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Experimental evidence regarding the pressure dependence of fission track annealing in apatite

J.S. Schmidt [a,*], M.L.M.V. Lelarge [a], R.V. Conceicao [a], N.M. Balzaretti [b]

[a] Instituto de Geociências, Universidade Federal do Rio Grande do Sul, Av. Bento Gonçalves, 9500 Porto Alegre, RS, Brazil
[b] Instituto de Física, Universidade Federal do Rio Grande do Sul, Av. Bento Gonçalves, 9500 Porto Alegre, RS, Brazil

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ABSTRACT

The main purposes of fission track thermochronology are unravelling the thermal histories of sedimentary basins, determining uplift and denudation rates, identifying the structural evolution of orogenic belts, determining sedimentary provenance, and dating volcanic rocks. The effect of temperature on fission tracks is well known and is used to determine the thermal history; however, the effect of pressure on the stability of tracks is still under debate. The present work aims to understand the role of pressure on the annealing kinetics of apatite fission tracks. The samples of Durango apatite used in our experiments were chosen for their international recognition as a calibration standard for geological dating. Neutron irradiation of the samples, after total annealing of their spontaneous tracks, produced induced tracks with homogeneous densities and lengths. The effect of pressure associated with temperature on fission track annealing was verified by experimental procedures using a hydraulic press of 1000 t with a toroidal chamber profile. The experiments consisted of a combination of applying 2 and 4 GPa with 20, 150, 190, 235, and 290 °C for 1 and 10 h. The annealing rate was analysed by measuring the lengths of the fission tracks after each experiment using optical microscopy. The results demonstrate that the annealing of apatite fission tracks has a pressure dependence for samples subjected to 2 and 4 GPa. However, when extrapolated to pressures of ⩽150 MPa, compatible with the normal geological context in which apatite fission track methodology is broadly used, this dependence becomes insignificant compared to the temperature effect.

1. Introduction

The fission track method relies on the spontaneous fissioning of the ^{238}U isotope, which is present in certain minerals, e.g., apatite, zircon, and titanite, in a continuous manner over geological time and on the thermal susceptibility of these defects (Fleischer et al., 1975; Naeser, 1967; Wagner, 1968, 1969).

During fission, the ^{238}U nucleus is split in two or rarely in three fragment nuclei, releasing a large amount of kinetic energy with the launch of new-born nuclei in opposite directions. Those ionising particles interact electrostatically with the mineral crystalline lattice nuclei, creating a narrow trail along its trajectory. The resulting damage consists of defects with cylindrical symmetry known as latent tracks. Fission tracks form continuously over time at a constant rate, allowing the density of the tracks to be used to estimate the amount of time elapsed since they began to accumulate (Gallagher et al., 1998; Wagner and Van den Haute, 1992).

It is noteworthy that the defects that form latent fission tracks anneal at all temperature above zero Kelvin, however the annealing visible under a microscopy as the progressive length reduction of etched fission track only occur if the sample is exposed to certain temperatures. This progressive length reduction when observed on confined track is widely used as a diagnostic tool for thermal history analysis (Gleadow et al., 1986).

Because age determination depends on two-dimensional samplings of tracks intersecting an internal surface of mineral, a reduction in the track length leads to a reduction in the track density. This relationship has allowed many experimental determinations of the track stability in terms of temperature and time, culminating in the development of models leading to reliable extrapolations of laboratory length evolution to geological time intervals (Carlson, 1990; Ketcham et al., 1999; Laslett et al., 1987).

The lithostatic pressure, assumed as principal stress that rocks are subject during burial, is a function of depth multiplied by rock density and gravity acceleration, typical lithostatic pressure under 3 km depth is about to 0.1 GPa (Gutenberg, 1959). Some rocks have a pore fluid pressure that is assumed usually as hydrostatic, in general 10 MPa/km, this will reduce the resulting effective pressure throughout the rock mass by over one-third (Ahrens, 1995;

* Corresponding author. Present address: PETROBRAS S.A., CENPES R&D Center, Rua Horácio Macedo 950, Rio de Janeiro, RJ, Brazil.
E-mail address: jaques.schmidt@ufrgs.br (J.S. Schmidt).

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Vidal et al., 2003). In some cases due the low permeability of the rock sequence the fluid cannot escape fast enough from the system, when compared with the lithostatic pressure increase, thus generating an over-pressured section. In this section pressure is higher than normal hydrostatic pressure gradient, an increment that can cause reduction by over one-sixth in the effective pressure (Nelson, 2012).

Early works concerning pressure effects on fission track stability within a host mineral (Ahrens et al., 1970; Fleischer et al., 1965, 1974, 1975; Lakatos and Miller, 1970; Naeser and Faul, 1969) indicate that, under geological conditions, they are insignificant in comparison to temperature. Fleischer et al. (1965), for example, showed that the effects of hydrostatic pressure are much less important than the effect of temperature for mineral samples.

Later, however, Wendt et al. (2002) claimed that the available data were only qualitative and therefore unreliable. On this note, they proposed the establishment of a systematic experimental procedure for the quantitative study of the annealing of fission tracks in minerals. Their main conclusion was that the annealing of apatite fission tracks is actually extremely sensitive to pressure and stress, implying that applying current annealing models to geological problems introduces significant errors in the derivation of ages, exhumation rates, and erosion rates. Pressure dependence was also reported for argon diffusion in phlogopite and Fe–Mg geothermometers in olivine (Behrends and Zhang, 2001; Giletti and Tullis, 1977; Jaoul et al., 1995). An approximate equation for the apatite fission track annealing considering the effect of pressure, presented in Vidal et al. (2003), is reproduced below to illustrate the possible importance of pressure in the annealing process: $\ln(1 - l/l_0) = 0.219 * \ln(t) + 3.87 - (35\,090 + 1.2 * (P - 1))/(8.3144 * T)$; where $t(s)$ is time, P (bar) is pressure, T (K) is temperature, l_0 is the initial length, and l the length of tracks after annealing.

