Ion tracks in apatite: Does annealing rate depend on pre-anealing sample preparation?

Igor Alencar,* Sandro Guedes, and Julio Cesar Hadler
Departamento de Raios Cósmicos e Cronologia - Instituto de Física "Gleb Wataghin"
Universidade Estadual de Campinas, Unicamp - Rua Sérgio Buarque de Holanda, 777, CEP 13083-859, Campinas-SP, Brazil

The resistance of radiation damage (latent fission tracks, metamictization) in minerals has been studied for decades. The aim of these studies is the employment of such materials as nuclear waste ceramics and thermo-geochronometers. The last is based on the knowledge of annealing kinetics and the Principle of Equivalent Time, which states that the reduction of damage by a thermal treatment (annealing) is independent from previous thermal history. We present preliminary results for reduction in projected length for fossil and pre-annealed apatite samples irradiated with ^{152}Sm ions (≈150MeV and 45°). It was found that projected length in fossil samples are more resistent to annealing.

Solid State Track Detectors (SSTDs) are materials capable of registering charged particles. In this sense, many minerals are able to register swift heavy ion passage [1, 2]. Particulary, apatite is employed in many studies. It is the most abundant naturally ocurring phosphate, it possesses an hexagonal structure and the following ideal chemical formula:

$$Ca_5(PO_4)_3(F,Cl,OH)$$

where the end-member (fluorine, chlorine or hydroxyl) is aligned with the c-axis. More detailed information about apatite structure and composition could be found in geochemical reviews [3, 4].

The registration of charged particle is a consequence of its energy deposition into the target. This energy deposition leads to the formation of distorted (damaged) regions in the crystal structure. These regions are called latent tracks and they can fade, mainly by thermal treatment (annealing), restoring the original bulk structure. This property is interesting for two research branches: immobilization of high-level nuclear waste [5] and thermo-geochronology. The latter is performed with a revelation process (chemical etching) and ordinary optical microscopy. Chemical etching is based on the lower density of damaged regions, which creates an enhanced etching rate as compared to the bulk one, enlarging the (etched) tracks. In the microscope it is possible to measure surface track areal density, surface track projected length and horizontal confined track full length. The full length is measured when a confined track parallel to the observation plane intercep a surface track, the so-called Track IN Track (TINT) [6].

Mineral dating is possible due to uranium decayment. Uranium is present in trace amounts, substituing calcium at the cation site. The exponential decay law can be written as:

$$P\,(e^{-\lambda t}-1) = D$$

where P represents the amount of remaining parent atoms, D the amount of produced daughter atoms, λ the decay constant and t the elapsed time. For fission track dating, D could be related to the spontaneous ^{238}U fission track areal density and P with thermal neutron induced ^{235}U fission track areal density. At first order in time, the mineral age is [7]:

$$t = \left(\frac{\sigma_{th}}{\lambda_F\ \eta}\right)\phi_{th}\left(\frac{\rho_S}{\rho_I}\right)$$

where σ_{th}, λ_F and η are the thermal neutron ^{235}U fission cross-section, ^{238}U spontaneous fission decay constant and the ^{238}U to ^{235}U isotopic ratio, respectively. These are constants and for dating it is necessary to measure the thermal neutron flux, spontaneous and induced fission track areal densities (ϕ_{th}, ρ_S and ρ_I, respectively). There are some methods to obtain spontaneous and induced fission track areal densities and they were discussed by Gleadow [8].

As tracks fade by annealing, full length distributions can be used to infer thermal histories of the host rock. This is based on: annealing kinetic models (*e.g.* [9, 10]); Principle of Equivalent Time [11]; and inverse modelling [12]. Annealing models are calibrated with laboratory annealing experiments and are extrapolated to geological timescales. The Principle of Equivalent Time states that track reduction by a thermal treatment is independent from the previous thermal history experienced by the sample, which opens the possibility to describe annealing with variable temperature. Finally, inverse modelling is required, because the consequence of the phenomenon is measured (length distribution), while it is attempted to discover the cause (thermal history). It is worth noting that inverse problems possess more than one solution.

Laboratory experiments are usually performed in neutron irradiated samples after a pre-annealing preparation. This pre-annealing is used to erase all spontaneous tracks. This work concerns with the question if this pre-annealing sample preparation affects the annealing kinetics of tracks. For this purpose, many slices from natural apatite crystal where obtained by a coarsely cut in the $\{10\bar{1}1\}$ plane. Slices were separated in two aliquots: one of them was pre-annealed (45N) while the other retained the fossil tracks (45F). These slices were mounted in epoxy resin, grinded and polished to expose a flat, smooth surface. Then, samples were irradiated with ^{152}Sm ions (≈150MeV and 45°) at GSI (Darmstadt, Germany). Samples were enclosed in pairs (one 45F together with one 45N) and were put into a furnace for thermal runs. These runs were performed at constant temperature for fixed times (10 or 1000 hours). Afterwards, each sample pair was etched together in diluted nitric acid. Details of etching experiments will be published elsewhere [13].

