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SAXS study of ion tracks in San Carlos olivine and Durango apatite

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ABSTRACT

Ion tracks were generated in crystalline San Carlos olivine $(Mg,Fe)_2SiO_4$ and Durango apatite $Ca_{10}(PO_4)_6F_2$ using different heavy ions (^{58}Ni, ^{101}Ru, ^{129}Xe, ^{197}Au, and ^{238}U) with energies ranging between 185 MeV and 2.6 GeV. The tracks and their annealing behavior were studied by means of synchrotron based small angle X-ray scattering in combination with *in situ* annealing. Track radii vary as a function of electronic energy loss but are very similar in both minerals. Furthermore, the annealing behavior of the track radii has been investigated and preliminary results reveal a lower recovery rate of the damaged area in olivine compared with apatite.

1. Introduction

Ion tracks are long narrow defect structures that are generated by the high electronic excitations that swift heavy ions cause as they traverse through a solid. Ion tracks in minerals and their behavior upon thermal annealing provide the critical basis for geochronology/thermochronology, archeology, and astrophysics. In natural apatite and olivine, ion tracks have been extensively investigated for their applications in fission track dating and for identification of cosmic rays in meteorites, respectively [1,2]. At elevated temperatures, ion tracks shrink in size and fragment into sections until they are completely annealed. Thus, the length and morphology of fission tracks can be used to determine the thermal history of Earth's crust and meteorites [3–5].

Different techniques have been employed to study ion tracks and their properties in minerals, including chemical etching [6], transmission electron microscopy (TEM) [7–9], and Rutherford backscattering spectroscopy [10]. In addition, different groups have studied the annealing properties of tracks in apatite [11–14] and olivine [4,15]. These experiments predominantly utilize chemical etching and investigate empirically the effect of annealing on the number density and length distribution of the etched tracks [11–13]. Etching, however, erases the primary damage structure of the track, thus, essential information on the actual scale of the underlying radiation damage is irrevocably lost. Small angle X-ray scattering (SAXS) can be used as an alternative technique to study ion track damage as it measures density changes at the nanometer scale [16]. Recent SAXS measurements provided details of the track structure in amorphous SiO_2 [17,18] and natural apatite [19]. Using *in situ* and *ex situ* annealing experiments, it was demonstrated that SAXS is well suited for monitoring the annealing kinetics of ion tracks.

In this work we have applied SAXS to measure track radii in olivine and Durango apatite, the latter of which complement earlier results [19]. Moreover, we show preliminary results on isothermal annealing of ion tracks in olivine and compare them to previous results on apatite [19].

2. Experiment

Crystalline samples of Durango apatite and San Carlos olivine (95% Mg_2SiO_4) with thicknesses of 30–50 μm were irradiated with swift heavy ions. The irradiation experiments were performed using ^{58}Ni, ^{101}Ru, ^{129}Xe, ^{197}Au, and ^{238}U ions with energies between 0.6 and 2.6 GeV at the UNILAC accelerator at GSI in Germany. At the ANU Heavy Ion Accelerator Facility, apatite samples were irradiated with 185 MeV ^{197}Au ions. Irradiation was performed at room temperature under normal incidence. Details of the irradiation parameters are given in Table 1, where the values for the surface electronic energy loss dE/dx, and the ion range were calculated using SRIM 2008 [20]. Irradiation fluences ranged from 5×10^{10} (well separated ion tracks) to 2×10^{12} ions/cm^2 (overlapping tracks).

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Table 1
Irradiation parameters and track radii extracted from SAXS measurements.

Sample	Sample thickness (μm)	Fluence (ions/cm^2)	Ion	Ion energy (MeV)	Electronic energy loss (keV/nm)	Projected range (μm)	Radius (nm)	Radius polydispersity (nm)	Reference
Olivine	40	2×10^{11}	^{58}Ni	644	6	88	1.8 ± 0.2	0.5 ± 0.2	Present work
Olivine	40	5×10^{10}	^{101}Ru	1121	13	81	2.7 ± 0.1	0.4 ± 0.1	Present work
Olivine	40	5×10^{10}	^{197}Au	2187	26.2	90	4.6 ± 0.1	0.3 ± 0.1	Present work
Olivine	40	5×10^{10}	^{238}U	2642	34.4	87	5.3 ± 0.1	0.4 ± 0.1	Present work
Apatite	50	5×10^{10}	^{238}U	2047	37.5	66	5.5 ± 0.1	0.3 ± 0.1	Present work
Apatite	40–50	5×10^{10}–1×10^{12}	^{197}Au	185	23.4	15	5.5 ± 0.1	0.5 ± 0.2	Present work
Apatite	30	2×10^{12}	^{58}Ni	644	6.3	83	1.8 ± 0.1	0.3 ± 0.1	[19]
Apatite	30	5×10^{10}	^{101}Ru	1121	13.5	76	2.9 ± 0.1	0.2 ± 0.1	[19]
Apatite	40	5×10^{10}	^{129}Xe	1432	18.6	75	3.6 ± 0.1	0.3 ± 0.1	[19]
Apatite	30	5×10^{10}	^{197}Au	2187	27.3	85	4.8 ± 0.1	0.2 ± 0.1	[19]