In the following year, Donelick et al. (2003) presented new annealing experiments showing no significant effect of pressure under the studied conditions. Most importantly, they could not reproduce the experiments by Wendt et al. (2002) under conditions of 168 h at 250 °C subjected to pressures of 0.1 MPa to 100 MPa.

Two years later, Cruz and Chadderton (2005) extended the line of research proposed by Wendt et al. (2002) and presented a theoretical investigation by estimating the changes in the stopping power cross section due to pressure-induced changes in the electronic structure of idealised “random” fluorapatite. According to their study, the effect of pressure on the range reduction for tracks in apatite is less profound than expected.

Raman studies of Durango apatite for samples irradiated by swift heavy ions with and without pressure are described by Liu et al. (2008), Schouwink et al. (2010), and Weikusat et al. (2011). They concluded that the Raman spectrum of an irradiated sample at pressures up to 10.5 GPa suggests that pressure does increase the radiation stability of the crystalline lattice.

Thereafter, Lang et al. (2008) carried out experiments by irradiating zircon with 35 GeV relativistic heavy ions within a diamond anvil cell at 0.75 GPa and 250 °C. They reported a small increase in track length relative to artificially produced tracks at room pressure.

In the present work, lengths of etched induced fission tracks were measured, as performed in most experimental annealing studies (Barbarand et al., 2003a, 2003b; Carlson, 1990; Carlson et al., 1999; Crowley et al., 1991; Donelick et al., 1999; Laslett et al., 1987). Samples were subjected to various pressures and temperatures for different durations to determine the effects of each variable on the annealing process. An extrapolation of the experimental results over geological time is presented, permitting a better understanding of the effects of pressure under natural conditions.

2. Materials and methods

Samples consisted of slabs, 2 × 1.5 × 3 mm, from a single Durango apatite crystal from Mexico (Young et al., 1969) cut parallel to the crystallographic *c*-axis by a diamond saw. Durango apatite is a reasonable representative of fairly chlorine rich apatites with Cl = 0.12 anion per formula unit, it also presents anomalous content of Fe, As, REE and Th (Barbarand et al., 2003b), and it has been widely studied experimentally (Green et al., 1986; Green, 1988). Prior to ion irradiation, all samples were annealed at 500 °C for 24 h to eliminate existing tracks from spontaneous fission. The thermal neutron irradiation used to produce induced fission tracks was performed in the reactor at the Instituto de Pesquisas Energéticas e Nucleares (IPEN). The adopted parameters are defined in Lelarge and Goulart (2005) as follows: a flux of 2.8×10^{12} n/cm^2/s for 18 min, resulting in a fluence of $\approx 3 \times 10^{15}$ n/cm^2. The neutron dosimetry was verified with Kapton® external detectors coupled to a CN5 Corning® glass dosimeter, described in Hurford and Green (1983) and Hurford (1990), located at the top and bottom of the stack, which presented a variation of less than one standard error. Samples were ground into cylinders with a 1.5 mm radius and 2 mm height for placement inside the pressure apparatus. After experiments, sample preparation for microscopic analysis was performed in the Laboratório de Geologia Isotópica of the Universidade Federal do Rio Grande do Sul (UFRGS). Apatite fragments were embedded in epoxy resin and ground on carborundum papers (grades 220, 1200, and 4000) and then polished using 3 and 0.3 µm alumina and finally 1/4 µm diamond paste on cloth on lapwheels.

All samples were etched in a bath of 5 M HNO_3 at room temperature at 20 ± 1 °C for 20 s and then rinsed in distilled water to arrest etching, washed with water, and rinsed in alcohol, during etching the temperature was monitored with a thermometer. Fission track analysis was performed using track-in-track (TINT) confined tracks in the crystal plane parallel to crystallographic *c*-axis of the mineral, characterised by elongated etch pits parallel to the *c*-axis. The abbreviation convention proposed by Barbarand et al. (2003b) was used; therefore, the mean track length is denoted MTL and the mean track angle MTA. Horizontal confined tracks were localised with a ZEISS® Axioplan 2 optical microscope at a true magnification of 1000× with a 100× dry objective. To perform length measurements, the microscope was fitted with an Autoscan® 3-axis AS3000B stage, Trakscan® software, and the necessary hardware and associated electronics. The lengths and orientations of the TINT horizontal confined tracks were measured with a computer screen magnification of 2000× using a cursor to mark the beginning and the end of each track using the easy-length® sub function of Autoscan® software. The measuring system was calibrated with a 5 mm graticule stage with 10 µm divisions. Its precision was estimated at ±0.24 µm (>99%), obtained from 4 analyses of 100 measurements made in different orientations, totalling 400 measurements of 10 µm divisions. All length measurements were performed without prior knowledge of the run conditions and were executed at the center of the field of view within the calibration zone. Some samples were double checked by M.L.M.V. Lelarge.

3. Experimental configurations

Experiments were designed to examine pressure and temperature conditions of 2 to 4 GPa at 150 to 290 °C, with runtimes of 1 and 10 h. Apatite is stable at temperatures up to 2300 °C and pressures below 12 GPa, as described by Murayama et al. (1986) and Williams and Knittle (1995). Thus, the observed track annealing cannot be assigned to a phase change in the crystal structure hosting the fission tracks.