*igorav@ifi.unicamp.br

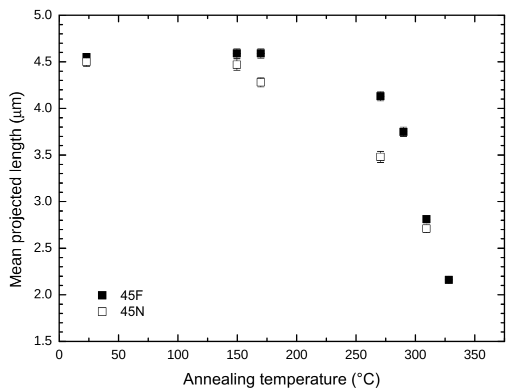

Figura 1: Samarium mean projected length reduction in apatite for an isochronal annealing experiment (10 hours).

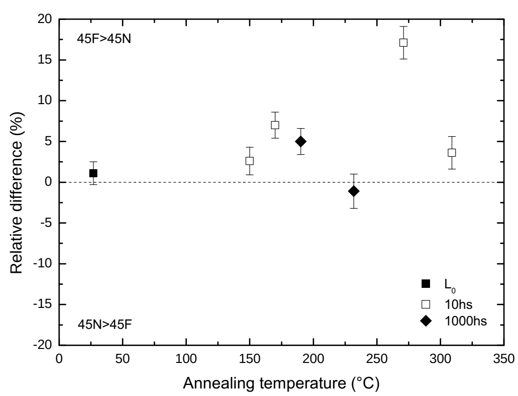

Figura 2: Relative difference in projected length measurements from 45F and 45N samples.

Measurements were performed in a Zeiss© Axioplan 2 microscope at 1000× nominal magnification. Results for 10 hours isochronal experiment are shown in figure 1. In this figure, it is possible to see a faster projected length reduction for pre-annealed samples between 150°C and 310°C. As our experiment allows us to make a direct comparison between 45F and 45N samples annealed at the same temperature, figure 2 presents the relative difference of projected length from both samples. This relative difference was calculated taking the difference between the projected lengths for 45F and 45N samples and dividing it by the combined mean. Thus, positive values indicate projected length from 45F samples are greater than 45N samples, while negative values indicate the opposite. These preliminary results show that 45F samples retain longer projected lengths and, therefore, the annealing rate does depend on pre-annealing sample preparation. This conclusion re-opens the discussion on the validity of the presently used proxies (reduction on neutron induced lengths after pre-annealing preparation) for annealing extrapolation to geological timescale and on the Principle of Equivalent Time hypothesis. Further work with prismatic ($\{10\bar{1}0\}$ planes) and basal ($\{0001\}$ planes) orientations are in progress to confirm this trend.

This work is financially support by Brazilian foundation CNPq. The authors are in debt with Dr Raymond Jonckheere for fruitful discussion about annealing experiments and Dr Christina Trautmann for ion irradiations at GSI.

[1] Fleischer, Price, Walker. Nuclear Tracks in Solids: Principles and Applications. *University of California Press*, 1975.

[2] Wagner & van den Haute. Fission-Track Dating. *Kluwer Academic Publishers*, 1992.

[3] Hughes & Rakovan. *Rev. Mineral. Geochem.* 48:1-12, 2002.

[4] Pan & Fleet. *Rev. Mineral. Geochem.* 48:13-49, 2002.

[5] Ewing. *Proc. Natl. Acad. Sci.* 96:3432-3439, 1999.

[6] Lal, Rajan, Tamhane. *Nature* 221:33-37, 1969.

[7] Price & Walker. *J. Geophys. Res.* 68:4847-4862, 1963.

[8] Gleadow. *Nucl. Tracks* 5:3-14, 1981.

[9] Guedes, Hadler, Oliveira, Moreira, Iunes, Tello. *Radiat. Meas.* 41:392-398, 2006.

[10] Guedes, Moreira, Devanathan, Weber, Hadler. Improved fission-track annealing model based on reevaluation of annealing data, *in preparation*.

[11] Duddy, Green, Laslett. *Chem. Geol. (Isot. Geosci. Sec.)* 73:25-38, 1988.

[12] Lutz & Omar. *Earth Planet. Sci. Lett.* 104:181-195, 1991.

[13] Alencar, Guedes, Jonckheere, Trautmann, Soares, Moreira, Curvo, Tello, Nakasuga, Dias, Hadler. Projected length studies through chemical etching and annealing experiments of ^{152}Sm ion tracks in apatite, *in preparation*.