Title
PROCEDURES FOR FINDING FISSION TRACKS IN MICA AND MYLAR.

Authors
Kleppe, Lowell M.
R.S.M., Sister Mary Roger

Publication Date
1966-08-25

UCRL-17075

University of California

Ernest O. Lawrence Radiation Laboratory

PROCEDURES FOR FINDING FISSION TRACKS IN MICA AND MYLAR

Research and Development

UCRL-17075
UC-34 Physics
TID-4500 (49th Ed.)

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

PROCEDURES FOR FINDING FISSION TRACKS IN MICA AND MYLAR

Lowell M. Kleppe and Sister Mary Roger, R. S. M.

August 25, 1966

Printed in USA. Price $1.00. Available from the Clearinghouse for Federal
Scientific and Technical Information, National Bureau of Standards,
U. S. Department of Commerce, Springfield, Virginia.

PROCEDURES FOR FINDING FISSION TRACKS IN MICA AND MYLAR

Lowell M. Kleppe and Sister Mary Roger, R. S. M.

Lawrence Radiation Laboratory
University of California
Berkeley, California

August 25, 1966

ABSTRACT

A procedure is described for recording, etching, and counting the tracks of fission fragments in mica and Mylar resulting from the neutron bombardment of various heavy elements. Sensitivity comparisons as well as annealing and etching conditions for both mica and Mylar are described in detail.

Tracks of fission fragments in mica and Mylar can be used to locate uranium and thorium in thin rock sections; mica and Mylar can be used with metals as threshold detectors for neutrons of different energies.

I. INTRODUCTION

Almost all insulating solids, including natural minerals, glasses, and plastics, are capable of recording tracks of fission fragments. These tracks may be formed by the "violent coulombic repulsion of the positive ions remaining after the scattering away of electrons by an energetic, massively charged particle."[7]

Upon being etched, these tracks may take any of several different forms. Diamond-shaped tracks are visible in mica. Dark spots are made in Mylar. Elliptically shaped or spike-like tracks can be found in various glasses.

These tracks may be counted by using an optical microscope, and the count may then be compared with those from known exposures to find what dose was given to various samples.

Our main interest in studying the tracks of the fission fragments was to investigate the possibility of using this method for threshold detectors for fast-neutron surveys around accelerators. We were also interested in studying thin slices of rock samples containing the heavy elements uranium and thorium, and using fission tracks to locate the positions of these elements in the rock.

II. CHOICE OF MATERIALS

One can find fission fragment tracks in several materials. According to the literature,[2, 6, 7, 8] one should be able to find fission tracks in almost any insulator, provided one uses the proper etching solutions, etching temperatures, and etching times.

Several insulating materials were investigated, including mica, Mylar, Toshiba dosimeter glass, pure silica quartz, Lucite, cellulose acetate, cellophane, neutron and gamma film badge backings, Pyrex, lime glasses, and vinyl chloride.

Fission tracks seemed to be most easily observed in the first four of these substances, and the first two proved to be most convenient and economical. The discussions in this report refer to experiments and etching procedures for mica (supplied by Victory Mica Mfg. Co. Inc; 1313 39th St.; Brooklyn, N. Y.) and Mylar (supplied by DuPont), as these were the two types of material studied most extensively. We would have liked to study other insulating materials, especially Lexan (supplied by General Electric), but were unable to obtain these materials during the time available to us for this study.

III. PROCESSING PROCEDURES AND OBSERVATIONS

The method for processing the mica and Mylar samples is simple and is fairly well described in the literature.[1, 2, 4, 6, 7] We have experimented with several variations of these procedures, and discuss them in the context of this report. It should be remembered that there is no one set way of etching the materials, as differences in irradiations and applications may necessitate variations. Some of these variations follow.

A. Etching Solutions

The basic method for developing the tracks in the mica is to etch the exposed sheet in 48% HF for about 3 hours at room temperature. Mylar etches nicely in 20% NaOH for about 30 minutes at 70°C.

These concentrations of the etchants seem to be quite satisfactory, and no variation seems necessary here.