Transmission SAXS measurements were performed at the SAXS/WAXS beamline at the Australian Synchrotron with an X-ray energy of 12 keV and a camera length of approximately 1600 mm. Mounting the samples on a three-axis goniometer allowed for precise alignment of the ion tracks with respect to the X-ray beam. Measurements were taken with the ion tracks tilted by 0°, 5° and 10° with respect to the X-ray beam and spectra were collected with a Pilatus 1 M detector with exposure times of 5 and 10 s. Scattering from un-irradiated samples was measured for background removal and the absolute scattering was calibrated using a glassy carbon standard [21].

In order to study the ion track recovery, isothermal annealing was carried out *in situ* on the olivine and apatite samples irradiated with 2.2 GeV Au ions. A hot-air heater was positioned underneath the sample, and the temperature was monitored using a thermocouple at sample height. The samples were kept at 350 °C for about 6 h, and SAXS measurements were taken approximately every 40 s. A more detailed description of track annealing in apatite is reported in Ref. [19].

3. Results and discussion

Fig. 1(a) and (c) show isotropic scattering images from the ion tracks in olivine and apatite, respectively, when the ion track axis is nearly parallel to the X-ray beam. Tilting the sample by 10°, for example, results in highly anisotropic scattering in the form of narrow streaks that are shown in Fig. 1(b) and (d). This anisotropy results from the high aspect ratio of the ion tracks, which are only a few nanometers wide and up to tens of micrometers long. X-ray intensities of radial sectors perpendicular to the streaks in the anisotropic images resemble those of unirradiated samples. This suggests the lack of significant density fluctuations on the nanometer length scale along the ion tracks.

Scattering intensities extracted from the streaks for olivine samples irradiated with different ion/energy combinations are plotted in Fig. 2. From the strong oscillations, largely monodisperse radii and sharp transitions between the track densities and the surrounding matrix material can be inferred. The best model that adequately fits the observed scattering for all irradiation conditions is a simple cylinder model with a constant density, different from that of the matrix material. The form factor of this model can be written as:

$$f(q) = 2\pi LR\rho_0 \frac{J_1(Rq)}{q}$$

where q is the scattering vector, L the length of the track, R the track radius, ρ_0 the density difference between track and matrix, and J_1 is the first order Bessel function. A narrow Gaussian distribution of

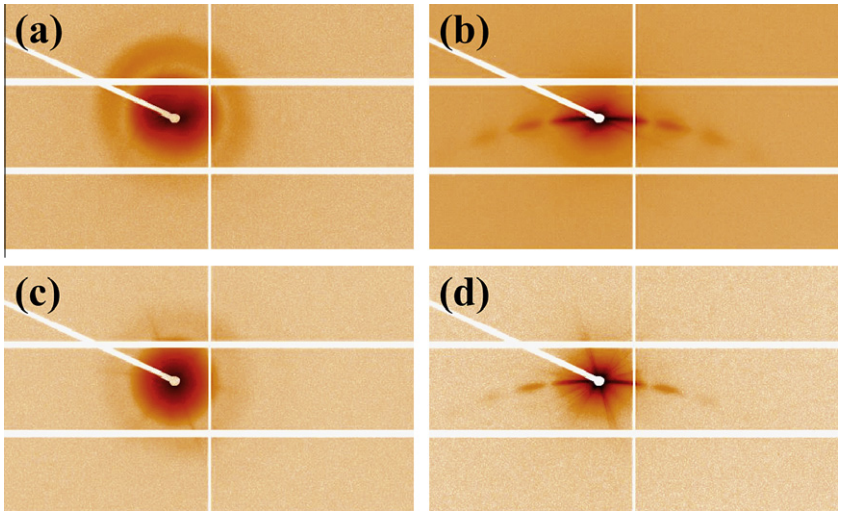

Fig. 1. Scattering image of samples irradiated with 2.2 GeV Au ions (a) olivine with the X-ray beam parallel to the ion tracks, (b) with the tracks tilted by ~10°, (c) apatite with the X-ray beam parallel to the ion tracks, (d) with the tracks tilted by ~10°.