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Ion track annealing in quartz investigated by small angle X-ray scattering

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ABSTRACT

We report on the reduction of cross-section and length of amorphous ion tracks embedded within crystalline quartz during thermal annealing. The ion tracks were created via Au ion irradiation with an energy of 2.2 GeV. The use of synchrotron-based small angle X-ray scattering (SAXS) allowed characterization of the latent tracks, without the need for chemical etching. Temperatures between 900 and 1000 °C were required to see a notable change in track size. The shrinkage in cross-section and length was found to be comparable for tracks aligned perpendicular and parallel to the *c*-axis.

1. Introduction

Swift heavy ions can create amorphous defect structures, called ion tracks, along their trajectory in a target material [1,2]. They result from the interaction of high-energy heavy ions with the electrons of a target material. Although these tracks are generally only a few nanometers in diameter, they can reach lengths of up to tens to hundreds of micrometers, leading to extremely large aspect ratios [3]. At elevated temperatures, they are known to shrink in size [4]. Ion tracks have a wide range of applications in materials science and nanotechnology, nuclear physics, geo- and thermochronology, archaeology, and interplanetary science. In nature, these tracks occur as a result of the fission of traces of ^{238}U in minerals such as apatite and zircon and are employed for dating and constraining the thermal history of geological material [4]. Quartz is used as track detector in extreme environments due to the temperature resistance of the ion tracks [5] or for optical waveguides [6].

Ion track characterization historically utilizes chemical etching to enlarge the tracks such that they can be imaged by optical microscopy; however, this process completely erases their initial structure [3] such that important information about the track structure is lost. Furthermore, interpretation of the results is difficult as the track dimensions are not only determined by the latent track structure but also by the etching kinetics. Recently, we have shown that small angle X-ray scattering (SAXS) is well suited to study the cross-section of un-etched tracks in quartz [7] and other materials by utilizing the difference in density between tracks and the undamaged host matrix [8–10]. Furthermore, we have demonstrated its ability to study the track annealing kinetics in minerals with *in situ* annealing [7].

Here, we investigate un-etched ion tracks in synthetic quartz and the reduction in cross-section and length during annealing. Previously, using chemical etching of annealed tracks in natural quartz, Sandhu et al. reported track lengths to shorten significantly faster for tracks oriented perpendicular to the *c*-axis, as compared to those oriented parallel to the *c*-axis [11]. Similar observations were reported by Sawamura et al. for synthetic quartz at lower temperatures studying the track shrinkage [12]. However, in direct contrast Aframian found no significant anisotropy for track annealing in natural quartz [13], also using etching experiments. The annealing experiments in the present work on un-etched tracks indicate no significant difference for the shrinkage of in cross-section and length for tracks oriented parallel and perpendicular to the quartz *c*-axis.

2. Experiment

For our experiments we used commercial synthetic single-crystalline quartz [c-SiO_2] wafers of *Y* [$10\bar{1}0$] and Z-cuts

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[0001], all of dimension 10 mm × 10 mm × 0.5 mm. The wafers were irradiated at the GSI Helmholtz Centre (Darmstadt, Germany) with 2.2 GeV ^{197}Au ions to a fluence of 5×10^{10} ions/cm^2. The incident ion direction was normal to the polished surface of the respective wafer, leading to tracks perpendicular (⊥c) as well as parallel (‖c) to the *c*-axis of quartz. The electronic energy loss (d*E*/d*x*) of the Au ions at the surface was calculated using the SRIM-2008 [14] package to 24 keV/nm. The resulting ion tracks have a length of approximately 90 μm, estimated using the same code assuming a track formation threshold of 2 keV/nm [15]. The ⊥c and ‖c-samples were subsequently mechanically polished to thicknesses between 65 and 90 μm.

For characterization of the track radii, transmission small angle X-ray scattering measurements were performed at the SAXS/WAXS beamline at the Australian Synchrotron in Melbourne, Australia. The energy of the X-rays was 11 keV and the camera length ~1.6 m. The spot size of the X-ray beam was approximately 0.04 mm^2. For calibration of the *q*-axis, silver behenate was used. The absolute scattering intensity was normalized using glassy carbon [16]. *In situ* annealing experiments were performed using a LINKAM TS1500 furnace under ambient atmosphere. The scattered intensity was collected using a Pilatus 1 M detector taking images once every minute with an exposure time of 2 s. each. The sample surface normal or equivalently the axis of the ion tracks was orientated slightly tilted (5°) with respect to the X-ray beam. An unirradiated sample provided data for background removal. As demonstrated earlier, the track structure is unaffected by X-rays even after prolonged exposure [7]. A more detailed description of the SAXS experiments can be found elsewhere [2,8].

