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SSTR MATERIALS FOR USE IN LIGHT WATER REACTOR-PRESSURE VESSEL SURVEILLANCE EXPOSURES

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"Solid State Track Recorder Materials for Use in Light Water Reactor-Pressure Vessel Surveillance Exposures"

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ABSTRACT

Solid State Track Recorders (SSTR) have been extensively used in reactor neutron dosimetry. Further applications of SSTR in the U. S. Breeder Reactor, Light Water Reactor, and Magnetic Fusion Energy Reactor programs have been planned. Extension of high accuracy SSTR techniques to high fluence irradiations requires careful attention to many experimental details. One very important aspect of this work is the selection of materials that comprise the SSTR.

A variety of solid state track recorder materials have been examined for Light Water Reactor-Pressure Vessel Surveillance applications. Emphasis has been placed on SSTR characteristics which are most relevant to high fluence irradiations. The SSTR materials investigated include eleven different types of muscovite mica from worldwide geographical locations and different commercial suppliers, synthetic and natural quartz crystals, and quartz glass. Activatable impurities are appreciable only in the case of mica SSTR, and relative isotopic impurity concentrations are reported. Fissionable impurity concentrations in mica and quartz are also reported. The properties of mica and quartz SSTR subjected to a high fluence (3×10^{21} n/cm^2), high temperature (800°F) irradiations have been investigated. Recommendations regarding suitable types of mica and quartz are made.

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Some twenty years have elapsed since the discovery of radiation damage tracks in dielectric materials by Young.(1) In his original work, Young demonstrated that a large increase in chemical etch rate existed in the region of damage created by energetic and highly ionizing charged particles such as fission fragments. In spite of this rather profound and pioneering discovery, little if any work was pursued along these lines until well into the decade of the sixties. Since then, however, the use and applications of the track-etch technique have expanded exponentially as is well documented in the recent text of Fleischer, Price and Walker,(2) who are perhaps the most notable contributors to this field of endeavor.

The range and scope of applications for this technique now cover almost the complete breadth of the natrual and physical sciences, as well as certain limited applications in biological science. In nuclear applications, detectors using this technique have become commonly known as Solid State Track Recorders (SSTR). Such detectors offer fundamental advantages for in-core reactor dosimetry, health physics, and environmental science. Judging from recent past history, it is not difficult to predict that the relevance of this measurement method will continue to increase significantly, especially in support of existing nuclear programs of national and even international scope.

Solid State Track Recorders (SSTR) provide a precise highly sensitive technique for absolute fission rate measurments. This method has been used for a broad range of nuclear applications by many workers throughout the world for more than a decade now. The SSTR dosimeters are comprised of a fissionable deposit (or fission fragment source) pressed in firm contact against a suitable transparent dielectric track recorder material, as shown in Figure 1. Any appropriate fissionable nuclide can serve as the fissionable deposit and a broad range of plastic and mineral track recorder materials can be utilized. Fission fragments generated in the SSTR source enter the track recorder material creating radiation damage trails or tracks, which can subsequently be enlarged with suitable etching techniques. As an example, the microphotographs displayed in Figures 2 and 3 are fission tracks observed in mineral recorders, namely mica and quartz crystal, respectively.

Fission rate measurements with SSTR have been placed on a precise absolute basis using optical microscopy and manual scanning techniques.(3) By systematic refinement of the SSTR technique, relative experimental error of absolute fission rate observations has been reduced to a few percent (1-2%) at the 1σ level. This foundation has provided for the accurate application of the SSTR method to a diversity of neutron dosimetry and related experiments as attested to by efforts in the following areas:

1. In-core reactor physics.(4-7)
2. Spontaneous fission half-lives.(8-11)
3. Absolute fast neutron fission yields.(12-14)
4. Health physics and environmental neutron dosimetry.(15-18)

The general success of this technique in the study of fission and neutron related phenomena rests upon the following facts:

1. SSTR are small in size, offering the capability of high spatial resolution in the measurement of heavy charged particle emission, and produce small pertubations when inserted into the medium of interest.

2. SSTR provide time-integrated observations and give a permanent data record that can be evaluated by different data reduction techniques.

3. SSTR have an enormous range of sensitivity, which can be adjusted by the strength or geometry of the source of fission fragments or of alpha particles. The dynamic range of observable track density varies at least from approximately one event/cm^2 to approximately 10^8/cm^2.(2) A comparable dynamic range is also afforded by variation of source strength. These two factors can be applied in consort to provide SSTR techniques with a range of sensitivity that is virtually unmatched.

4. SSTR record events with high efficiency (if desired), and afford absolute measurements, after suitable calibration.(3)

5. SSTR, even in the present state-of-the-art for recording fission fragments, yield high accuracy(3) of approximately 1 to 2% (1σ). Actually differences in routine SSTR measurements of absolute fission rates as compared with the NBS fission chamber have been reported at the 2-3% level.(19) In routine SSTR applications such differences can be expected, since SSTR work at the best state-of-the-art requires attention to many refinements.

6. SSTR yield absolute fission rates independent of the knowledge of any nuclear cross section and of any other neutron standards.

7. SSTR are highly selective in recording the events of interest in a mixed radiation field. Although the influence of high gamma radiation on the track recording characteristics of various SSTR requires further study,(20-22) these are not likely to be sources of concern, except for measurements at high power. Muscovite mica, for example, is so selective in recording fission fragments as against alpha particles, that distinct fission fragment tracks have been observed(11) in a background of 4×10^{13} alpha particles/cm^2, and this limit may be somewhat higher.

8. Some SSTR, such as natural quartz crystals, can be used to record and retain fission fragment tracks at elevated temperatures,(2) thus introducing the possibility of making absolute fission rate measurements in the core of a reactor operating at full power.

9. In many applications the source thickness used with SSTR is so small that neutron flux depression and self-shielding problems are essentially nonexistent.

Actual LWR applications will require dosimetry in a fluence region extending from roughly 10^{16} n/cm^2 up to 10^{19}n/cm^2 and higher. Work with SSTR in such higher fluence irradiations has been very restricted indeed. A program has been initiated at the Hanford Engineering Development Laboratory (HEDL) to extend SSTR techniques to regions of higher fluence for dosimetry measurements in Light Water Reactor, LWR, Breeder Reactor, BR, and Magnetic Fusion Energy Reactor, MFER environments.(23) Although some high fluence observations have been attempted, such as efforts to measure thermal neutron fluence,(24) these investigations have been very limited in scope, detail and results. Because of the paucity of techniques for accurate neutron dosimetry in environments of unquestionable relevance to our national energy programs, the full potential of the SSTR method must be adequately assessed.

