

Fission Track Analysis of Electrodeposited Uranium Alpha Sources

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Introduction

Fission track analysis (FTA) is a highly sensitive and reliable analytical technique for mapping the spatial distribution of trace levels of fissile isotopes. The FTA technique uses the neutron induced fission reaction for the detection of the isotopes and follows a three step process that includes: sample preparation, irradiation, and etching. In this technique, a sample is closely packed with a polymer detector and irradiated with thermal neutrons in a nuclear research reactor to induce a fission reaction. The energetic fission fragments created in this reaction tunnel into the detector, creating tracks along their flight path [1]. These tracks correspond to the location of the fissile element in a sample. To make the tracks more visible, the detector must be chemically etched. The damaged areas created in a detector have different chemical and physical characteristics than an undamaged section [2]. Due to these characteristic differences, the damages on the detector dissolve faster than the rest of the region during the etching process [3]. Faster dissolution of a damaged part leads to an enlargement of the track and makes it visible under an optical microscope. The number of tracks created during FTA is proportional to the number of fissile atoms, the cross section for thermal neutron induced fission, and the neutron flux [4].

Experimental

The FTA experiments used to evaluate the distribution of uranium on electrodeposited sources were performed at the Pennsylvania State University Radiation Science and Engineering Center (RSEC). The Breazeale Nuclear Research Reactor at the RSEC provided multiple irradiation positions to perform the irradiations. Two positions, dry tube and beam port 4, were used for these experiments. The two positions had different neutron flux levels of approximately 1.7×10^{13} cm²/s and 3×10^7 cm²/s, respectively, to generate ideal number of tracks from each sample.

The FTA irradiation time was set based on the fissionable uranium content to allow the formation of 1,000 to 20,000 fission tracks per sample. Equation 1

was used to estimate the time required to create the desired number of tracks.

$$t = \frac{N_t}{\epsilon \Phi \sigma_f N} \quad \text{Equation 1}$$

N_t is the number of fission tracks per particle, N is the number of fissionable atoms in a sample, ϵ is the track formation efficiency, σ_f is the fission cross-section, and Φ is the thermal flux. Based on multiple test runs, the efficiency (ϵ) was estimated to be 0.65 for the beam port irradiation.

The custom designed polyethylene sample holder (Figure 1) was used to keep samples at a stationary position during the irradiation. It was designed to hold multiple samples for a single irradiation to increase productivity and enable the irradiation of multiple samples using identical conditions.



FIGURE 1: Polyethylene sample holder used in the dry tube irradiation

Electrodeposited uranium samples were placed in the sample holder with Lexan detectors. Once the irradiation was completed, the detectors were developed with 6 M NaOH at 70 °C. The Lexan detectors were observed under an optical microscope and track locations were recorded using ImageJ software.

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The track analysis software divides track locations into rectangular sections and counts tracks per section. The percent distribution of tracks was also calculated by the program. The program performed relatively well; however, there was an inherent limitation in dividing a circular image into rectangular sections. The outer sections will only have portions of their surfaces covered by the tracks, as shown in the Figure 2. The gray section represents an area where tracks will be located and the rectangles represent the sections. The probability of tracks being located in the outer sections will obviously be lower than that of the inner ones. So only the sections with tracks present on their entire surface had any value in this study. These sections were referred to as the effective sections.

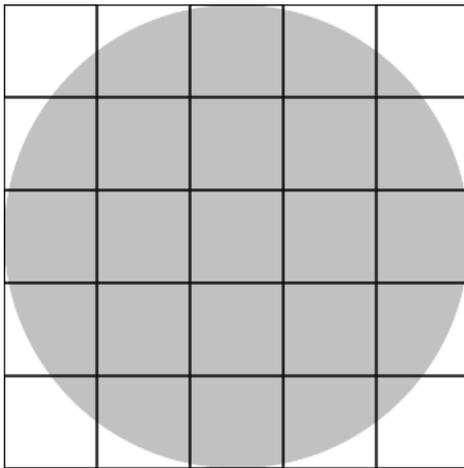


FIGURE 2: Illustration of the sectional track analysis

Results and Discussion

The ^{233}U sources were prepared by punching out 4.76 mm diameter subsamples from a larger titanium disk. The samples had enough fissionable particles to be irradiated in the beam port 4 position. The total neutron flux level was managed by adjusting the sample distance from the neutron beam source and the irradiation time. 91M1B-1 and 91M1C-1 sources were irradiated twice at different flux levels, 3.70×10^7 and 1.28×10^8 neutrons/($\text{cm}^2 \cdot \text{s}$), to generate the ideal number of tracks for the analysis. The neutron flux levels were measured by irradiating a gold wire. The number of tracks generated in the process was directly proportional to the neutron fluence and the sample activity as expected. One track was generated per every 10^9 to 10^{10} uranium atoms.

The overall image appeared to have fairly evenly distributed tracks. A sample track image is shown in Figure 3. Close visual inspection of the image also revealed no localized high concentration of tracks. For the sectional analysis using the track count software, images were divided into five vertical and five horizontal sections, totaling 25 sections.

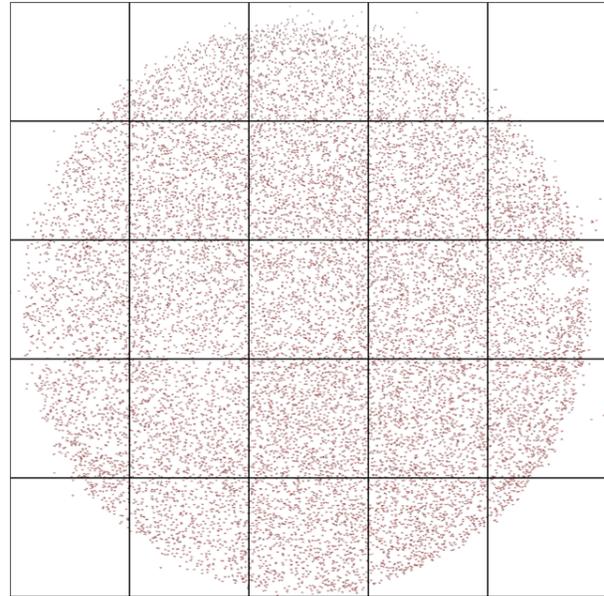


FIGURE 3. ^{233}U sample fission track image

The number of effective sections was different for each sample due to the different degree of distortion in the overall images. This fact was put in consideration for the analysis. In most cases, about 6 to 7% of tracks were located in each of the effective sections. Due to the stochastic nature of the fission reaction, there were some variations in track concentration. However, the variations were statistically within the 95% confidence level of each other, which indicated that the track distribution was uniform among the effective sections.

Conclusions

The fission track analysis procedure was effective for evaluating the spatial distribution of uranium on the surface of electrodeposited sources. The procedure was developed and tested on uranium samples but should be applicable to any other fissionable samples. The data analysis routine included removing defects on a detector, counting tracks, and analyzing the distribution. Except for the defect removal, entire process was automated.

Even actinide distributions were observed for both natural uranium and ^{233}U sources. The sectional track distribution analysis showed that about 6 to 7% of tracks were located in each of the sections. These variations were statistically identical at the 95% confidence level of each other. Also, surface defects and imperfections had no effect on the electrodeposited actinide distribution. The uranium did not use surface imperfections as nucleation sites. Tracks were distributed evenly in the Lexan detector, and no difference was found in the track density or homogeneity between the damaged and regular samples.

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