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SELECTED ETCHING AND ANNEALING PROPERTIES OF BRAZILIAN QUARTZ CRYSTALS FOR SOLID STATE TRACK RECORDER APPLICATIONS

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SELECTED ETCHING AND ANNEALING PROPERTIES OF BRAZILIAN QUARTZ CRYSTALS FOR SOLID STATE TRACK RECORDER APPLICATIONS

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ABSTRACT

The etching and annealing properties of Brazilian quartz crystals are under investigation to determine their suitability for use as solid state track recorders (SSTR) and damage monitors in nuclear reactor environments, where temperatures and neutron fluences are high. Observer objectivity in counting fission tracks has been established at the 1-2% level, and a method of standardizing chemical etching from one sample of quartz to another has been found.

A method has also been found to make corrections for track loss due to thermal annealing in terms of the effect of the annealing on the track size in the direction of maximum bulk etch rate parallel to the 100 plane, provided the fractional track loss does not exceed ∿40%.

KEYWORDS

Solid state track recorder (SSTR); track counting; thermal annealing; bulk etch rates.

INTRODUCTION

Brazilian quartz crystals are known for their high purity and resistance to thermal annealing of fission tracks (Fleischer, Price, Walker, 1975). To investigate the solid state track recorder (SSTR) properties of Brazilian quartz, crystals cut as disks and polished in the 100 and 001 planes have been investigated.

These crystals were pre-etched for 100 minutes in 49% HF at room temperature, and then in 65% boiling NaOH for 25 minutes. Selected pre-etched crystals were then exposed to fission fragments from ^{244}Cm or ^{252}Cf. One face of the disk was placed in direct contact with one of these thin sources, and the other face was exposed in a vacuum to normally incident fission fragments.

After exposure some crystals were etched in 65% boiling NaOH for various times, and the fission tracks incident isotropically were manually counted. Other samples were thermally annealed for various times and temperatures before etching so that changes in track density and size could be investigated.

CHEMICAL ETCHING AND FISSION TRACK COUNTING

The reproducibility and observer objectivity of manual fission track counting for exposures in the 100 and 001 planes after various etching times in boiling 65% NaOH has been investigated. After the pre-etching in HF and NaOH and exposure to ^{244}Cm as described above were completed, two samples were etched for 10.0 minutes in the 65% boiling NaOH. Fission tracks in each SSTR sample, one cut in the 100 plane, and the other in the 001 plane, were counted by five observers, using Nikon LKE microscopes. Each of the two samples was etched for an additional 10.0 minutes, and the counting repeated. This procedure was repeated again for total etching times of 25.0 and 30.0 minutes.

In addition to the track counting after each etch, the lengths <L> of the etched holes in the 001 and in the 100 planes in the direction of maximum bulk etch rates using normally incident fragments were measured with a digitized filar micrometer eyepiece.

The results for the track counting and for the <L> measurements are presented in Table 1. Results for track counting is generally better for the 100 plane than for the 001 plane. Incidentally, observers were not instructed regarding what to count. Since in both planes, surface imperfections exist, the results indicate that even without significant experience with quartz crystals, agreement is at the 1-2% level for the 100 plane. If the data showing best agreement for four observers is chosen, the sigmas for the 100 plane etched 20 and 25 minutes are 0.73 and 0.79%, respectively. Results comparable to that obtained for muscovite (Gold, Armani, Roberts, 1968) are possible.

The data for <L> as a function of etching time t for the 100 and 001 planes are plotted in Fig. 1 and fitted to linear functions. The significance of the linearity

Table 1

RESULTS OF FISSION TRACK COUNTING AND TRACK SIZE MEASUREMENTS IN NATURAL QUARTZ CRYSTALS CUT IN THE (a) 100 PLANE AND IN THE (b) 001 PLANE

Part (a) 100 Plane

Etching Time (min.)	<L>† (μm)	(V_g) max μm/min	Tracks Counted By Each Observer* 1	2	3	4	5	Average
10.0	3.94±0.11	0.394±0.011	2151	2361	2398	2391	2283	2316±103(4.5%)
20.0	8.05±0.27	0.403±0.014	2469	2426	2496	2446	2454	2458±26(1.1%)
25.0	10.12±0.21	0.405±0.008	2435	2390	2406	2419	2496	2429±41(1.7%)
30.0	11.39±0.33	0.380±0.011	2438	--	2454	2388	2445	2431±30(1.22%)

Part (b) 001 Plane

Etching Time (min.)	<L>† (μm)	(V_g) max μm/min	Tracks Counted by Each Observer* 1	2	3	4	5	Average
10.0	--	--	2067	2111	2174	2258	1788	2080±178(8.6%)
20.0	1.80±0.13	.090±.007	2231	2139	2237	2210	2146	2192±47(2.1%)
25.0	2.30±0.16	.092±.006	2258	2177	2165	2213	2244	2211±41(1.8%)
30.0	2.66±0.18	.089±.006	2154	--	2205	2040	--	2133±84(4.0%)

† Measured for tracks incident perpendicular to the plane along which the crystal was cut

*Observers are numbered 1 through 5.

of $\langle L \rangle$ vs t is that a predetermined value of $\langle L \rangle$ can always be achieved. First one uses a relatively short etch time t_s to determine $\langle L \rangle_s$, then by re-etching one can bring $\langle L \rangle_s$ up to the predetermined value, thus standardizing the etch from sample to sample. This linearity also demonstrates the reproducibility of the etching technique being used and that "step etching" is a reliable procedure for quartz crystals. Further studies are in progress.