For high fluence applicability, many aspects of the SSTR method will have to be quantitatively examined. In particular, the selection of materials which comprise the SSTR for high fluence LWR neutron dosimetry applications is focused upon in this presentation. The SSTR characteristics singled out for special emphasis at high fluence are:

1. Optical quality, such as transparency, surface imperfections, and fossil track density.
2. Bulk etch rates.
3. Track recording properties as a function of neutron fluence.
4. Impurities, such as U or Th, which could lead to unacceptable fission track backgrounds.
5. SSTR constituents and/or impurities which, when activated, could produce unacceptable personnel dose rates.
6. Thermal stability of tracks.
7. Background tracks from neutron scattering recoils (plastic SSTR only).

The significance of these characteristics will be more fully elaborated upon below. Experimental tests based on these criteria are then presented. On this basis, preliminary recommendations are advanced for materials that will afford the most accurate SSTR neutron dosimetry in high fluence LWR applications.

Criteria for Selection of SSTR Materials

There are a variety of materials that can be used as SSTR, each one possessing its own useful range of application. Natural minerals and glasses can be used for fission fragment detection, but not for alpha particles, since they possess too high a radiation damage threshold to form etchable tracks with alpha particles. Plastics, on the other hand, have a much higher sensitivity to etchable damage, and some, such as polycarbonate makrofol E, can be used to form tracks of low energy protons.(25) Since the plastics are composed of light elements such as hydrogen, carbon, and oxygen, tracks produced by recoil nuclei during fast neutron irradiation produce a background if fission fragment detection is desired; on the other hand, these very recoils can be used to give information on neutron-fluence spectra.(26) Plastic SSTR are also more sensitive to thermal effects and to high gamma dose,(20) so that the minerals and glasses are needed to detect fission fragments at elevated temperatures and these SSTR are also preferable in very high gamma fields. High levels of impurities of such elements as Fe, Co, Ni, etc., are also of concern in the application of mineral SSTR for high neutron fluence applications, because of personnel radiation exposure which can accrue in handling activated materials.

The backing materials of fission deposits must also have low impurity levels of actinide elements in order to keep the background of fission fragments from these impurities at a negligible level.

Details concerning the criteria for the selection of suitable SSTR and of suitable fission deposit backing materials are discussed in the remainder of this section, whereas actual data for these characteristics are presented in the section on experimental results.

Optical Quality

Most SSTR materials are illuminated with transmitted light when tracks are manually counted with an optical microscope; the SSTR material must thus be transparent to visible light. The SSTR surface must also be free of imperfections which may be confused with fission or alpha tracks. The surface must also be free of any other kind of defects or imperfections which make the particle tracks difficult to identify and count.

Natural minerals which contain actinide impurities may contain latent "fossil" fission tracks which are revealed on chemical etching. If the density is low, these can be discriminated against by pre-etching the SSTR for a time large compared to the etching time used to reveal the tracks of interest.(3) These fossil tracks are then much larger than the irradiation induced tracks and can be discriminated against. Fossil tracks can also be removed by pre-annealing the SSTR. For Muscovite mica, pre-annealing at 600°C for 6 hours effectively removes the fossil background. However, for good natural quartz crystals, the background of fossil tracks is extremely low so that the pre-annealing is not usually necessary.

In the case of high quality quartz glass, the surface appears free of defects that might be confused with fission fragment tracks; however, when a sample of quartz glass that has not been exposed to fission fragments is etched in hydrofluoric acid, defects are revealed that may be interpreted as fission tracks. In order to avoid this problem, the quartz glass is pre-etched in 49% HF for 5 minutes; the surface density of these defects can then be determined, making possible the pre-irradiation selection of good samples. When etched again for 5 minutes in 49% HF after irradiation by fission fragments, the defects are too large to be confused wth fission tracks.

A mechanically polished natural quartz crystal may appear to be free of optical defects that would be troublesome in fission track counting. Many surface defects may be revealed, however, when etched with HF or sodium hydroxide. A satisfactory procedure for selecting good quartz crystal cut along either the 001 or 100 plane is to etch the surface in 49% HF for 10 minutes at room temperature. This produces a number of shallow triangular pits which can usually be removed by etching the crystal in boiling 65% sodium hydroxide solution for 25 minutes. Good samples are then quite free of defects, and poor samples can be rejected. After this chemical polishing and subsequent exposure to fission fragments and re-etching in the boiling 65% NaOH for another 25 minutes, the optical quality of the quartz crystal is highly satisfactory for reliable fission track counting.

Bulk Etch Rates

Other important parameters that characterize various SSTR are the track etch rate, V_T, and the bulk etch rate, V_G. V_T is the time rate at which the radiation damaged material along the direction of the track is removed by the etching solution, whereas V_G is the corresponding etching rate of the material not damaged by the passage of a heavy charged particle. In order for a track to form, the condition $V_T \sin \Theta > V_G$, must be patisfied, where Θ is the angle between the direction of the charged particle and the surface of the SSTR.(2,23)

For amorphous plastics and glasses, V_G is isotropic. For crystalline minerals such as mica and natural quartz crystals V_G is a function of the direction relative to the various crystalline planes.(27) For muscovite mica, $V_G = 0$ in a direction perpendicular to the cleavage planes. This leads to a high value for the optical efficiency η of mica, which is the ratio of the tracks counted per unit area to the fission fragments incident per unit area. For isotropic incidence of the fragments, η has been measured for mica to be 0.948 ± 0.0053. If the fragments are incident perpendicularly, theoretically $\eta = 1.00$ provided $V_T > V_G$ in a direction perpendicular to the surface.

A microphotograph of tracks produced in mica by fission fragments incident perpendicularly to the cleavage planes is shown in Figure 4. Since the tracks are diamond shaped, it is clear that V_G has different values in different directions in the cleavage planes. The bulk etch rate that can be measured with best sensitivity corresponds to the direction along the larger diagonal of the diamond shaped track. If ℓ is the length of this diagonal, then $V_G = \frac{\ell}{2t}$ where t is the etching time.

The results obtained for V_G in this direction for various samples of mica are shown in a subsequent section. Similar measurements will be carried out for quartz crystals.

