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ABSTRACT

In order to use Solid State Track Recorders (SSTR) in environments at elevated temperatures, it is necessary to know the thermal annealing characteristics of various types of SSTR. For applications in the nuclear energy program, the principal interest is focused upon the annealing of fission tracks in muscovite mica and in quartz. Data showing correlations between changes in track diameters and track densities as a function of annealing time and temperature will be presented for Amersil quartz glass. Similar data showing changes in track lengths and in track densities will be presented for mica. Time-temperature regions will be defined where muscovite mica can be accurately applied with negligible correction for thermal annealing

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INTRODUCTION

Whereas most thermal annealing studies in solid state track recorders (SSTR) have been undertaken because of the importance of this effect in fission track dating,[(1)] the research reported in this paper is being pursued to determine the conditions for which selected SSTR can be used to measure fission rates at elevated temperature such as exist in nuclear reactors. Thermal annealing of unetched tracks causes radiation damage sites to be removed or modified, resulting in the reduction of etch rates along the damage trails. The reduced track etch rates cause some tracks to be difficult to observe and may cause some tracks to be reduced below the optical threshold for observation. It is thus important to make corrections for track losses due to annealing, or at the very least to define a time-temperature region for which no track losses occur. Of primary interest are tracks produced when a fission source thin relative to the range of fission fragments is placed in direct contact with the SSTR.

SSTR materials selected for these studies consist of muscovite mica* from India, natural Brazilian quartz crystals, and Supra II quartz glass produced by Amersil Corp.† The etching characteristics of these SSTR materials are quite different. Mica and crystalline quartz are very anisotropic, in contrast to quartz glass, for which track and bulk etch rates are isotropic.

EXPERIMENTAL PROCEDURES

To obtain full energy fission fragments incident isotropically on the surfaces of the SSTR, thin sources of ^{244}Cm or ^{252}Cf were placed in direct contact with the surfaces. Exposures were controlled so that timing uncertainties were <1% from sample to sample. For exposures to fragments incident at 45° or at 90° to the SSTR surface, the SSTR were placed in a vacuum chamber designed to expose several samples simultaneously.

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* The mica used was India Ruby mica supplied by United Mineral Corp.

† (Address)

Exposed SSTR of quartz glass and mica were placed between sheets of mica in metal capsules. The capsules were then inserted into a furnace in which the temperature was controlled to approximately ± 1°C. Annealing studies were first carried out with the quartz glass. The samples were first pre-etched for 5 minutes in 49.2% HF at room temperature, so that pre-selection of good samples was possible. The imperfections that were present in selected samples were easy to distinguish from fission fragment tracks. The samples were then exposed isotropically to full energy fission fragments from ^{244}Cm. Each sample was then heated for a different time period at a particular temperature. The heated samples, along with an unheated sample, were then etched for 5.0 minutes at 22°C in the HF. Fission tracks were counted manually with an optical microscope, and the track count in the heated samples could be compared with that in the unheated samples to determine track loss due to annealing. The diameters of circular tracks were measured with a filar micrometer eyepiece.

In the case of mica, selected samples were exposed to isotropically incident fission fragments from thin sources of ^{244}Cm or ^{252}Cf. Additional samples were also exposed in a vacuum chamber to normally incident fragments and to fragments incident at 45°. After heating, the samples exposed to normal and 45° incident fragments (along with an unheated control sample) were etched from 15 to 30 minutes in 49.2% HF at 23.0 ± 0.3°C. The samples exposed isotropically were etched 90 minutes* in the same etching solution and at the same temperature. Projected track lengths were measured with a digitized filar micrometer eyepiece.

DISCUSSION AND INTERPRETATION OF RESULTS

1. Quartz Glass

   In the case of quartz glass, track-etch kinetics similar to those used by Somogyi and Nagy[2] applied to problems in fission track dating can be used. In our case, however, analysis is simpler, since all tracks originate at the surface of the SSTR. The same procedures cannot be readily applied to anisotropic crystals such as mica and natural quartz.

*Many of the samples were first etched for 30 minutes, so that the range of "flat" tracks could be measured, and then etched for an additional 60 minutes prior to counting.

