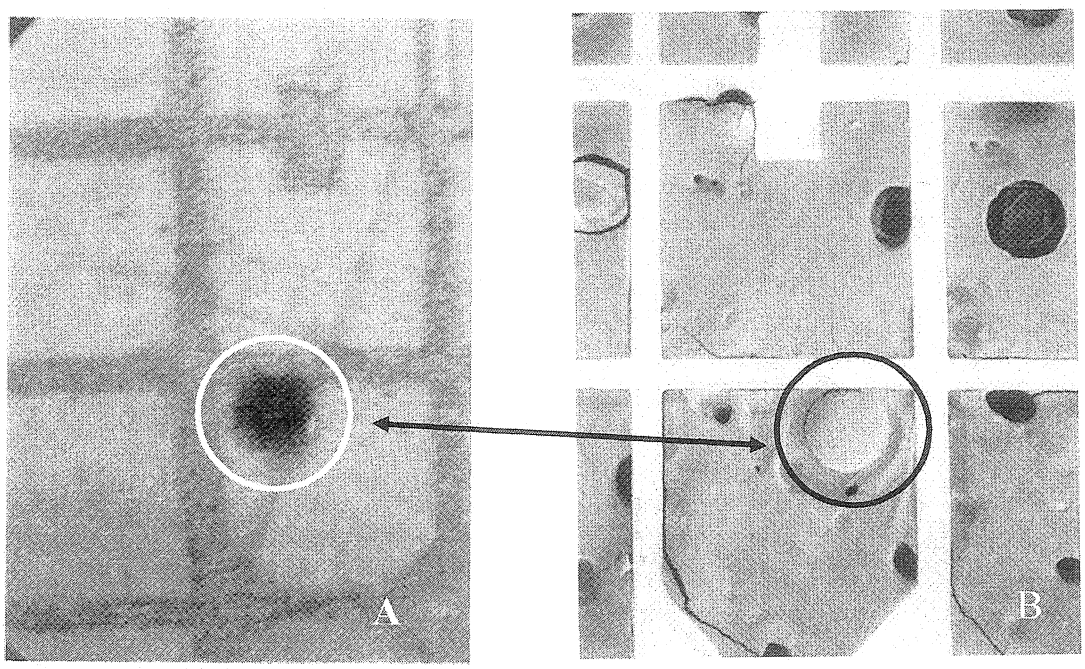

Figure 3 A. Image recorded in a LEXAN track detector. The grid system was produced by alpha tracks, as shown in Fig. 2. The dark spot that is circled was produced by fission tracks from the ^{235}U-ladened ion exchange bead (see text). **B** SEM image of the sample used to produce the fission track image shown in Fig. 3A. The ^{235}U-ladened ion exchange bead is circled. The dark circles are imperfections in the tape used as the backing to hold the sample (see text).

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