Chemical Geology 260 (2009) 129–137

Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

Annealing kinetics of Kr-tracks in monazite: Implications for fission-track modelling

Carsten Weise [a,*], K. Gerald van den Boogaart [b], Raymond Jonckheere [a], Lothar Ratschbacher [a]

[a] Geologisches Institut, Technische Universität - Bergakademie Freiberg, Bernhard-von-Cottastraße 2, 09599 Freiberg (Sachsen), Germany
[b] Institut für Stochastik, Technische Universität - Bergakademie Freiberg, Prüferstraße 9, 09599 Freiberg (Sachsen), Germany

ARTICLE INFO

Article history:
Received 4 March 2008
Received in revised form 28 November 2008
Accepted 13 December 2008

Editor: D. Rickard

Keywords:
Monazite
Ion tracks
Annealing kinetics
T–t-path modelling

ABSTRACT

This paper reports data related to the potential of the mineral monazite for reconstructing the low-temperature thermal histories of geological samples from confined fission-track length measurements. The data are derived from isothermal annealing experiments on 300 MeV ^{86}Kr tracks. Simplified versions of the favoured annealing models for apatite are adequate for describing the dataset. A non-compositional model predicts that: (1) track accumulation sets in when a sample cools under ~100 °C; (2) a substantial reduction (~2.5 μm) takes place at ambient and lower temperatures on a geological timescale; (3) an additional reduction of similar magnitude (~2.5 μm) takes place during exposure for a geological time span to the annealing conditions in the earth's upper crust; and (4) etching at elevated or room temperature is expected to affect the length of induced fission tracks but not that of fossil tracks.

1. Introduction

Monazite [Ce,La,REE(PO_4)] contains a suitable amount of uranium for isotopic and chemical dating with the U/Th–Pb and U/Th–He methods but, unlike other uranium bearing minerals such as apatite, titanite and zircon, it is not used for fission-track dating. Monazite is thus a promising mineral for both multi-method and multi-mineral dating that also presents a number of other interesting properties. It is a common trace constituent in igneous, metamorphic and vein rocks and occurs concentrated in river and beach placers at great distances from the source. Such fission-track ages as have been determined are young compared to independent estimates (Shukoljukov and Komarov, 1970; Gleadow et al., 2005), indicating that the track retention temperature is low. A number of monazites of unknown provenance were mounted, polished and etched in preparation for this investigation. Their low fossil track densities ($<10^7$ cm^{-2}) and uranium concentrations of the order of ~1% also argue for a low track retention temperature and for the practical feasibility of fission-track etching and counting. A similar conclusion can be attached to the results of Shukoljukov and Komarov (1970). The supposed high annealing rate is supported by the fact that the mean length (~10 μm) of the fossil confined tracks in ~1 Ma monazite is much shorter than the calculated length of the latent tracks (~19 μm; SRIM; Ziegler et al., 2008).

Fig. 1 shows that etching of the fossil tracks is isotropic. This is not thought to be due to an isotropic etch rate resulting from partial metamictization, as in titanite (Gleadow, 1978). Monazite is almost never found in a metamict state despite the high doses of self-irradiation that the lattice sustains due to its high uranium and thorium concentrations. The needle-like – as opposed to cone-shaped (Gleadow, 1978) – tracks give no indication of an increase or levelling of the bulk etch rate. Thus, the isotropic track orientations in monazite appear to be “original” and not the result of lattice deterioration, indicating that monazite is like apatite in its etching as well as its annealing properties, and that these are related to a capacity for self-repair that is not shared by the silicates. It is indeed known that both minerals are capable of a considerable measure of self-repair at low temperatures. Weber et al. (1997, 1998) determined that the rate of repair of lattice damage resulting from 1.5 MeV Kr-ions exceeds the highest damage production rates at >150 °C for monazite and >200 °C for apatite; the corresponding temperature for zircon is nearer ~1000 °C. These considerations suggest that length measurements of confined fission tracks in monazite hold significant potential for temperature–time path modelling of processes affecting the earth's upper crust.