V_G, the bulk etch rate for the quartz glass was measured to be (1.17 ± 0.10) μm/min in 49.2% HF at 22°C. The efficiency, η, for track registration in unheated samples from the batch used at isotropic incidence was found to be (70.4 ± 1.0)%. It can be readily shown that $\eta = 1 - V_G/\langle V_T\rangle$, where $\langle V_T\rangle$ is the average track etch rate. If $\langle V_T\rangle$ is modified by heating to a value $\langle V_T'\rangle$ the corresponding fraction f of tracks counted is given by $f = 1 - V_G/\langle V_T'\rangle$. Tracks making an angle $\leq \theta_c$, where $\sin\theta_c = V_G/\langle V_T\rangle$ do not register. For normal incidence, $\tan\theta = \dfrac{R}{(\langle V_T\rangle - V_G)t}$, where θ is the cone angle, R the track radius, and t the etching time. Solving for R, we have $R = V_G\left(\dfrac{\langle V_T\rangle}{V_G} - 1\right) t \tan\theta$.

Since $\tan\theta = \dfrac{1}{\left[\left(\dfrac{\langle V_T\rangle}{V_G}\right)^2 - 1\right]^{1/2}}$, we have for the track diameter

$$D = 2R = \frac{2V_G\left(\dfrac{\langle V_T\rangle}{V_G} - 1\right)t}{\left[\left(\dfrac{\langle V_T\rangle}{V_G}\right)^2 - 1\right]^{1/2}} . \qquad (1)$$

The same formula applies for the heated sample, except that $\langle V_T\rangle$ becomes $\langle V_T'\rangle$ and D becomes D', the diameter of the normally incident tracks in the heated sample. Since $f = 1 - \dfrac{V_G}{\langle V_T'\rangle}$, we can calculate D' from the measured f and V_G, and compare it with the measured D'.

Also if D and D' are measured in samples etched in the same way and if η has been measured one can readily show that $f = \dfrac{2\mu}{1+\mu}$, where $\mu = \dfrac{\eta}{2-\eta}\left(\dfrac{D'}{D}\right)^2$.

The experimental results and the calculated values of $\langle V_T\rangle/V_G$ and of D or D' and of f are all shown in Table I. Most of the results demonstrate the correctness of the model, within the limits of the accuracy of the measurements. The track registration efficiencies in quartz glass as a function of heating time for various annealing temperatures are shown in Figure 1.

2. India Ruby Muscovite Mica

Since track etch kinetics are greatly complicated by anisotropies in the bulk etch and track etch rates for crystalline SSTR such as mica, the method used for quartz glass does not apply. Therefore, an empirical approach has been developed to determine the time-temperature region in which no annealing corrections are needed for fission track registration when mica SSTR are exposed to isotropically incident full energy fragments.

Fission fragments incident normally to the cleavage planes in mica produce diamond shaped etch pits when etched in HF, indicating that the bulk etch rate parallel to the cleavage planes is highly anisotropic. The bulk etch rate perpendicular to these planes is essentially zero. Since charge pickup causes the rate of energy loss in fission fragments to decrease when the fragments slow down, it is expected that the damage site density will be least at the ends of the tracks. Thus, the tracks will anneal out first at the end and tend to shrink in etchable length as the annealing proceeds.

The empirical method selected was to relate the length of selected tracks to the registration efficiency for isotropic incidence. One method consisted of measuring the projected length of 45° incident fission fragment tracks in samples which had the same annealing history as samples exposed isotropically. The samples exposed at 45° were etched in 49.2% HF at 22.7°C for periods from 15-30 minutes. Another method consisted of etching the sample exposed isotropically for 30 minutes, and then measuring the length distribution of "flat" tracks; the sample was then etched an additional 60 minutes before the tracks were counted. For both cases, the average projected track length, $<\ell_o>$, in an unannealed sample and the average projected track length, $<\ell>$, in an annealed sample etched in the same way can be used to calculate the ratio $<\ell>/<\ell_o>$. One can also count the tracks N_o in the unannealed sample and N in the annealed sample to determine N/N_o.

A plot of N/N_o vs. $<\ell>/<\ell_o>$ for given etching conditions yields the desired empirical relation which can be used to obtain annealing corrections. For mica, such curves are shown in Figure 2. Note that these curves are composites obtained from different time-temperature histories, rather than a single isothermal annealing experiment. Such plots therefore. support the contention that the registration efficiency is simply a geometric function of track size or track size distribution. It is clear that no annealing correction is required for "flat" tracks in the region $\ell/_{<\ell o>} \geq 0.3$. No corrections have been made for the contribution to track lengths due to the bulk etch rate.