Impurities

In a typical neutron spectrum in the ex-vessel cavity beyond the pressure vessel in a LWR, the average fission cross section σ_f for ^{235}U, ^{238}U, ^{237}Np, and ^{232}Th are 4, 0.082, 0.41, and 0.020 barns, respectively. A typical fluence (for the entire spectrum) to which SSTR are expected to be exposed in LWR-PVS irradiations is roughly 2×10^{16} neutrons/cm^2. The mass of deposits of these nuclides can be adjusted so that the total number of fission tracks/cm^2 will be about 2×10^5. This track density represents roughly the upper limit for accurate SSTR measurements employing manual scanning techniques.(3) Consequently, it is desirable to keep the background fission tracks from all sources (impurities in the SSTR and backing material) to less than 1%, or $<2 \times 10^3$ tracks/cm^2. If fissions from the backing material are for the moment neglected, an upper limt of uranium impurity in SSTR can be estimated as follows.

It has been shown that the effective "asymptotic" sensitivity(28) is approximately one-half the average range R of fission fragments in mica; $\overline{R}$ is about 14 μm. If B is the background tracks per cm^2, and if N is the number of atoms of uranium per cm^3, then

$$N = \frac{B}{(.0072\ \bar{\sigma}_5 + 0.993\ \bar{\sigma}_8)\left(\frac{\bar{R}}{2}\right)(\phi t)}, \qquad (1)$$

where σ_5 and σ_8 are the spectrum average fission cross sections for ^{235}U and ^{238}U respectively, ϕt is the total neutron fluence, and the factors .0072 and 0.993 are the atomic abundances of ^{235}U and ^{238}U in natural uranium, respectively.

Substituting in the above numbers, one finds $N \cong 1 \times 10^{15}/cm^3$. Since a typical density of mica is 2.77 g/cm^3, this uranium impurity limit is 1.8×10^{-7} g uranium/g mica, or 0.18 ppm.

In addition to intrinsic impurities in the SSTR and fission deposit backing material, the surface of the mica or other SSTR must be clean to give no more than the background track density calculated above. The mass of a deposit of a given fissionable nuclide can then be adjusted to give the desired track density for each spectrum/fluence combination. For fluences lower than 2×10^{16} n/cm^2, the fission deposits have greater mass and the background requirements are not so stringent. It is for higher fluence, extending to 10^{21} n/cm^2 and above, that severe requirements are placed upon both fission deposit preparation and background track density requirements.

After exposure it is necessary to disassemble the SSTR, etch them, and count the tracks; consequently, radiation safety guidelines place an upper limit on the concentration of impurity atoms that SSTR should contain.

Thermal Stability of Tracks

When SSTR are heated to elevated temperatures, radiation damage resulting from the passage of heavy charged particles may be annealed so that some fraction of the latent tracks do not etch. Such annealing can therefore result in a loss in efficiency; in fact, if a given SSTR material is heated to a sufficiently high temperature for a long enough time, the damage may be sufficiently removed so that no tracks prove etchable. A summary of some of the annealing characteristics of various SSTR is given in the recent treatise on track recorders.(2) Since more detailed information is needed for the materials to be used in the LWR-PVS program, a program to determine the annealing characteristics of mica and quartz is underway at HEDL. Naturally, the safest procedure would be to define a maximum temperature and associated maximum heating time for a given SSTR, so that corrections due to annealing effects will be very small or even unnecessary.

Track Recording Properties as a Function of Neutron and Gamma Fluence

Not only are the track recording properties of SSTR affected by temperature; exposures to high fluences of neutrons and/or gamma-rays also are known to produce changes in the ratio of the track etch rate to the bulk etch rate.(29) For this reason, experiments are underway at HEDL to study these effects. An intense gamma-ray facility containing 2.6 x 10^5 curies of ^{60}Co is available to study the effect of high gamma-ray exposures, and SSTR are being exposed to high fluences of neutrons and gamma-rays in the Experimental Breeder Reactor II (EBR-II) and in the Oak Ridge Research Reactor (ORR). Effects on the bulk etch rate of natural quartz crystals have been observed in a recent exposure in EBR-II to a neutron fluence of 3 x 10^{21} n/cm^2. Further experiments are currently in progress in both EBR-II and ORR. Fortunately, the fluence levels anticipated in LWR-PVS irradiations are not expected to present problems for mineral or glass fission fragment SSTR. However, the use of plastic SSTR for the detection of alpha particles or protons needs to be carefully evaluated.

Neutron Induced Recoil in SSTR

Recoils resulting from fast neutron scattering are not a problem for LWR-PVS exposures of fission fragment SSTR, but they present a very serious problem for plastics. For personnel dosimetry, however, this problem has been turned into an advantage, in that these recoils are now used to measure exposures to fast neutrons.(26) Nevertheless, in using plastic SSTR to measure alpha particle production from various nuclides, care must be used to distinguish recoils from the alpha particles. This is possible by a combination of range and track diameter measurements,(25) but the technique needs further study, and the accuracy that can be obtained has yet to be established.

Experimental Results and Discussions

Optical and "Fossil" Fission Track Density

Each of the eleven types of mica has been scanned to determine the density of fossil fission tracks. The fossil track densities for the different types of mica are shown in Table I. In general, the African, South African, and Angolan micas have high fossil track densities and also poor optical quality. The Brazilian and Indian micas have the lowest fossil track densities and also have good optical quality.

Bulk Etch Rates

The value of V_G previously described has been measured for two disks of India Ruby mica punched from a piece supplied by Asheville-Schoonmaker Co. The mica was exposed in a vacuum chamber to ^{252}Cf fission fragments incident perpendicular to the cleavage planes. The mica was etched in 49.2% hydrofluoric acid at a temperature of $(22.7 \pm 0.1)°C$ for 1.00 hour under gentle agitation. A digitized 10x filar micrometer attached to a LKE Nikon microscope was used to measure ℓ, the larger diagonal of the diamond shaped tracks. A 100x oil immersion lens and cover glass were used to provide high magnification. Both sides of the mica were exposed to the fission fragments to see if V_G was the same on both sides. The large diagonal ℓ was measured for 20 tracks on each side for both samples, so that a total of 80 measurements were made. No difference in V_G was found for the two sides and for the two disks of mica. The mica was then etched for three additional 30 minute periods, and the values of ℓ were measured again after each 30 minute period.

The results are shown in Table II. As expected, V_G is found to be independent of time in the time range measured as attested to by the linear least squares fit shown in Figure 5. Improved experimental error for longer etching times may result primarily from the improved precision in the measurements of when it is larger, but they may also reflected statistical fluctuations in V_G which decrease as ℓ increases.

The measurement of V_G as described above can be used to select the best etching time at a given etching temperature to bring tracks up to a selected standard size. A disk of the mica punched from the same piece being used in a given experiment can be exposed to perpendicularly incident fission fragments and etched for about one hour with disks used in the experiment. Measurements of ℓ in the control sample can be used to determine how much longer the samples should be etched to give tracks of the selected standard size.

