Detailed Fission-Track Radiography Methods

Background

Fission-track radiography can be done in combination with petrography and scanning electron microscopy (SEM) to determine the association of uranium within aquifer sediments at the microscopic scale. Uranium concentrations in the solid phase at typical U.S. Department of Energy Office of Legacy Management (LM) sites are commonly too low (< 15 mg/kg) to be easily detected using more traditional techniques often used for uranium ores. These traditional techniques include direct mineral identification through petrography of thin sections or the identification of bright spots with SEM backscatter electron imaging (BSE) due to the presence of uranium as a heavy element. In addition, the uranium at LM sites is often sorbed to organic carbon or amorphous coatings, making it necessary to first identify any microscopic locations of elevated uranium concentrations. Fission-track radiography makes this possible as a first step before doing mineral petrography and SEM work. For this work, petrography was completed using plain light, oblique light, reflected light, and crossed Nicol prisms. Petrography and SEM work focused more on the identification of grain coatings, cements, carbon and pyrite that were associated with elevated uranium, than on detailed mineral identification, since this project is looking more at secondary sorbed uranium than primary uranium associated with mineral grains.

Theory

Fission-track radiography involves the irradiation of a uranium-bearing sample with a flux of thermal neutrons that causes induced fission of the highly fissionable uranium-235 isotope. Fission fragments recoiling from the sample surface can be detected by covering a polished thin section with a suitable detector material. This detector material is typically muscovite mica or Lexan plastic. Fission fragments entering the detector cause structural disturbances that can be developed for optical inspection by subsequent etching of the detector with hydrofluoric acid (for mica) or sodium hydroxide (for Lexan). For this study, only muscovite mica was used for fission-track detectors, as prior work concluded that the mica detector was superior to the Lexan detector (DOE 2018). The developed fission tracks observed under a microscope reveal sources of uranium in the thin section much as autoradiographs would, but with much greater sensitivity and resolution. Thorium is a possible source of fission fragments, but its sensitivity to thermal neutron fission is approximately 100 times less than that of an equal concentration of uranium. References that discuss the above theory and the procedures discussed below include: Kleeman and Lovering, 1967; Fleischer et al., 1975; Zielinski and Rosholt, 1978; Zielinski and Budahn, 1998; and DOE 2018.

Glass Slide Preparation

Glass slides were prepared by polishing a thin section of an epoxy-impregnated sample of interest. Standard sized-thin section slides (27 x 46 mm) were modified by grinding the edges slightly (25 x 46 mm) to fit in plastic containers for irradiation. Polished thin sections are preferred because they permit the most intimate planar contact between the thin section surface and the detector. Poor contact during irradiation will produce poorer spatial resolution of uranium sites and a fuzzy image of fission tracks.

Mounting of mica detector

Thick books of mica sheets were progressively thinned and separated by inserting a razor knife along an edge and twisting. These actions expose fresh cleavage faces to be inspected for clarity, uniformity of surface, and freedom from scratches. The thickness of the final mica sheet used for a detector is not critical but should be sufficient to avoid curling, allow easy handling, and permit clear distinction of upper and lower surfaces under the microscope. This freshly exposed surface was again inspected for freedom from scratches or other obvious defects. The chosen sample of mica was further trimmed with scissors to the proper rectangular size, applied fresh face down on the thin section, and affixed at the edges with clear tape. In an area without tape, two marking holes were bored along one edge of the mica and thin section and one hole was bored along the other edge. This is done to allow for orienting the mica and the thin section when they are later separated. These holes were completed with a small drill bit and went completely through the mica and slightly into the thin section to create a mark only (taking care not to crack the thin section). Once marked, the mica is completely covered in tape and attached to the thin section to create the best contact possible between them.

Having a good mica detector requires obtaining relatively pure muscovite mica from sheet stock. The mica was checked for low uranium content by irradiating a thin sheet of it without a thin section, as a blank, under typical irradiation conditions, etching it, and checking for the presence of induced fission tracks (an alternative is looking for lack of fission tracks where the mica is not contacting any sample, e.g., thin section air bubbles).

Packaging for Irradiation

The prepared thin section slides were placed in polyethylene tubes and irradiated in a research reactor at the U.S. Geological Survey in Denver, Colorado. These tubes are cylindrical and are specially designed and sized for use in a research reactor (TRIGA; Training, Research, Isotopes, General Atomics). After grinding the long edge by about 2 mm, the thin section was placed into a piece of polyethylene foam of 1-inch diameter that is partially slit lengthwise to enclose the thin section and mica, and then inserted into the polyethylene tubes. This foam enclosure places some pressure on the thin section to provide good contact of mica and thin section surface during irradiation. Samples were irradiated at a neutron flux of 2×10^{12} neutrons per square centimeter per second for 8 hours for a total maximum neutron dose of ~11.6×10^{16} neutrons (less time and/or a lower neutron flux is required for higher concentrations of uranium). After irradiation, the samples are allowed to decay in storage for about 6 weeks or more to allow for the decay of some shorter-lived neutron activation products.