Of further interest is the relationship between $<\ell>/<\ell_o>$ and annealing time for each annealing temperature. These curves are shown in Figures 3 and 4. Note the large change in $<\ell>/<\ell_o>$ for short annealing times (<8 hours) between 460° and 500°C. This suggests a high density of states (well depths) in the region $kT \approx 7 \times 10^{-2}$eV. Investigations are underway to see if a more basic model for annealing based on quantum statistics can be developed, but there are many complicating factors.

3. Quartz Crystals

Experiments on the thermal annealing of fission fragment tracks in natural Brazilian quartz crystals are in progress. Crystals cut and polished on the 100 plane appear to give the best track registration characteristics. Full energy fragments incident isotropically and normally on this surface are being used to produce the tracks. It appear that the maximum etched length in the 100 plane for normally incident fragments may prove to give a good thermal annealing correction to the registration efficiency for the fragments incident isotropically, but this needs to be confirmed through further studies.

CONCLUSIONS

The use of track etch kinetics appears to give a reliable approach to apply thermal annealing corrections to fission track densities for isotropic incidence in the case of isotropic-amorphous materials like quartz glass. In the case of crystalline anisotropic materials such as muscovite, an empirical relationship has been established to determine time-temperature regions for which no thermal annealing corrections are needed.

Annealing observations in these totally different SSTR media support a geometric model of registration efficiency, namely that SSTR registration efficiency is a geometric function of track characteristics and is independent of the manner in which these given characteristics have been produced. Studies similar to those pursued here, but in which fission fragment sources thick relative to the range of the fragments are used, may prove of value to those interested in thermal annealing corrections applied to fission track dating.

In terms of SSTR observations in nuclear reactors, the next issue to be addressed concerns overall applicability in high power environments. Besides annealing, three additional effects also produce limitations in SSTR applicability for high power reactors, namely (1) radiation damage, (2) background track production, and

(3) track pile-up at high track density. All of these different effects can act in consort, rather than independently, to define the actual high power-high fluence limitation of a given SSTR material. Studies of such interactive effects are currently in progress.

ACKNOWLEDGEMENTS

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TABLE I

Thermal annealing data of fission fragment tracks in Supra II (Amersil) quartz glass, and calculated values of the registration efficiency f, track diameter <D>, and $<V_t>/V_g$ from track etch kinetics model. The value of η used in the calculation was 0.739 ± .049.

Annealing Temp. °C	Annealing Time (hrs)	N	f	f Calc.	<D>(μm)	<D>Calc(μm)	$<V_t>/V_g$
350	0	625	0.739	0.739	9.2 ± .4	9.0 ± .8	3.8 ± .3
"	0.5	540	0.60 ± .03	0.64 ± .05	8.2 ± .4	7.7 ± .6	2.5 ± .1
"	1.0	478	0.53 ± .03	0.57 ± .05	7.6 ± .4	7.0 ± .6	2.1 ± .1
"	2.0	466	0.52 ± .03	0.54 ± .05	7.25 ± .5	6.9 ± .6	2.1 ± .1
"	4.0	367	0.41 ± .02	0.39 ± .07	5.9 ± .7	6.0 ± .5	1.7 ± .1
"	8.0	291	0.32 ± .02	0.38 ± .05	5.8 ± .5	5.1 ± .5	1.5 ± .1
402	0	723	0.739	0.739	8.7 ± .4	9.0 ± .8	3.8 ± .3
"	1.0	391	0.44 ± .02	0.37 ± .05	5.4 ± .5	6.2 ± .5	1.8 ± .1
"	2.22	252	0.28 ± .02	0.31 ± .05	4.9 ± .5	4.7 ± .5	1.4 ± .1
"	4.0	252	0.28 ± .02	0.29 ± .03	4.7 ± .31	4.7 ± .5	1.4 ± .1
"	8.0	196	0.22 ± .02	0.21 ± .05	3.9 ± .5	4.1 ± .5	1.3 ± .1
380	0	684	0.739	0.739	8.5 ± .5	9.0 ± .8	3.8 ± .3
"	1.15	550	0.61 ± .03	0.69 ± .10	8.0 ± .9	7.8 ± .6	2.6 ± .1
"	2.0	505	0.56 ± .03	0.65 ± .12	7.7 ± .6	7.3 ± .6	2.3 ± .1
"	4.5	506	0.56 ± .03	0.55 ± .07	6.8 ± .6	7.3 ± .6	2.3 ± .1
"	8.3	423	0.47 ± .02	0.50 ± .06	6.4 ± .5	6.5 ± .5	1.9 ± .1