3. Results and discussion

Fig. 1 shows scattering images of SAXS measurements for quartz ⊥c (Y-cut) and ‖c (Z-cut) at different stages during the annealing process. The intensity represents the scattering in *q*-space, with dark colors corresponding to large intensities and *vice versa*. At room-temperature (first image) the scattering of X-rays with ion tracks in the tilt geometry leads to the oscillating streaks, due to the high aspect ratio of track radius to length (1:10,000). The strong oscillations indicate a sharp density transition between track interior and the matrix. SAXS utilizes this clear density contrast and the large number of tracks (about 10^7 for given beamsize and ion fluences in this work) to probe the structure of the 'individual' tracks with excellent precision. This is possible as the mono-energetic ion irradiation normal to the surface produces virtually identical, parallel tracks. The X-ray penetration depth within SiO_2 clearly exceeds the specimen thicknesses, allowing SAXS also to probe the tracks along the entire length. For moderate temperatures (<800 °C) no apparent change of the signal is seen. However, at higher temperatures, a clear reduction of the intensity is noticeable, making the entire streak appear shorter, yet with increasing distance between the minima of the oscillation. Finally, at 1040 °C the streak-signal is hardly visible anymore and the image is dominated by the isotropic background.

The intensities of the oscillating streaks were extracted from each scattering pattern, as they contain information about the radial electron density of the tracks. Fig. 2(a and b) shows these intensities (open circles) as a function of the magnitude of the scattering vector *q*. For comparison, alignment of tracks ⊥c (Fig. 2a) and ‖c (Fig. 2b) are shown. The solid lines show the respective fits. Best results in fitting the spectra were obtained using a cylinder model with constant density. The form factor can be expressed as $f(q) = \pi R L \Delta\rho 2J_1(Rq)/q$, where L is the track length, R the track radius, $\Delta\rho$ the density difference between track and the material of the host crystal, and $J_1(x)$ the first order Bessel function. The track radius is varied by a polydispersity parameter σ_R [8,17], to account for a variation of the track radius with depths and between individual tracks [18]. This parameter was typically found to be within the range of 0.3 nm or around 10%. The scattering intensity $I(q)$ is expressed as

$$I(q) = C|f(q)|^2 = C\left|\pi R^2 L \Delta\rho\right|^2 |2J_1(R \cdot q)/R \cdot q|^2 \quad (1)$$

with proportionally constant C. For $q \to 0$ the latter term becomes $|2J_1(R \cdot q)/R \cdot q| \to 1$ and the first term is defined as fitting parameter I_0:

$$I_0 = I(q \to 0) = C|\pi R^2 L \Delta\rho|^2 \quad (2)$$

The optimal fitting parameters and their uncertainties were determined iteratively by a least square algorithm. It produces a radius of the best fitting cylinder that directly relates to the average track radius. The radius is only dependent on the *q*-positions of the oscillations and independent of all other fitting parameters. Note that for higher annealing temperatures the minima shift towards larger values of *q*, which are associated with smaller radii.

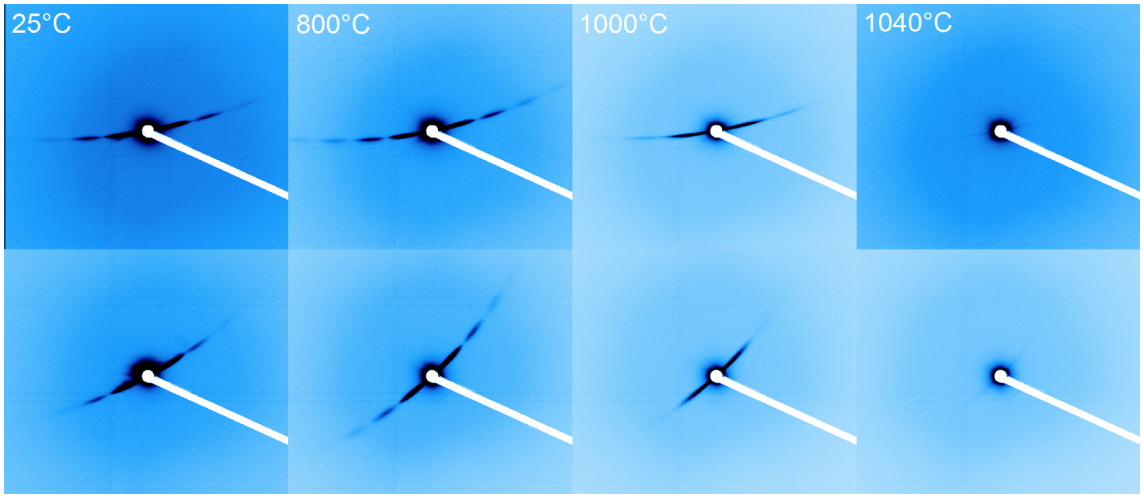

Fig. 1. [Top row: tracks *c*-axis shown, bottom row: tracks ‖ to *c*-axis] SAXS scattering patterns at RT and after annealing at 800, 1000 and 1040 °C. The oscillating "streaks" are a result of the high aspect ratio of the ion tracks.