B. Etching Temperatures

The temperature of the etching solution is an important consideration when etching Mylar. No tracks are visible after a 30-minute etching in 20% NaOH unless the temperature is above 50°C. Temperatures between 70°C and 80°C seem to give the best results; the maximum number of counts occurs after samples are etched in this temperature range.

For etching mica, the temperature of the etching solution does not appear to be nearly so critical; room temperature seems adequate.

C. Etching Times

If the expected number of tracks is large, it is often desirable to shorten the etching time. One difficulty in doing this is that the tracks are still very small and may not be clearly defined. However, unless the etching time is shortened, there may be severe overlapping of tracks that would cause errors in the counting of the tracks.

Overdeveloping the tracks also may cause counting errors due to the overlapping of the tracks. In Mylar a type of "mushrooming" (uneven growth) of the tracks results, and this, too, causes errors in counting.

D. Sample Preparation

There are several methods for preparing, irradiating, etching, mounting, counting, and preserving the samples of mica and Mylar. The steps that follow are the methods used by us that have proved most successful in obtaining reproducible results. (Some of these points are discussed in more detail later in this paper.)

1. The samples are cut to uniform size (in our work, approximately 1×2 cm).

2. A corner (for our work, the lower right-hand corner) of the sample is notched for identification so that, when reading the number of tracks, one will always be looking at the surface of the sample on which the fission fragments were incident.
   a. For mica, either face of the sample may be notched.
   b. For Mylar, the notch is made with the shiny side of the Mylar up. (Mylar has two types of surface when viewed with a microscope. The choice of the shiny side as the top surface was arbitrary on our part, as there was no apparent difference between surfaces in the sensitivity in recording tracks.)

3. The samples should be prewashed with alcohol to remove any oil deposited by the fingers in handling the samples.

4. The samples are then attached to a 3×5-inch card with Scotch brand Magic Tape so that the top face of the mica (see item #2 above) is next to the metal that is to be the source of the fission fragments.

5. After irradiation, the sample is removed from the cardboard, washed in alcohol, and placed in the etchant for the desired length of time.

6. After it has been etched, the sample is again washed with water and alcohol to remove the etchant and stop the etching process.

7. The samples are then viewed with an optical microscope and a count is made of a suitable number of randomly chosen fields of view over the entire top surface of the sample (except that portion of the sample that had been covered with tape when it was attached to the card).

8. For permanent retention the sample is mounted on a microscope slide with Duco cement and covered with a cover glass also cemented to the slide.

IV. COUNTING PROCEDURES AND OBSERVATIONS

A. Track Descriptions

The tracks of the fission fragments in the mica samples may vary in appearance from short rodlike spikes, when the etching time is very small, to clearly defined and easy-to-count diamond-shaped pits. These diamond-shaped tracks were the type that we studied most, and are shown in the photomicrograph of Fig. 1.

The tracks in the Mylar samples were less clearly defined than those in the mica. These Mylar tracks are shown in the photomicrograph of Fig. 2. The size, shape, and optical density of the track depend upon the etching time and etchant temperature. When a large number of tracks is expected, a shorter etching time is desirable, and the resulting tracks are small and dark and somewhat difficult to count. When the etching time is extended, one notices a "mushrooming" that results in larger but less dense and less well defined tracks. These larger tracks are generally easier to count than the smaller tracks, and yield satisfactory results as long as the number of tracks per field is less than about 40.