Development of fission tracks

Thin sections and polyethylene foam packing need to be carefully removed from the plastic tubes, e.g., using tongs with long arms that slide down along opposite sides of the inside diameter. Before using the tongs, a long thin spatula to break the seal between the foam and tube walls may be needed, trying to avoid contact of the spatula and tongs with any parts of the glass slide. The muscovite mica detector is removed from the recovered thin section with a razor knife that is carefully slid along and around the borders of the mica to cut any of the clear tape. The mica detector and the adherent tape on its upper surface are immersed in reagent-grade hydrofluoric acid (48%) for 10 minutes in a Teflon crucible. After about 5 minutes, the tape can easily be peeled off the mica with tweezers and removed. After 10 minutes, the mica is removed

and transferred to a large plastic beaker containing water. The mica is then washed with a strong stream of water and then isopropyl alcohol (70%) and set to dry overnight. The dried mica is mounted on a fresh glass slide using minimal amounts of clear tape placed along the ends. The mica is mounted original side down, in the same orientation as during irradiation, as indicated by comparison to the paired original thin section. To aid in slide comparison of the fission-tracked mica and the original thin section, mounting of the mica on the new glass slide should be as close as possible to the orientation of the thin section on the original glass slide.

Observation of fission tracks

The distribution of uranium in the original thin section is indicated by the distribution of fission tracks in the mica detector. To aid in locating fission-track areas compared to thin section areas, both the fission-track slide and the thin section slide were imaged with a high-resolution scanner. The backlight feature on the scanner provided better image brightness. These scans provide a "reference map" for finding and zooming into any areas of interest either digitally or with a microscope. Using the fission-track image, areas of interest with high fission tracking were identified. Digital overlays of the fission-track image and the thin section image were used to determine where fission tracking corresponds with mineral grains. This was achieved by allowing the fission-track image transparency to change while looking at the thin section image on a computer screen. Thus, the fission tracking could be compared with the underlying mineral grains. Care must be used to make sure the fission-track and thin section images are exactly aligned, best done using the three orientation holes. Areas of fission-tracking interest and the corresponding thin section locations were identified before using the microscope.

With the microscope, the focus is adjusted to view the underside of the mounted mica that was the surface in contact with the thin section. Etched fission tracks appear as linear features approximately 5 microns long. The prior scanned images are used to zoom into areas of interest with higher magnification. For both the fission-track and the thin section scanned images, pattern recognition of features (such as individual grains, air bubbles, orientation holes, etc.) become an invaluable guide for finding areas of interest on the microscope with subsequent magnification.

Paired areas of fission tracks and thin section were photographed for documentation using cameras that were mounted to the microscopes. This can be done with one microscope or with two microscopes with direct image projections to a video screen for more immediate comparison. Once identified, areas of relatively high fission-track density (and therefore relatively high uranium concentration) were revisited and further characterized in the original thin section using a scanning electron microscope. Distinct areas of high fission-track density were noted and prioritized before doing any SEM work. Note that the detection limit for uranium by SEM is on the order of several hundred to one thousand parts per million (ppm), so the greater sensitivity of the fission-track technique provides distinct insights for many samples of low-to-moderate uranium concentrations.

A tight cluster of fission tracks radiating from a central core could be caused by a very tiny uraniferous grain that is much smaller than the fission tracks. Fission track length can vary depending on the angle of incidence to the overlying detector and the presence of any separation between detector and thin section. Fission track images near bored holes in the detector and detector edges may be blurred because of increased separation. Most epoxy used to impregnate thin sections is very low in uranium, so this does not cause any interference.

Observations with the scanning electron microscope

When going to the SEM, the thin section slide is carbon coated and then a BSE image is created of the whole slide. This creates another "reference map" similar to the high-resolution scanning of the fission-tracked mica. With visual pattern matching of grains, the user can zoom into areas of interest with the SEM based on the prior identification of high fission-track areas. Thus, the prior "reference maps" from the fission-track mica and the thin section petrography become very useful guides. At higher magnification, the prior images of grains or coatings with higher fission-track density and the associated petrography become guides for locating those areas with the SEM. The efficiency of this approach is quickly realized, as without the fission-track mica images, locating areas with elevated uranium concentrations in grains or coatings with the SEM only is a "needle in a haystack" approach with overall low uranium concentration samples.

Once individual grains or coatings are located, the energy dispersive X-ray function of the SEM provides elemental analyses at that specific location. It is then up to the user to interpret those analyses. However, coatings that are mainly iron hydroxides vs. clays vs. Al/Si precipitates are relatively easy to identify.

References

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