TABLE II
THERMAL ANNEALING DATA FOR MUSCOVITE MICA

Temperature (°C)	Heating Time (hrs)	$\langle \ell \rangle$ μm	$\langle \ell \rangle / \langle \ell_o \rangle$	N	N/No □
402*	0	5.565 ± .06	0.99 ± .02	2313	1.00 ± .03
"	0.5	5.06 ± .10	0.90 ± .02		
"	1.0	4.83 ± .10	0.86 ± .02	2328	1.00 ± .03
"	2.0	4.94 ± .10	0.88 ± .02	2256	0.97 ± .03
"	4.4			2329	1.00 ± .03
"	8.1	4.59 ± .10	0.82 ± .02	2373	1.02 ± .03
"	25.5	4.40 ± .09	0.78 ± .02	2337	1.01 ± .03
"	95.0	3.91 ± .09	0.70 ± .02	2318	1.00 ± .03
459	0	10.05 ± .22	1.01 ± .03	2234	1.05 ± .04
"	0.5	7.26 ± .16	0.73 ± .02		
"	1.0	7.08 ± .16	0.71 ± .02	2128	0.99 ± .04
"	2.0	7.13 ± .16	0.72 ± .02	2067	0.97 ± .04
"	4.0	7.12 ± .16	0.72 ± .02		
"	8.0	6.87 ± .15	0.69 ± .02		
"	16.2	6.10 ± .13	0.61 ± .02		
"	32.0	5.32 ± .12	0.53 ± .02		
"	64.0	4.14 ± .09	0.42 ± .01		
"	146.0	4.06 ± .09	0.41 ± .01	2123	0.99 ± .04
"	263.25	2.50 ± .06	0.25 ± .01		
"	627.8	2.28 ± .05	0.23 ± .01	2069	0.97 ± .04
501	0	10.25 ± .23	1.02 ± .03	2035	0.98 ± .03
"	0.5	6.99 ± .15	0.70 ± .02	2100	1.01 ± .03
"	1.0	5.34 ± .12	0.53 ± .02	2095	1.01 ± .03
"	2.0	3.29 ± .07	0.33 ± .01	2116	1.02 ± .03
"	4.0	2.77 ± .06	0.28 ± .01	2041	0.98 ± .03
"	8.0	2.21 ± .05	0.22 ± .01	2092	1.01 ± .03
"	16.2	2.47 ± .05	0.25 ± .01	1992	0.96 ± .03
"	32.2	1.76 ± .04	0.18 ± .01		
"	89.4	1.93 ± .04	0.19 ± .01		
"	146.5	1.77 ± .04	0.18 ± .01	2107	1.01 ± .03
"	337.7	1.74 ± .04	0.17 ± .01		
"	508.5	1.14 ± .03	0.11 ± .01	1968	0.95 ± .03
518	0			4064	1.00 ± .02
"	23.6			4015	0.99 ± .02
"	167.7			3842	0.95 ± .02
"	311.2			3718	0.92 ± .02
"	456.5			3779	0.93 ± .02
"	617.5			3680	0.91 ± .02
540	0	10.02 ± .22	1.00 ± .02	2230	1.00 ± .02
"	311.6	1.08 ± .03	0.11 ± .01	1962	0.88 ± .03
600	0	10.04 ± .22	1.00 ± .03		
"	0.6	2.85 ± .06	0.28 ± .01		
"	1.2	2.63 ± .05	0.26 ± .01		
"	2.4	2.26 ± .04	0.23 ± .01		
"	5.0	2.11 ± .04	0.21 ± .01		
"	7.8	1.77 ± .03	0.145 ± .004		

* For the data at 402°, $\langle \ell \rangle$ is the average projected length for 40 tracks incident at 45° with the mica surface. Etching is for 15 minutes in 49% HF at (23 ± .3)°C For all other annealing temperatures $\langle \ell \rangle$ is the average length of "flat" tracks etched for 30 minutes at the same temperature. Comparisons of the two methods at 501°C for measuring $\langle \ell \rangle / \langle \ell_o \rangle$ for values above 0.7 show that they are the same within limits of error.