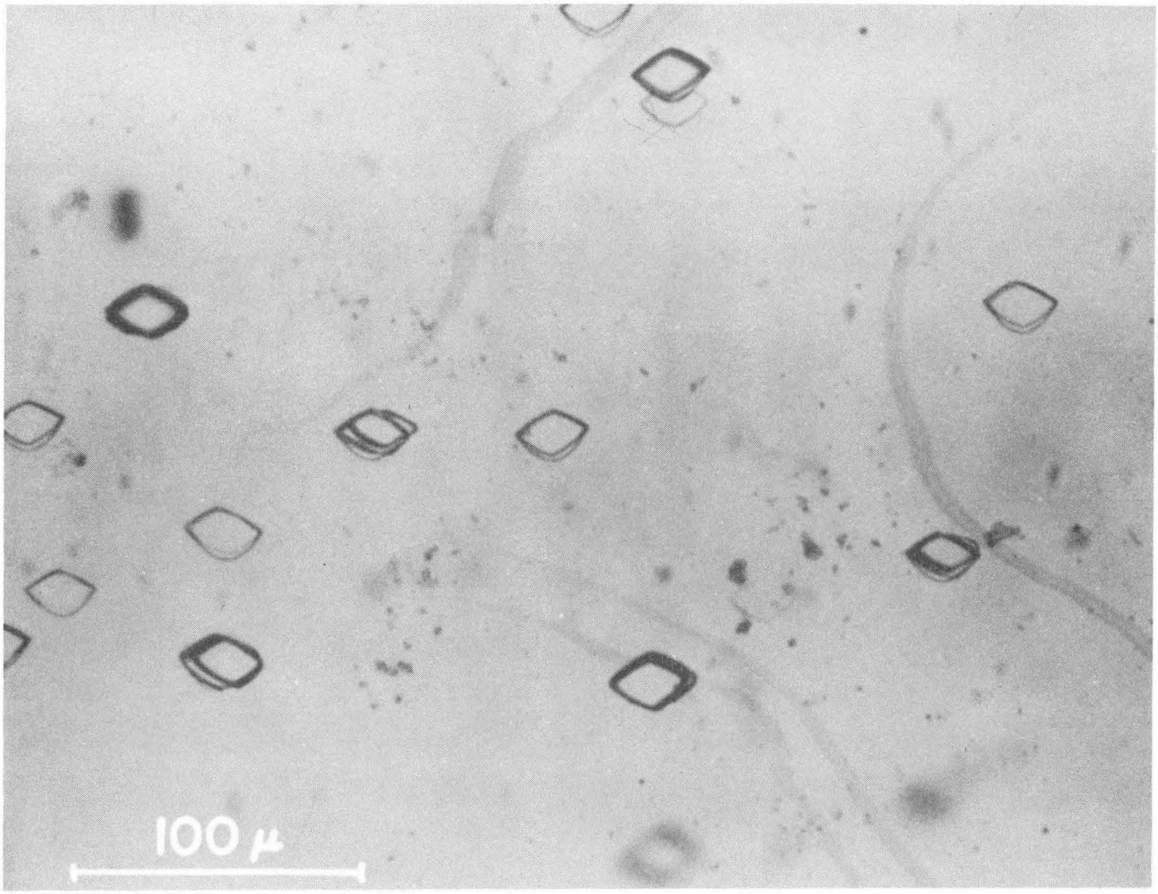
In both mica and Mylar there were things that looked as if they might possibly be tracks but either were unclear, were too close to another track, or looked a bit different from the other tracks in the sample. These were not counted.

B. Attenuation

It is important to place the sample on the slide in the orientation indicated by the notch. Fission fragments are attenuated in both mica and Mylar. In the Mylar, no tracks are visible on the surface that was not in contact with the metal in which the fission fragments originated. This same attenuation is also noted in mica. Because the mica is fairly clear, however, one can see tracks in it on either surface. It is important to count only the top surface of the sample.