In the etching procedure, the mica samples are placed vertically in the etching bath, with the surface fully exposed to the etchant. A group of mica disks punched from samples from various origins and suppliers were exposed in a vacuum chamber to fission fragments from ^{252}Cf incident perpendicular to cleavage planes. All were etched simultaneously in 49.2% HF for 90.0 minutes at $(22.7 \pm 0.1)^{\circ}$C. The maximum bulk etch rates in the cleavage planes were measured as before. The results are shown in Table III. It can be seen that the measured V_G varies over a factor of two for the types of mica studied.

Activatable Impurities

Exposure to high fluences will result in neutron activation of the SSTR and the radiation dose rate to be encountered when processing the SSTR must be considered. Mica can be represented by the chemical formula $KAl_2Si_3AlO_{10}(OH,F)_2$ and quartz by SiO_2. None of the elements represented by these formulas are converted to long-lived (1 day) activities as the result of neutron irradiation. However, trace impurities such as iron, rubidium, and cobalt can result in long-lived nuclides and an accompanying radiation dose. Samples of the eleven different types of mica from all over the world were subjected to a total fluence of 5 x 10^{18} neutrons cm/^{2} in order to determine the content of trace activatable impurities. The result of these analyses are shown in Table IV. In all cases the major activatable element present was iron, which results in the long-lived nuclides ^{59}Fe (45.6d) and ^{54}Mn (303d). Other elements, although detectable, result in a negligible contribution to the total dose. For all of the micas studied, the dose rate at contact for a 4 cm^{2} piece of mica (approximately 0.025 cm thick) was less than 1 mrem/hr measured two weeks after the end of the irradiation. In a separate experiment, mica and quartz SSTR were irradiated to a fluence of 3.4 x 10^{21} neutrons/cm^{2} over a period of 1.5 months and analyzed after 7 months. The mica samples studied all had dose rates less than 1 mrem/hr at contact and the quartz crystal and quartz glass SSTR were less than 0.1 mrem/hr.

It can be concluded on the basis of these results, that quartz crystals, quartz glass, and all of the types of mica result in acceptable dose levels with respect to personnel radiation safety, even after high fluence irradiations, provided that sufficient time (1-2 weeks) is allowed for the shorter-lived nuclides to decay. In the case of quartz crystals, the extremely high natural purity of this mineral is a very valuable characteristic for high fluence applications.

Fissionable Impurities

The eleven types of mica that were irradiated to a fluence of 5 x 10^{18} neutrons/cm^{2} were recleaved (to remove tracks produced on the surface by handling impurities), and after etching for 90 minutes in 48% HF were scanned for fission tracks along the freshly recleaved interior surfaces. Fission track densities were converted to uranium concentrations by assuming that all fission tracks originated from fission of the ^{235}U atoms in

Uranium of natural isotopic abundance. This is a reasonable assumption, since the irradiation was carried out in a highly thermalized neutron flux. The results of this analysis are shown in Table V. All of these values are well below the 1.8×10^{-7} g uranium/g mica limit, which provides less than 1% correction to the fission rate, as has already been established for an irradiation fluence of roughly 2×10^{16} n/cm^2. In fact, the Reliance South African mica, containing the highest concentration of uranium of the types studied, would result in a clearly negligible correction of 6×10^{-3}% to the fission rate in a typical LWR ex-vessel cavity exposure.

In other LWR-PVS exposures where fluences can be at least three orders of magnitude higher, background track density could be as large as roughly 6 percent for the poorest mica candidate, namely South African Mica from Reliance. Employing the better mica candidate materials listed in Table V would afford a reduction in background track density of roughly one order of magnitude. Consequently background track density due to impurities in mica can be rendered negligible for all LWR-PVS applications.

The solubility of uranium in quartz is known to be very low so that one can reasonably expect to find all quartz samples to have uranium concentrations in the sub parts-per-billion range, leading to corrections to the fission rate well below 1% for all anticipated LWR fluence irradiations.

Etching Properties - Fluence Dependence

In order to maintain better than 1% accuracy, the etching properties of the SSTR material must be well characterized so that the etching efficiency will be accurately known. For low fluence applications, mica has been shown to have a zero bulk etch rate(3) due to the fact that no etching occurs perpendicular to the cleavage planes. This may not be the case for higher fluence applications. Mica subjected to a total fluence of 3.4×10^{21} neutrons/cm^2 at a temperature of 433°C underwent dehydration, although the temperature at which this occurs under non-irradiation conditions is slightly less than 600°C.

The track recording properties of the mica were lost due to this change in optical quality and the markedly increased etch rate of the dehydrated mica compared to normal hydrated mica.

Quartz crystal, even when unirradiated, has a measurable bulk etch rate. The rate of etch depends on the cleavage plane being attacked and has been measured to be 10.8 ± 3.4 μ/hr for the (001) plane and 2.7 ± 0.5 /hr for the (100) plane in boiling 65% sodium hydroxide. The optical registration efficiencies, i.e. η, were found to be almost the same for these two planes. These results are in disagreement with the measurements of Khan and Ahmad who claimed a zero registration efficiency for the 001 plane.(27) When irradiated to a fast neutron fluence of 3 x 10^{21} neutrons/cm^2, the bulk etch rate increased to 11 ± 2 μm/hr, 18 ± 3.5 μm/hr. and 21 ± 4 m/hr along the three crystal axes (the orientation of the crystal was unknown in this experiment). Since the bulk etch rate of quartz crystal is fluence dependent at these high fluences, further calibration experiments must be performed in order to characterize the etching efficiency of quartz crystal SSTR.

Annealing Studies

Very limited effort has been undertaken to study annealing effects in SSTR. Most investigations that do exist are not concerned with reactor neutron dosimetry applications. Rather, they deal with determination of corrections for the fission track dating technique due to annealing effects in ancient specimens. The work of Storzer and Wagner(30) represents the most comprehensive of such studies.

As our program at HEDL to examine annealing effects in SSTR has only just been initiated, only preliminary results can be presented. Table VI present the effect of heating quartz glass SSTR in the region of 300-400°C for up to 8 hours. All of these quartz glass SSTR were prepared by surface contact irradiation with a ^{244}Cm spontaneous fission source. Irradiation with this source for equal time intervals produced specimens of equal fission track density within the statistical limits of these exposures (which was roughly 5%). As can be seen in Table VI, annealing not only decreases fission track density systematically, but SSTR size also decreases continuously.