□ The value of No used for each set of data was the average value of N for the subset indicating no track loss.

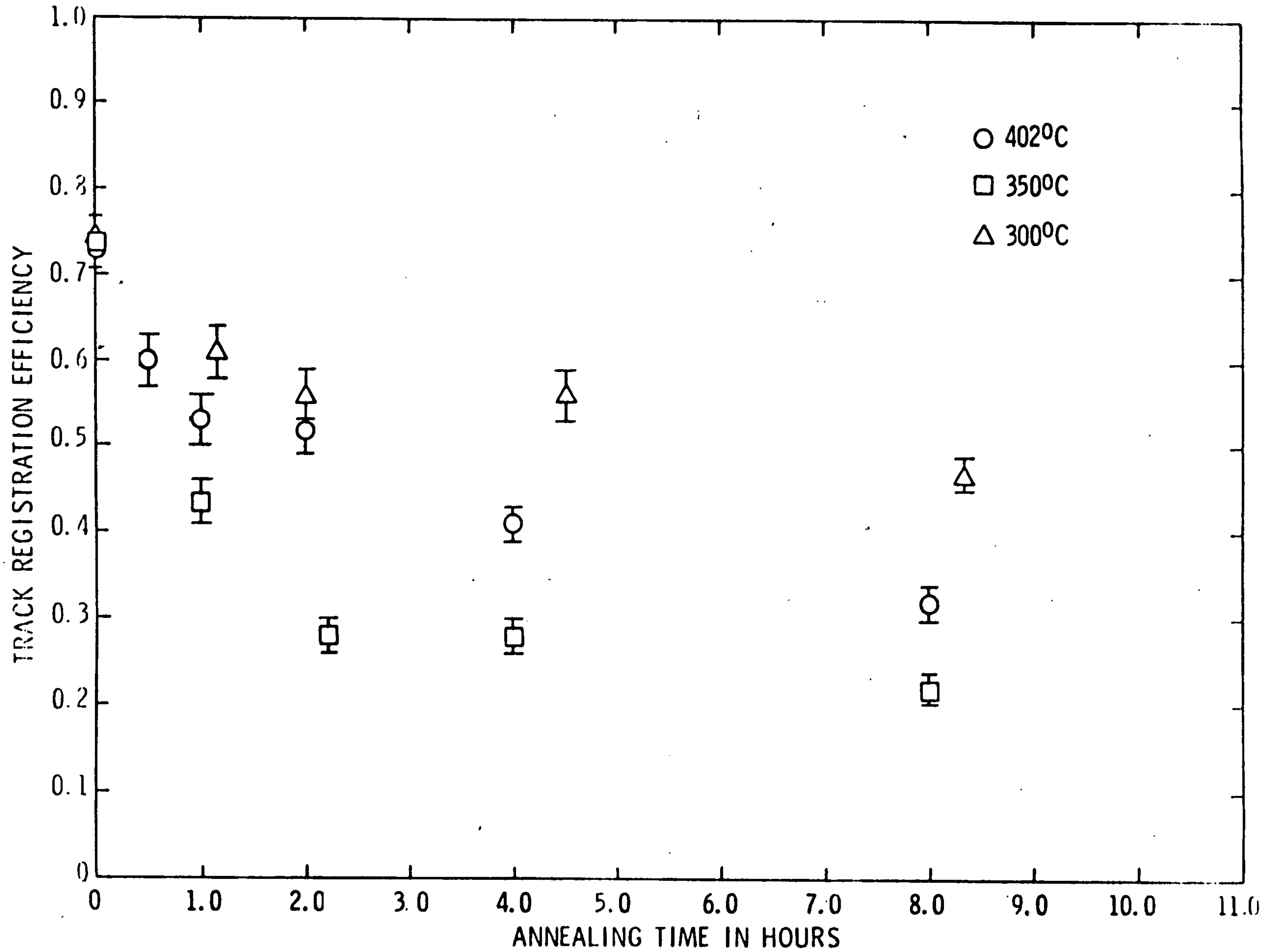

FIGURE 1. Thermal annealing characteristic of fission fragment tracks in quartz glass giving track registration efficiency as a function of time for 300, 350, and 402°C.