THERMAL ANNEALING CHARACTERISTICS

In the temperature range projected for using quartz crystals as SSTR in high power reactor environments, it is likely that no effects due to thermal annealing alone will be a problem. Combinations of thermal annealing and radiation damage due to high fast neutron fluences, however, will certainly place limits on the usefulness of quartz crystals as SSTR in high power reactor environments.

Only crystals cut in the 100 plane were used in the annealing studies. Samples were exposed to give about 2600 tracks from fission fragments incident isotropically. As before, the other side of the disk was exposed to normally incident full-energy fission fragments. SSTR samples to be heated after exposure were placed between nickel foils in metal capsules. Three samples were heated at a temperature of 857°C for 1.0, 2.0, and 4.0 hours, respectively. The samples were then etched for 20 minutes in 65% boiling NaOH. No tracks were found except in the sample heated for 1.0 hours. Antoher series was heated for 1, 2, and 4 hours at 837°C. After etching, tracks were found only in the samples heated for 1 and 2 hours. A third run was made at 812°C, with heating times of 1, 2, 4, and 16 hours. Tracks were found in the 1, 2, and 4 hour runs.

Data and results for these annealing experiments are given in Table 2. Tracks were counted by two observers, but the $\langle L \rangle$ were measured by one. The value of $\langle L \rangle$ was measured for 20 normally incident tracks in each sample. A plot of $\langle N \rangle / \langle N_0 \rangle$ vs $\langle L \rangle / \langle L_0 \rangle$ is shown in Fig. 2 for the quartz samples in which tracks were revealed. Here $\langle N \rangle$ and $\langle N_0 \rangle$ are the tracks counted in the annealed and unannealed samples, respectively, for the same exposure, and $\langle L \rangle$ and $\langle L_0 \rangle$ are the track lengths in the direction of the maximum bulk etch rates in the 100 plane for the annealed and unannealed samples, respectively. The behavior of $\langle N \rangle / \langle N_0 \rangle$ vs $\langle L \rangle / \langle L_0 \rangle$ is linear in the region $\langle L \rangle / \langle L_0 \rangle \geq 0.6$. In this limited domain $\langle L \rangle / \langle L_0 \rangle$ is found to be a good parameter to predict track loss.

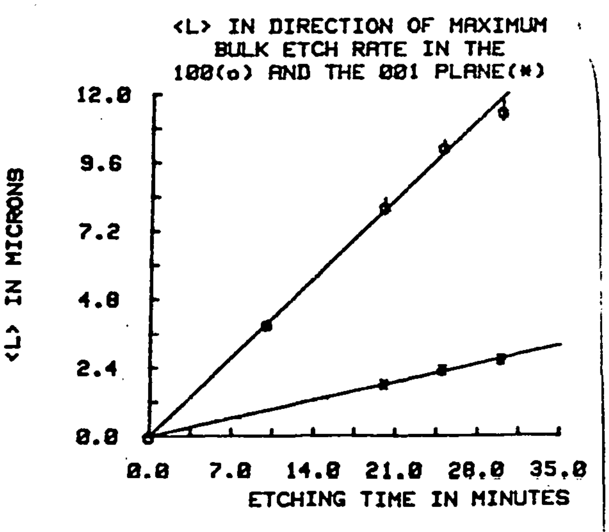

Figure 1

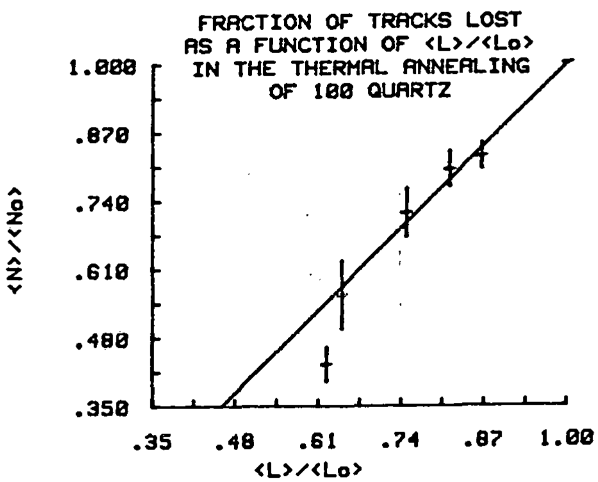

Figure 2

TABLE 2 Thermal Annealing Data for Crystals Cut in the 100 Plane

Sample Number	Heating Time (min)	Heating Temperature (°C)	Tracks Counted Obs. 1	Obs. 2	Average	⟨L⟩ (μm)
A0	0	857	2611	2635	2623	7.84±0.21
A1	60	857	2073	2111	2093	6.40±0.53
A4	0	837	2529	2535	2532	9.87±0.26
A5	63	837	2139	2013	2085	8.55±0.32
A6	123	837	1033	1134	1084	6.15±0.43
AN3	0	812	2606	2504	2555	8.52±0.27
AN1	120	812	1861	1793	1827	6.37±0.58
AN8	240	812	1459	1399	1429	5.51±0.86

Further work is in progress to give a more complete annealing characterization of fission tracks in natural quartz, including the combined effects of annealing and radiation damage due to high fluences of fast neutrons. It would appear that these techniques will be of value in correcting for track loss due to annealing in fission track dating research.

ACKNOWLEDGEMENT

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