It must be stressed that this data on quartz glass has been introduced as only a representative example of the annealing studies that will be required. Actually mica possesses vastly improved track retention characteristics compared with quartz glass. Fortunately, natural quartz crystal is even that much better than mica in this regard.

It has become customary to describe this annealing process with reaction rate theory applicable to chemical kinetics.(30) In most instances, simple Arrhenius equations are employed and experimenters attempt to deduce an "activation energy" corresponding to such a model of the annealing process. However, models based upon chemical reaction rate theory cannot be applicable here. It is clear from Table VI, that not only does annealing alter track density, but dramatically effects the very nature of the individual tracks themselves. Hence, rate constants cannot be introduced to describe entities such as tracks, which are themselves changing with time and temperature during the annealing process. Consequently, fundamental revisions are necessary to provide proper analysis of track annealing in SSTR.

Conclusion

Mica SSTR have been shown to have suitable properties for use in LWR-PVS exposures. High fluence irradiations result in negligible personnel dose rates during scanning for all the types of mica studied. Background track densities due to the fissionable uranium and thorium impurities are also negligible for all the types of mica studied. To date, no limitations have been uncovered which would restrict either the utility or accuracy of SSTR neutron desimetry for LWR-PVS applications.

The bulk etch rates of the different types of mica studied ranged over a factor of two. It is interesting to note the rough correlation between the bulk etch rate values of Table III and the fossil track density results of Table I and also with the uranium concentrations of Table V. No similar correlation is evident with the other trace elements observed (Table IV). A possible explanation is that the bulk etch rate is related to the stored radiation damage from the decay of the uranium impurities over geological times. A more vigorous assessment of these characteristics is currently being undertaken.

At extremely high fluences and temperatures, natural quartz crystal is a more suitable SSTR due to its higher annealing temperature and resistance to radiation damage. The inherently lower impurity concentrations of quartz crystal also lead to acceptable fission track backgrounds and dose rates at higher fluences. The bulk etch rate of quartz has been found to be fluence dependent, however, and calibration experiments must be performed to determine the etching efficiency of quartz for high fluence applications.

More data on the annealing properties of both quartz and mica are needed, and the current theories on annealing mechanisms must be subjected to more rigorous scrutiny. Such experiments are currently in progress.

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TABLE I

FOSSIL FISSION TRACK DENSITIES IN MICA SAMPLES FROM VARIOUS ORIGINS AND SUPPLIERS

Source	Supplier	Fossil Tracks/cm^2
India	United Mineral	28 ± 5
India	Asheville-Schoonmaker	38 ± 5
India	Perfection	78 ± 8
India	Reliance	79 ± 8
Brazil	United Mineral	86 ± 8
Angola	Asheville-Schoonmaker	115 ± 11
U.S.A.(Domestic)	Asheville-Schoonmaker	118 ± 12
Brazil	Asheville-Schoonmaker	121 ± 12
Africa	Asheville-Schoonmaker	143 ± 14
Brazil	United Mineral	193* ± 17
U.S.A.(Domestic)	Reliance	260 ± 26
Brazil	Reliance	278 ± 27
South Africa	Reliance	2680 ± 150

*The two United Mineral samples from Brazil were obtained at different times.

TABLE II

MAXIMUM BULK ETCH RATE* IN THE CLEAVAGE PLANE OF A SAMPLE OF INDIA RUBY MICA SUPPLIED BY ASHEVILLE-SCHOONMAKER

Etching Time (hr)	ℓ(um)	V_g(μm/hr)†
1.00	4.22 ± 0.13	2.11 ± 0.06
1.50	6.40 ± 0.15	2.13 ± 0.05
2.00	8.62 ± 0.11	2.15 ± 0.03
2.50	10.78 ± 0.12	2.16 ± 0.02

*Mica etched in 49.2% HF at (22.7 ± 0.1)°C.

†The experimental error is the standard deviation in the measurement of ℓ for 80 tracks, except for the 2.50 hour etch; in this latter case ℓ was measured for only 40 tracks.

TABLE III

MAXIMUM BULK ETCH RATE IN THE CLEAVAGE PLANS OF MICA SAMPLES FROM DIFFERENT ORIGINS AND SUPPLIERS

Mica Source (Origin)	Supplier	Density** g/cm^3	Number of Tracks* for which was measured	V_G (μm/hour)
India	United Mineral	2.73	20	1.97 ± 0.04
U.S.A. (Domestic)	Reliance	2.68	40	2.36 ± 0.05
India	Ashville-Schoonmaker	2.71	20	2.38 ± 0.03***
India	Perfection	2.74	40	2.51 ± 0.06
Brazil	Ashville-Schoonmaker	2.76	20	2.59 ± 0.05
U.S.A. (Domestic)	Ashville-Schoonmaker	2.75	40	2.66 ± 0.04
Brazil	United Mineral	2.68	40	2.67 ± 0.07
India	Reliance	2.79	40	2.83 ± 0.08
Brazil	United Mineral	2.68	20	2.90 ± 0.04
Africa	Ashville-Schoonmaker	2.78	40	2.94 ± 0.07
Angola	Ashville-Schoonmaker	2.75	40	3.36 ± 0.07
South Africa	Reliance	2.79	20	4.70 ± 0.08

The samples are arranged in the order of increasing V_G.

* When ℓ was measured 20 times, it was for tracks in the near vicinity of each other in a given mica disk. When measured 40 times, it was for 20 tracks in each of two mica disks punched from the same sample. If anything, the standard deviation is greater for the 40 measurements probably resulting from changes in V_G for the two different regions.

** The relative accuracy of these measurements is about one percent.

*** Note in Table II, V_G for mica from India supplied by Ashville-Schoonmaker was measured to be 2.13 ± 0.05 m/hr for a 90 minute etch. The two mica samples were cut from different pieces and etched at different times. The temperature of the etch both was the same within 0.1°C, but the concentration of the HF was not measured. It was nominally 49.2% in both etch baths.