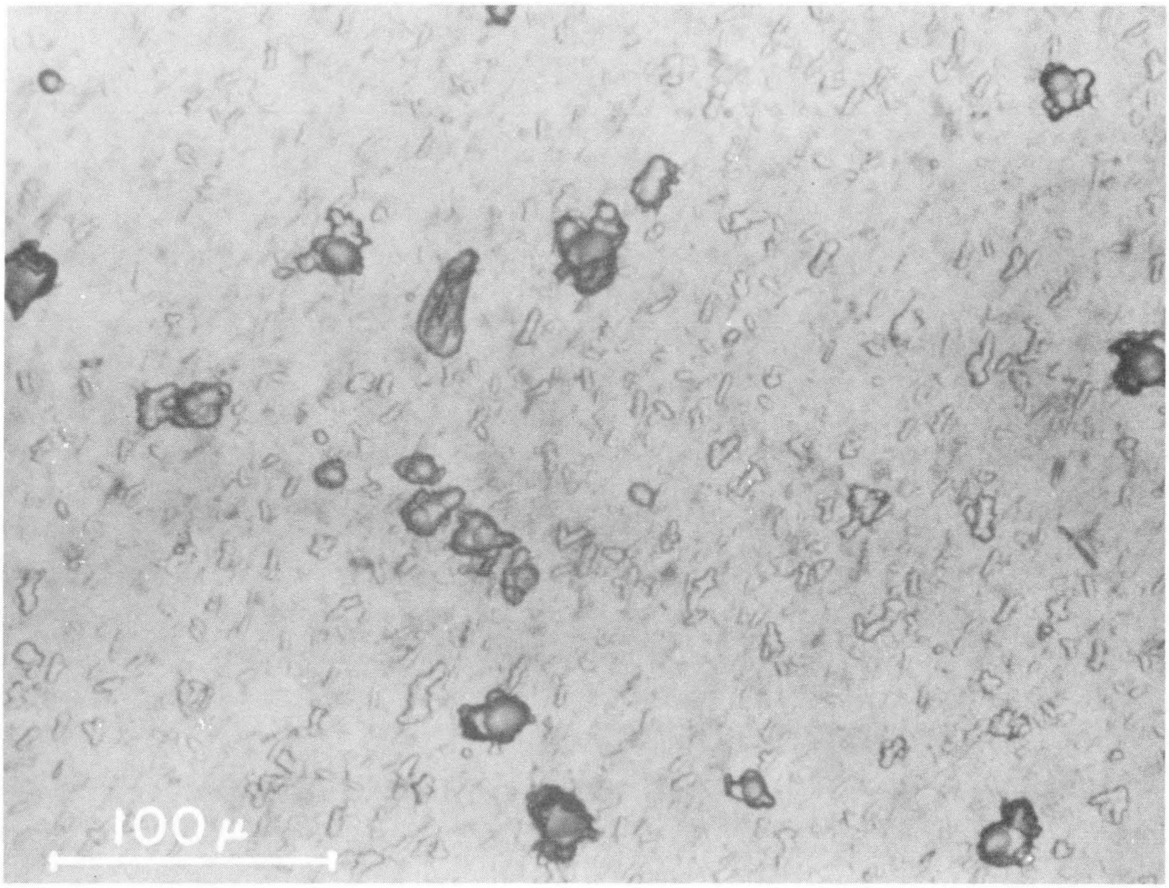
C. Scratches on the Mica

It was suspected that the number of tracks counted in mica samples could be changed if the surface of the mica were scratched. A slight decrease in the number of tracks was noted in the scratched areas, but the difference was very small. Scratched surface areas did interfere with the counting in that the optical clarity of the mica was diminished. Although scratching does change the number of tracks in the sample, the principal reason for keeping the surface as free of scratches as possible is to facilitate counting.



ZN-5746

Fig. 1. Fission tracks in mica.



ZN-5745

Fig. 2. Fission tracks in Mylar.

D. Background Tracks

In mica there is a track background due to naturally occurring impurities. Samples that were not irradiated were etched and counted. Three to four tracks per field were found when viewed at 210 ×. When the sample is irradiated, the contribution of fission fragment tracks from the impurities of uranium and thorium that occur naturally in the mica becomes important. A control sample is always included so that the contribution of fission tracks from these impurities can be estimated. This contribution is generally small. However, when a high neutron flux is used, a correction must be made by subtracting background from the total track counts in the mica samples.

In Mylar no background tracks exist naturally. However, some background has been observed when extremely high flux and energies are used, such as received by samples that have been placed in reactors or in the direct beam of the accelerator. For this reason, a control sample of Mylar is used whenever flux or energy level is high.

E. Annealing

An attempt was made to eliminate the naturally occurring tracks in mica. Annealing at temperatures up to 800°C for various periods of time up to 30 hours did not seem to reduce appreciably the tracks from the impurities in the mica. Some reduction was noted, but not enough to seem worth the effort. Prolonged annealing also caused deterioriation that reduced the optical clarity of the mica.