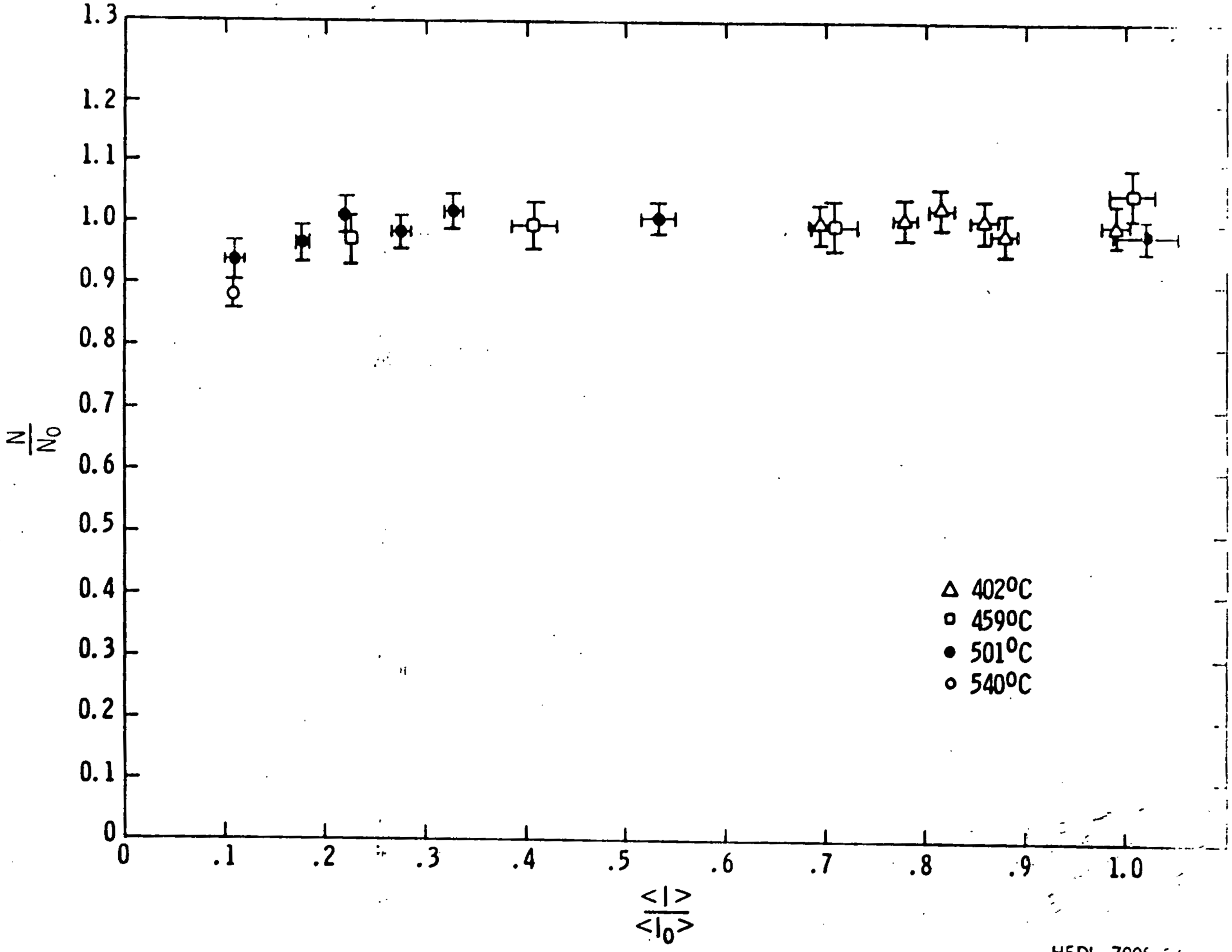

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FIGURE 2. Ratio of track densities in mica for given exposure to full energy fission fragments incident isotropically as a function of $\langle \ell \rangle / \langle \ell_o \rangle$ for "flat" tracks. The value of N_0 is the average of track densities for data at a given temperature for the region over which the curve is horizontal. No corrections are needed for $\langle \ell \rangle / \langle \ell_o \rangle > 0.3$.