Monazite presents a specific problem related to its high rare-earth contents, in particular the element Gd that has a thermal neutron absorption cross-section of 48,890 b (1 b = 10^{-24} cm^2; Mughabghab et al., 1981; Mughabghab, 1984; conventional thermal–neutron–fission cross-section of ^{235}U = 586 b; Holden and Holden, 1989). A test-irradiation of a monazite with 1.69 wt.% Gd with a thermal neutron fluence of 10^{13} n cm^{-2} produced no induced fission tracks due to complete fluence depression. The calculated induced track densities for 0 wt.% Gd are $N_i = \sim 2.4\ 10^9$ cm^{-3} and $r_i = 1.8\ 10^6$ cm^{-2}. None of the monazites in our collection has less than ~0.8 wt.% Gd. Published values have a wider range but no Gd-free monazite has been reported. It is conceivable to “burn up” the Gd in a monazite by neutron irradiation to clear the path for subsequent neutron-induced fission of ^{235}U. The calculations for estimating the required

---

* Corresponding author. Tel.: +49 3731 392064; fax: +49 3731 393599.
E-mail address: carsten.weise@geo.tu-freiberg.de (C. Weise).

0009-2541/$ – see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.chemgeo.2008.12.014

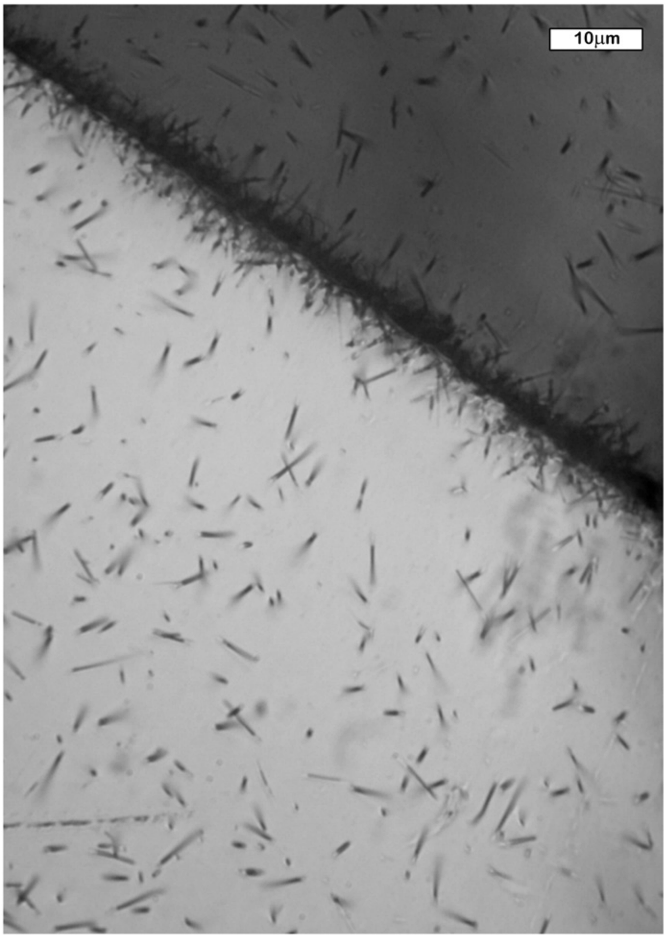

Fig. 1. Transmitted-light photomicrograph of fossil fission tracks in a ca. 1 Ma monazite of unknown provenance; note the isotropic track revelation; the relative excess of confined tracks intersecting the crack is thought to be due to natural etching by fluids circulating through the crack (Jonckheere and Wagner, 2000); it also provides independent support for the assumption the comparatively low track densities in the bulk are not the result of a low track production rate but instead of a high annealing rate.

neutron fluence are, however, forbidding and the stray activities from irradiation with such a high neutron fluence are unacceptable. Gd-free monazites can be grown (Meldrum et al., 1997; Boatner, 2002) but uranium would have to be built in for fission-track production. Is is also questionable whether the end-member compositions of artificial monazites are representative of natural monazites, as the example of He-diffusion suggests (Farley, 2007). Gd is not an obstacle to fission-track dating as such because non-track methods are available for measuring the uranium content (Hasebe et al., 2004; Fayon, 2005, 2008). Monazite fission-track dating thus necessitates neither induced-track counts nor neutron-fluence measurements, which are also problematic in the presence of Gd. Induced tracks are however also used for calibrating the annealing equations for T–t-modelling. In absence of induced fission tracks, the present annealing experiments were performed on 300 MeV ^{86}Kr-ion tracks from an ion accelerator and annealing equations were fitted based on assumed similarities with apatite.