TABLE IV

NEUTRON ACTIVATION ANALYSIS RESULTS FOR MICA SAMPLES FROM VARIOUS ORIGINS AND SUPPLIERS

Mica Source (Origin)	Supplier	Fe(g/g mica)	Co(g/g mica)	RG(g/g mica)	Ta(g/g mica)	Sc(g/g mica)	Cs(g/g mica)
U.S.A. (Domestic)	Reliance	3850 ± 1450	0.605 ± 0.018	230 ± 7	5.4 ± 0.1	1.42 ± 0.02	10.6 ± 0.4
India	Ashville-Schoonmaker	2270	0.08	239	5.86	1.97	10.2
India	Perfection	5615 ± 742	0.38 ± 0.28	250 ± 6	7.2 ± 0.3	4.5 ± 3.5	13 ± 2
Brazil	Ashville-Schoonmaker	9875 ± 714	0.17	889 ± 6	44.0 ± 0.8	3.4 ± 0.2	29.0 ± 0.7
U.S.A. (Domestic)	Ashville-Schoonmaker	9161	0.56	229	1.47	65.3	1.0
Brazil	United Mineral	9360	2.9	400	22	15.8	10
India	Reliance	9270 ± 3150	0.7 ± 0.2	299	14.9	18	12
Africa	Ashville-Schoonmaker	6250 ± 2350	0.80 ± 0.07	271 ± 23	6.7 ± 0.4	2.1 ± 0.5	2.1 ± 0.2
Angola	Ashville-Schoonmaker	8553 ± 93	0.82 ± 0.02	258 ± 10	6.7 ± 0.3	2.2 ± 0.2	2.0 ± 0.2
South Africa	Reliance	1783 ± 18	2.2 ± 0.2	342 ± 5	8.5 ± 0.3	7.6 ± 0.4	1.8 ± 0.1
Brazil	Reliance	7260 ± 155	0.8 ± 0.7	253 ± 6	270 ± 2	3.94 ± 0.04	2.4 ± 0.2

†Impurities are given in terms of microgram of element per gram of mica. Overall uncertainty in the neutron flux results in an error of ± 20%. The error when given represents the range of variation of two separate determinations. Where no error is given, only one sample was analyzed.

TABLE V

URANIUM CONTENT OF MUSCOVITE MICA SSTR MATERIALS BASED ON FISSION TRACK ANALYSIS

Origin	Supplier	Uranium Content* (g U/g mica)x10^{10}
Domestic (USA)	Reliance	1.0
India	Ashville-Schoonmaker	1.1
India	Reliance	2.1
India	Perfection	2.1
Brazil	United Mineral	2.2
Brazil	Ashville-Schoonmaker	2.6
Brazil	United Mineral	3.2
Brazil	United Mineral	3.6
Brazil	United Mineral	3.7
Brazil	United Mineral	4.2
Africa	Ashville-Schoonmaker	4.4
Angola	Ashville-Schoonmaker	5.5
Angola	Ashville-Schoonmaker	5.7
Domestic (USA)	Ashville-Schoonmaker	6.8
South Africa	Reliance	11

*The statistical accuracy of these observations is about 10 percent (1σ), whereas the absolute accuracy is roughly 20 percent (1σ).

TABLE VI

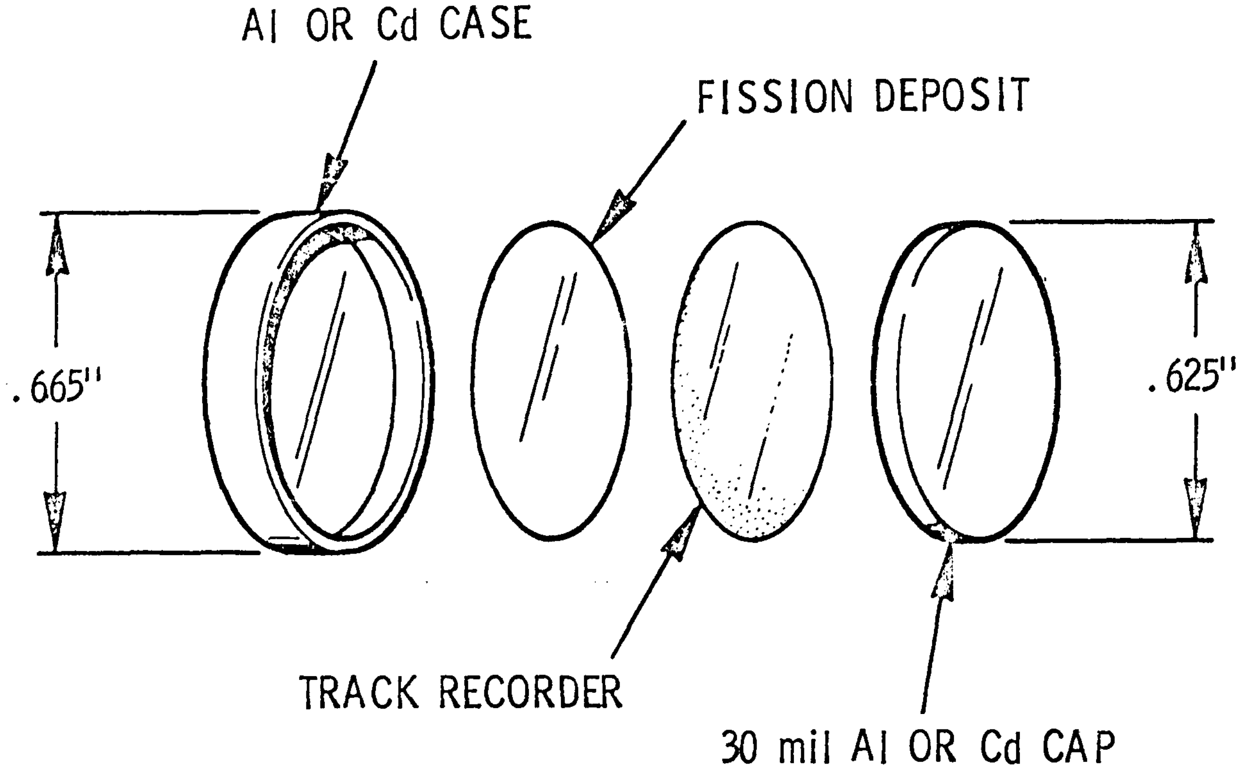
PRELIMINARY TRACK ANNEALING DATA ON QUARTZ GLASS†

Annealing Temperature	Annealing Time (hrs)	Tracks Counted**	Diameter of Circular* Tracks (μm)	Fraction of Tracks Remaining	Ratio of Annealed to Unannealed Diameter
300°C	0	684	8.5 ± 0.5	1.00	1.00
	1.15	550	8.0 ± 0.9	0.80 ± 0.05	0.94 ± 0.06
	2.0	505	7.7 ± 0.3	0.74 ± 0.04	0.90 ± 0.06
	4.5	506	6.8 ± 0.6	0.74 ± 0.04	0.80 ± 0.08
	8.33	423	6.4 ± 0.5	0.62 ± 0.04	0.75 ± 0.07
350°C	0	625	9.2 ± 0.4	1.00	1.00
	0.50	540	8.2 ± 0.4	0.86 ± 0.05	0.89 ± 0.06
	1.0	478	7.6 ± 0.4	0.76 ± 0.05	0.83 ± 0.06
	2.0	466	7.2 ± 0.5	0.75 ± 0.05	0.78 ± 0.06
	4.0	367	5.9 ± 0.7	0.59 ± 0.04	0.64 ± 0.08
	8.0	291	5.8 ± 0.5	0.47 ± 0.03	0.63 ± 0.06
402°C	0	723	9.0 ± 0.4	1.00	1.00
	1.0	391	5.4 ± 0.5	0.54 ± 0.03	0.60 ± 0.06
	2.22	252	4.9 ± 0.5	0.35 ± 0.03	0.54 ± 0.06
	4.0	252	4.7 ± 0.3	0.35 ± 0.03	0.52 ± 0.04
	8.0	196	3.9 ± 0.5	0.27 ± 0.02	0.43 ± 0.06

† Supplied by Amersil, Inc.