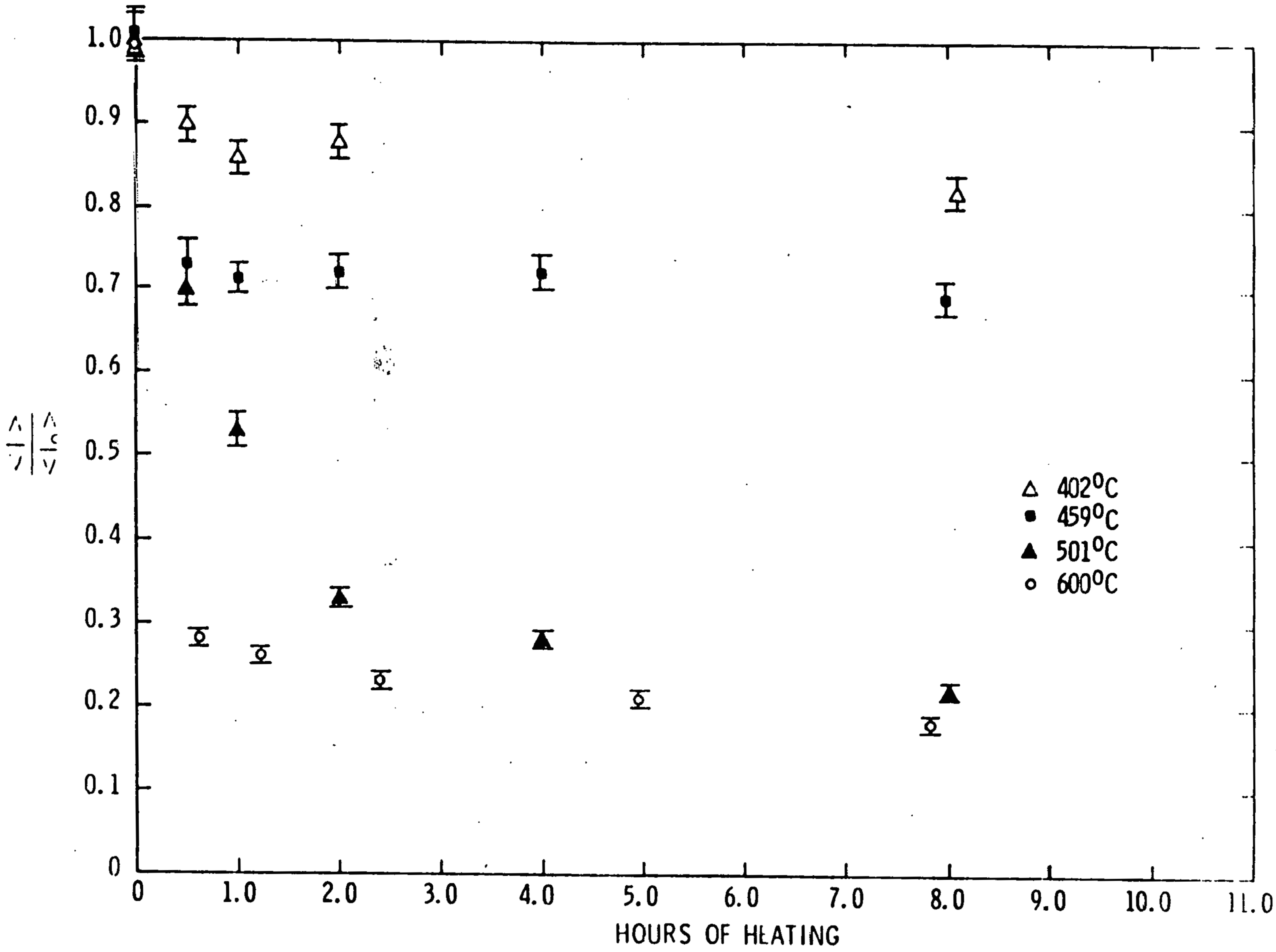

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FIGURE 3. $<\ell>/<\ell_0>$ for "flat" tracks in mica exposed to full energy fission fragments incident isotropically, as a function of heating time for temperatures of 402, 459, 501, and 600°C. The data shows the track annealing characteristics for the first ~ 8 hours of heating.