F. Pre-Etching

For some applications, it was found that a pre-etching of from 2 to 4 hours in 48% HF was desirable for the mica samples. The pre-etched tracks appeared larger than the tracks developed later, and were easily distinguishable in counting.

G. Staining

Various stains were used in an effort to make the surface tracks more easily distinguishable from the tracks lying deeper in the mica. Staining of the slides was eventually eliminated from the processing procedures, as it did not improve the ease of counting the tracks.

H. Track Distribution

The distribution of the tracks in the samples is generally not uniform over the entire surface of the sample. This nonuniformity requires that, for satisfactory counting statistics, a number of fields be chosen at random over the entire surface and counted. The entire top surfaces of a sample of mica and a sample of Mylar were counted. The results were graphed and studied. As a result, we feel that enough information can generally be obtained by counting 20 to 25 fields of view. However, when the number of counts per field is either very large or very small, the number of fields should be decreased or increased, respectively.

I. Sensitivity

There is a linear relationship between the number of tracks per field and the length of irradiation. Figure 3 shows this relation for both mica and Mylar. In this graph, the background correction has been made for mica. Irradiation of Mylar with no fission source in contact with it gave no tracks for these particular irradiations. The values shown in the graph are the result of irradiating depleted uranium with fast neutrons from a PuBe source with a yield of 8.6×10^7 neutrons/sec at a distance of 1.8 cm.

It can be noted from this graph that the sensitivity of mica and Mylar in recording fission tracks is nearly the same, the mica giving consistently a few more counts per field than does the Mylar for the same irradiation. A considerably smaller number of tracks appeared in the various glasses that received the same irradiation. This seems to agree with the results that have been obtained by other experimenters.[4]

The number of thermal neutrons necessary to cause one fission track in mica has been reported to be 2.3×10^4 thermal neutrons/track.[3]

Most of our work has been with fast neutrons. A very rough estimate of 3.6×10^7 fast neutrons/track has been made for our source and track-counting results.

J. Fading

There does not seem to be any fading in the number of tracks between the time of irradiation and the etching for either mica or Mylar unless the temperature is sharply increased. A loss of 50% of the tracks was noted in Mylar when samples were annealed for 30 minutes at 180°C. There was no change noted for samples of mica with this same amount of annealing. This fading is shown in Fig. 4.

K. Alpha Sensitivity

Some insulating materials are sensitive to alpha irradiation, and tracks resulting from alpha irradiation are visible in the samples. We did not observe any tracks of any shape or form in either the mica or the Mylar from alpha particles emitted by a sample of natural uranium.

L. Sample Thickness

The samples of Mylar that we found best were 0.010 in. thick. This is enough so that the fission tracks in the material are located on only one face of the sample, as the range of the fission fragments is considerably less than the thickness. The sample was not completely dissolved by the etchant when thicker samples were used, although this became a problem with thinner sheets of Mylar.

The thickness of the mica samples was not known as accurately as was that of the Mylar, as natural cleaving of the mica results in different thicknesses. A thickness great enough to prevent breaking when handled seemed to be adequate.