* The error is the standard deviation of ten track diameter measurements in each sample.

** All samples of quartz glass were exposed for one hour in close contact with a thin ^{244}Cm source. For a given annealing temperature, all samples were etched together in 48% hydrofluoric acid at room temperature.



HEDL 7805-196.1

FIGURE 1. Typical Configuration Used for SSTR Neutron Dosimetry.

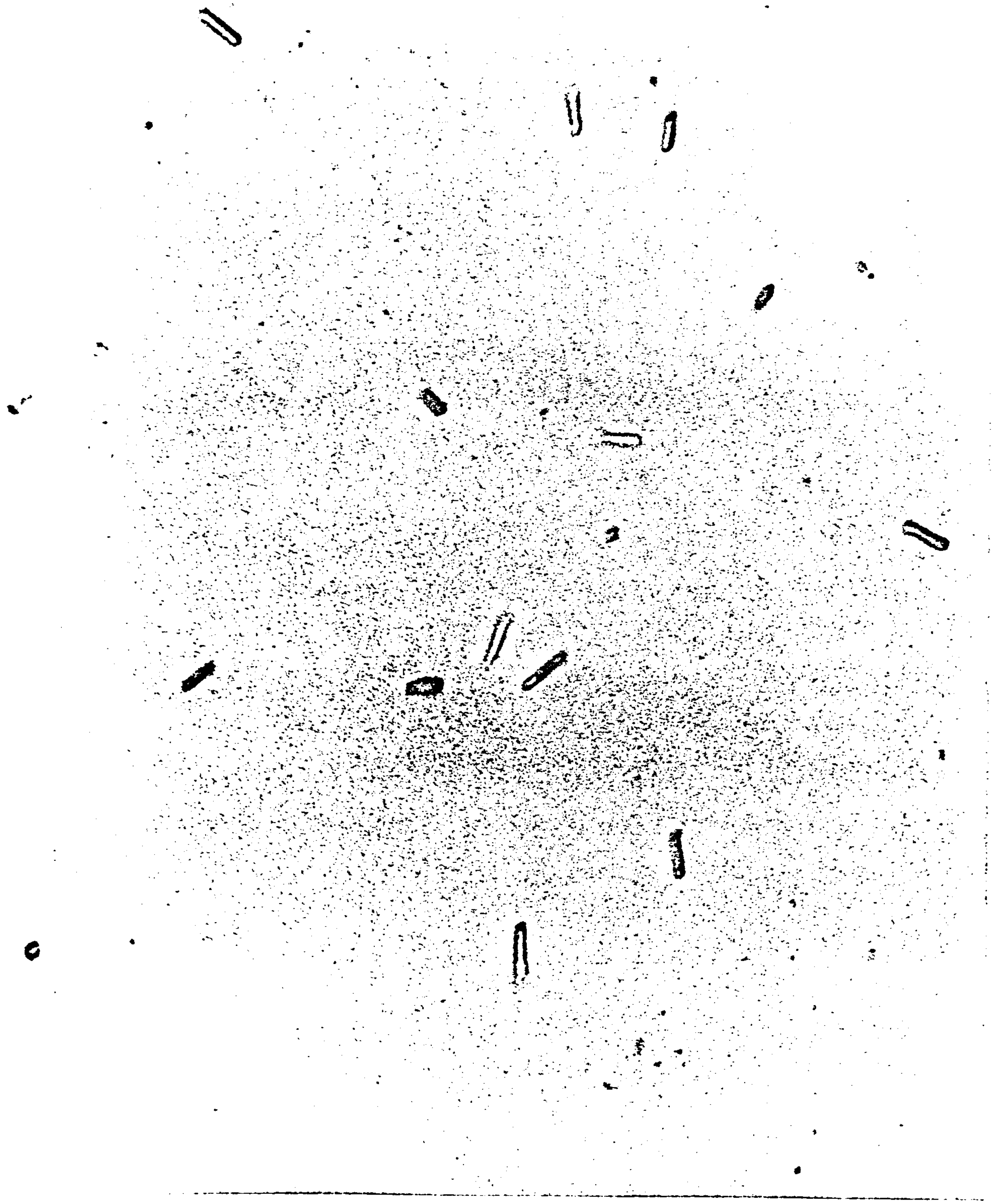


FIGURE 2. Microphotograph of Fission Fragment Tracks Observed in Mica.