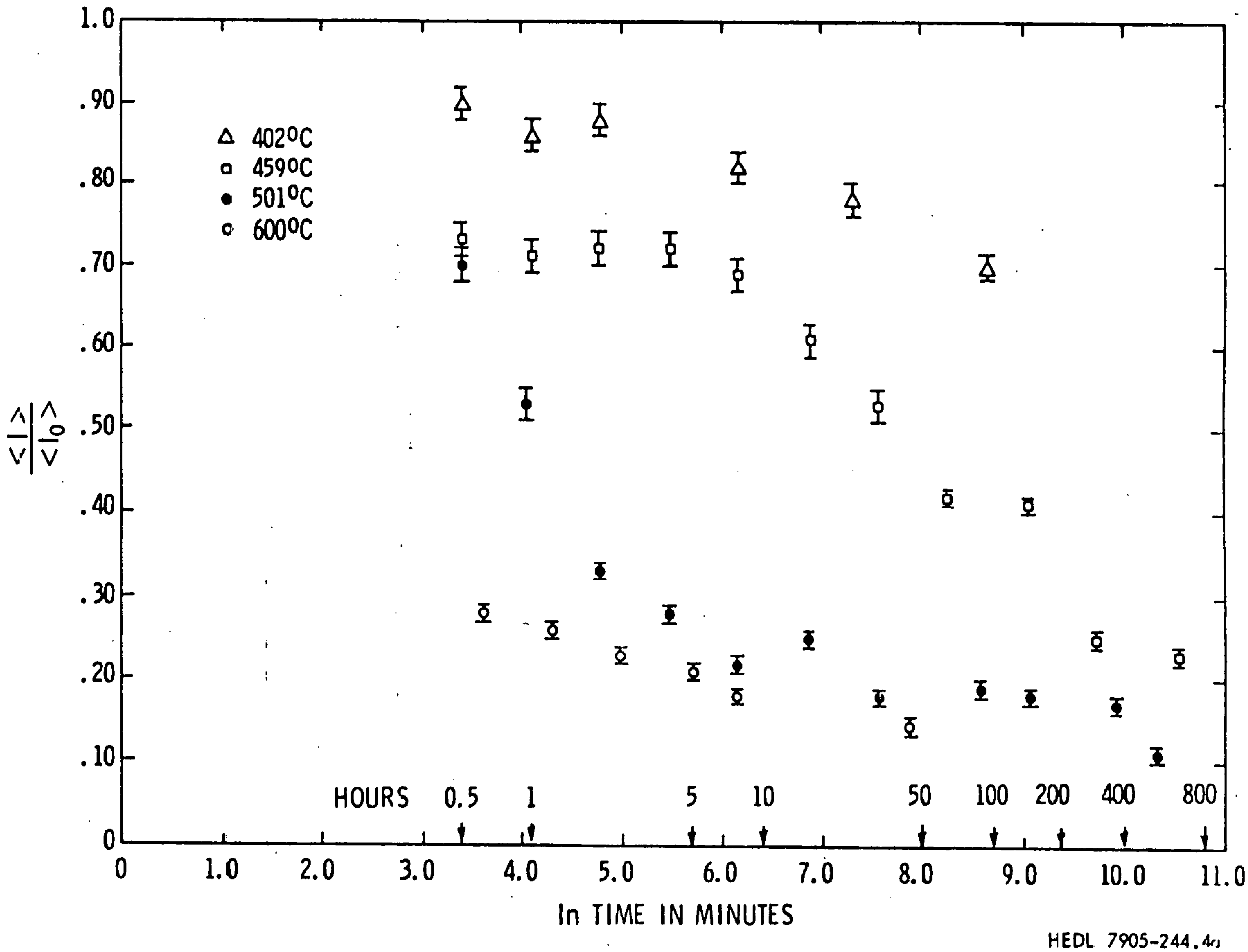

FIGURE 4. The same and additional data for $\langle \ell \rangle / \langle \ell_0 \rangle$ as in Figure 3, except that the abscissa is the ℓn of the heating time expressed in minutes. The heating time in hours is shown at various points on the abscissa.