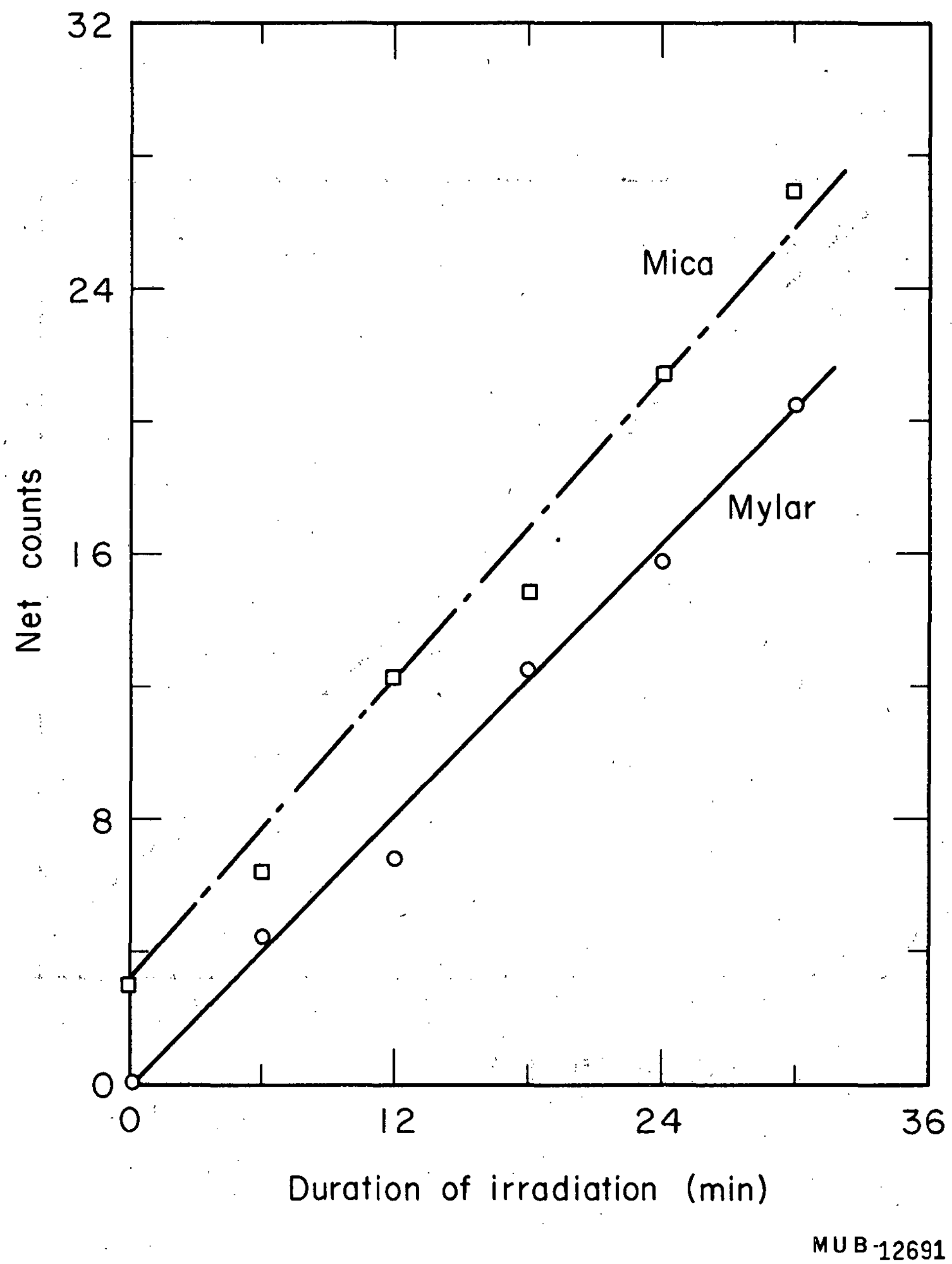


Fig. 3. Net counts for mica and Mylar as functions of irradiation time under natural uranium 1.8 cm from a PuBe source giving 8.6×10^7 n/sec.