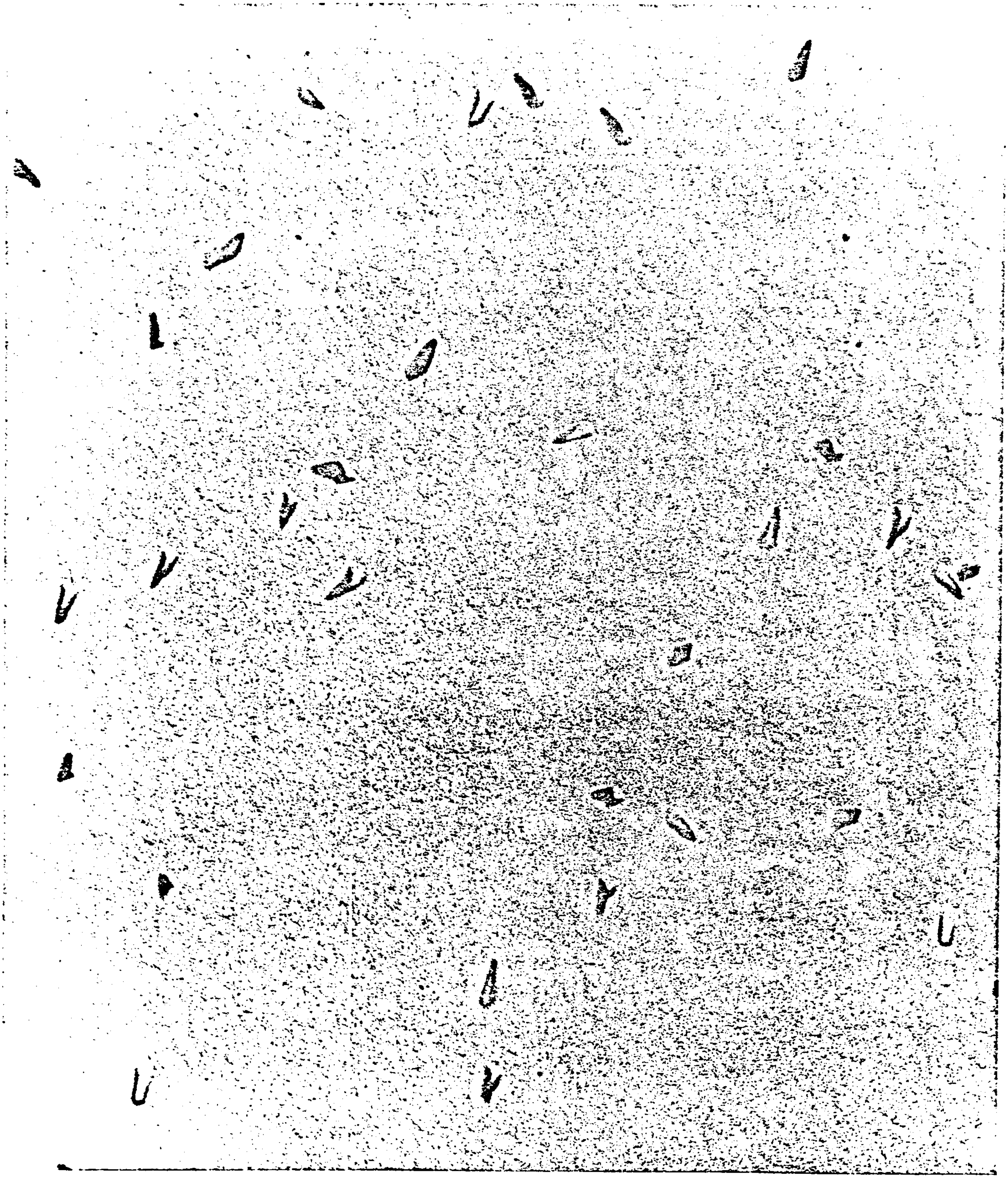


FIGURE 3. Microphotograph of Fission Fragment Tracks Observed in Natural Quartz Crystal (001 Plane).

FIGURE 4. Fission Tracks in India Ruby Mica Supplied by Asheville-Schoonmaker. The fission fragments entered normal to the surface. Etching was in 49.2% HF at 22.7°C for 2.50 hours. The average length of the longer diagonal (as observed from 40 tracks) was found to be (10.78 ± 0.12)μm.

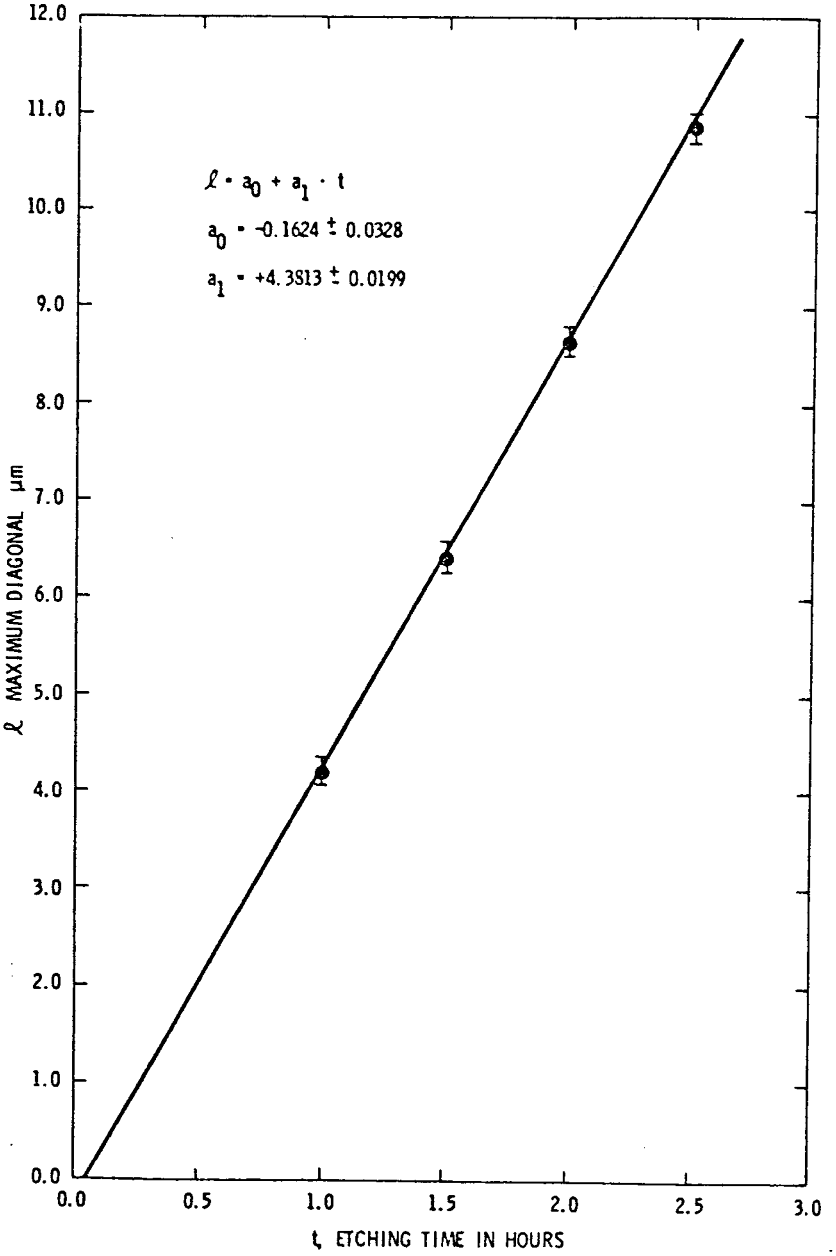


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Figure 5. The dependence of the larger diagonal, ℓ, of fission tracks in mica upon the etching time t. The smooth curve is a linear least squares fit of the experimental data.