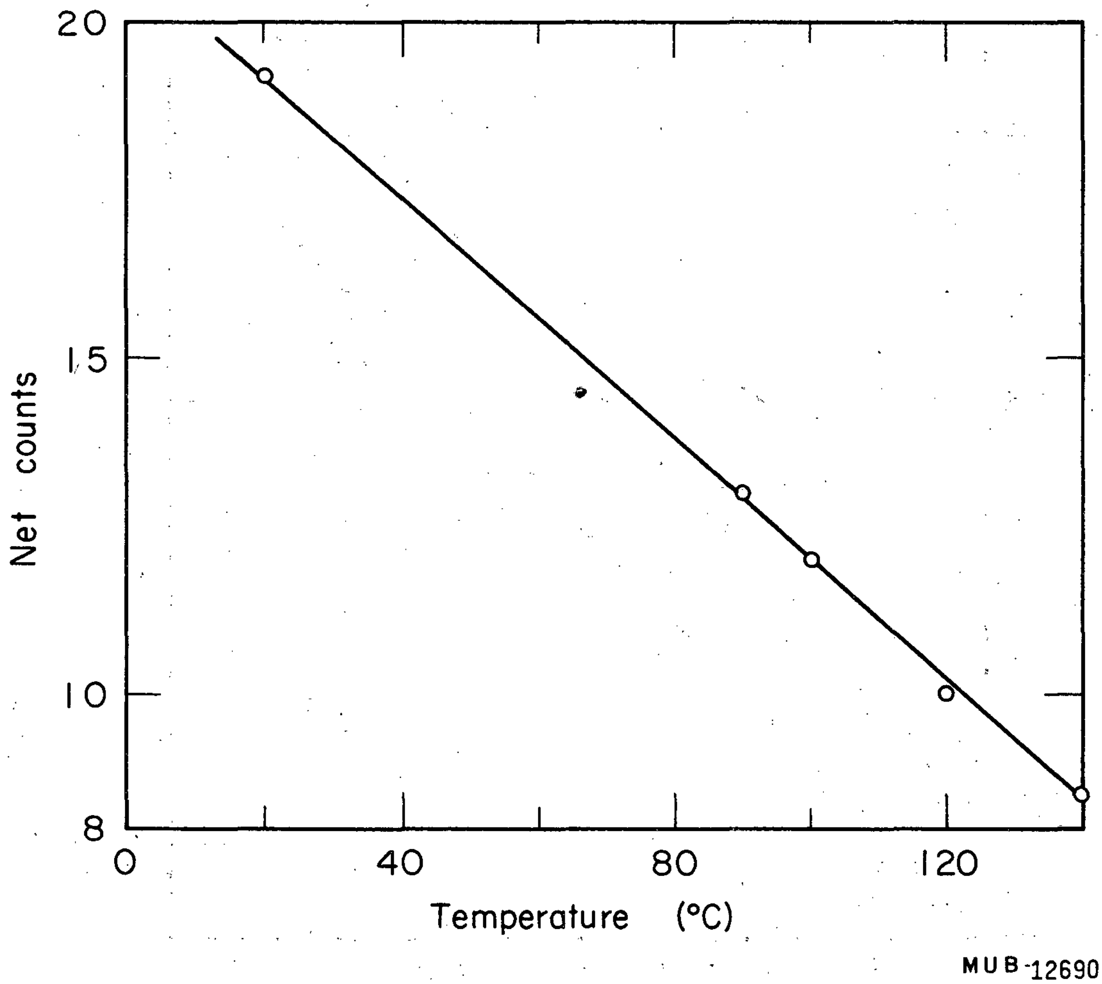


Fig. 4. Counts as a function of annealing temperature; Mylar, irradiated and annealed 30 min at various temperatures.

V. APPLICATIONS

A. Threshold Detectors

Several metals, each with different energies required for fission, were used as sources of fission fragments. These metals included aluminum, titanium, nickel, iron, copper, silver, lead, gold, bismuth, and depleted and natural uranium. Of these, gold, bismuth, lead, and depleted uranium were of the greatest interest to us for use in threshold detectors for fast neutrons.

The ratio of the number of tracks per field coming from foils of gold, lead, and bismuth exposed to high-energy neutrons in the 184-inch cyclotron was found to be approximately 1:2.5:5; depleted uranium was much higher.

Several surveys of the neutron spectrum near accelerators were made with these metals at different locations. The results of these surveys are currently being studied by the Health Physics Department.

B. Uranium and Thorium Detection in Rock Samples

An uncovered thin section (30 microns) of granodiorite from the Sierra Nevada containing uranium and thorium was placed in contact with mica and Mylar and irradiated in the Livermore pool-type reactor for 80 minutes in a fast-neutron flux of 3.7×10^{12} neutrons/cm^2-sec. It had previously been determined by γ-ray spectroscopy that uranium and thorium existed in the rock. The purpose of the exposures was to determine the sites of the uranium and thorium in the rock.

After etching, areas were found in the mica and Mylar that contained large clusters of tracks. These clusters were easily identifiable, and corresponded to grains of biotite located in the thin section.

The sensitivity of mica and Mylar were about the same, but the mica's optical qualities allowed easier comparison between the clusters of tracks and the sources of the fission fragments.

Currently, experiments are being done with lower fluxes of thermal neutrons in an effort to discriminate between the areas of uranium and thorium concentrations. The yield of fission fragment tracks resulting from irradiation with thermal neutrons should be less from the thorium concentrations than from the uranium.

The use of fission fragment tracks seems to be a useful tool in this type of study as well as other geological applications.[5,9]

C. Silver-Mica Capacitor

An experiment was done with a silver-mica capacitor to determine if tracks resulting from the fission of silver by fast-neutron bombardment in the 184-inch cyclotron could be located in the mica. Three capacitors were used in this study. One was used as a control while the other two were irradiated. The control capacitor's mica contained about 4 tracks per field viewed at 210 ×; the irradiated capacitor had 24 tracks per field in the areas of the mica that were under the silver and 6 tracks per field in the mica that was not in contact with the silver. No noticeable damage in the electronic performance was observed of the third capacitor following irradiation.

We felt that this experiment was interesting because of the increasingly widespread use of silver-mica capacitors, and because of the possible application of this technique in determining energies and flux present in the event of a nuclear excursion or accident.

VI. SOURCES OF ERROR

The main source of error in using fission tracks is in the recognition and counting the tracks in the samples. Several samples were given identical exposures and were etched for varying lengths of time. It was noted that overdeveloping either the mica or the Mylar resulted in a loss of counts due to the overlapping of the track locations. Underdeveloping the tracks made them difficult to identify and count. Etching within the parameters described earlier normally gives reproducible track counts. We have found it advisable to spot-check a sample as it is being etched to insure that this severe overlapping does not occur.

A second source of error is in having an extremely large number of tracks in a field of view. Calculations should be made prior to irradiation so as to prevent the complete covering of the samples with an uncountable number of tracks.

VIII. ACKNOWLEDGMENTS

The authors would like to thank Mr. H. Wade Patterson and the members of the Health Physics Department for their suggestions and assistance on this project.

We want to acknowledge the assistance of Mr. Frank Miller and Mr. Harold Wollenberg for their help with the geological studies that were made.

Sister Mary Roger, R. S. M., would like to express her thanks to the National Science Foundation for sponsoring her work as part of a Summer Research Participation Program for High School Teachers that was administered by the Lawrence Radiation Laboratory.

This work was done under auspices of the U. S. Atomic Energy Commission.

REFERENCES

1. K. Becker, Photographic, Glass, or Thermoluminescence Dosimetry, Health Physics 12, 955-964 (1966).

2. K. Becker, Nuclear Track Registration in Dosimeter Glasses for Neutron Dosimetry in Mixed Radiation Fields, Health Physics 12, 6 (1966).

3. R. C. Bhatt, Personal Neutron Dosimetry by Fission Fragment Damage in Insulators, Progress Report Biology and Health Physics Div., January 1 to March 31, 1966, AEC of Canada Ltd., Chalk River, Ontario, Report PR-B-69.

4. M. Debeauvais et al., Registration of Fission Fragment Tracks in Several Substances of Their Use in Neutron Detection, Intern. J. Appl. Radiation and Isotopes 15, 289-299 (1964).

5. R. L. Fleischer and P. B. Price, Techniques for Geological Dating of Minerals by Chemical Etching of Fission Fragment Tracks, Research Information Section, General Electric Research Laboratory, Schenectady, New York (The Knolls), Report No. 64-RL-3635M.

6. R. L. Fleischer, P. B. Price, and R. M. Walker, Solid-State Track Detectors: Applications to Nuclear Science and Geophysics, Annual Reviews.

7. R. L. Fleischer, P. B. Price, and R. M. Walker, Tracks of Charged Particles in Solids, Science 149 [3682], 383-93 (1965).

8. G. D. Kerr and T. D. Strickler, The Application of Solid-State Nuclear Track Detectors to the Hurst Threshold Detector System, Health Phys. 12, 8 (1966).

9. P. B. Price and R. M. Walker, Fossil Tracks of Charged Particles in Mica and the Age of Minerals, J. Geophys. Res. 68, 16 (1963).