The aim is not to establish definitive equations describing the fission-track annealing kinetics in monazite but to provide a first numerical estimate of the temperature range in which fission-track annealing takes place.

2. Experiment

The monazite crystal used in the experiments is from a pegmatite near Antsirabe (Madagascar), wherein it occurs in association with rose quartz, hatchettolite and blomstrandine (Overstreet, 1967). Its element composition (Table 1) shows it to be a (Ce(Nd,La))-phosphate with substantial substitution of P by Si. A monoclinic crystal was cut into sections parallel to the prominent (100)-face. These sections were heated for 24 h at 400 °C to erase the self-irradiation damage and fossil fission tracks, mounted in epoxy resin and polished. The sample set was irradiated with ~1.5 10^6 cm^{-2} 300 MeV ^{86}Kr ions at 50° (θ) from the normal to the sample surface at the HMI in Berlin (Fig. 2). Three isochronal (1 h; 20 h; 100 h) annealing sequences were executed. The shorter duration runs (1 h; 20 h) were carried out in a 9 mm diameter Carbolite tube furnace; the longer duration runs (100 h) were conducted in parallel in a Linn Electrotherm muffle oven equipped with an external rheostat to limit temperature fluctuations. A Dostmann dual channel digital measuring instrument equipped with two calibrated Pt-100 sensors allowed continuous temperature registration. Small samples (~1 mm^3) were wrapped in Al foil, attached to the tip of the temperature sensor and inserted into a pre-heated oven to avoid temperature gradients and ensure rapid equilibration of the sample and oven temperatures; the close contact of the sample with the sensor ensured accurate temperature measurements, irrespective of possible gradients.

The use of ion tracks as proxies for natural radiation damage in minerals is quite common; e.g. Weber et al. (1997, 1998) used 1.5 MeV Kr-ions to simulate alpha-recoil tracks in apatite, monazite and zircon (70–140 keV; ^{206}Pb–^{234}Th; Jonckheere and Gögen, 2001; Stübner and Jonckheere, 2006). Tracks from a range of accelerated ions have been used in studies related to fission-track annealing (Green et al., 1986: 220 MeV Ni tracks in apatite; Singh and Singh, 1989: 2.87 GeV Pb tracks in muscovite, garnet and quartz; Sandhu et al., 1990: 1.67 GeV Nb, 3.54 GeV Pb and 2.38 GeV U tracks in mica, apatite and zircon). The latter concluded from their experiments that the activation energies for annealing of GeV ion tracks were identical to those of fission-tracks from a ^{252}Cf source in the same mineral. In principle, this validates the use of ion tracks as proxies for fission tracks in annealing experiments, irrespective of whether or not physical significance can be attached to the calculated activation energies. The 300 MeV ^{86}Kr ions used in the present experiments are closer to fission fragments than those used in the cited studies (Table 2). ^{86}Kr is quite similar to a modal light fission fragment of ^{235}U (Lt). The mass difference is due to the neutron excess of the fission fragments and has little effect on the relative effective charge of the particles (Fleischer et al., 1975, p. 24), their respective Coulomb interactions with the lattice or, therefore, on the resulting tracks. Since nuclear charge is more important than mass, ^{86}Kr (Z=36) can even be considered a reasonable approximation of the modal heavy fission fragment, at least compared to the ions used in previous studies, which range from Ni (Z=28) to U (Z=92).

Table 1
Electron microprobe composition of the Antsirabe monazite in weight percentages (wt.%)

Element	Wt.%
Si	0.83
P	11.9
Ca	0.49
Y	2.09
La	9.16
Ce	21.2
Pr	2.77
Nd	9.60
Sm	2.88
Gd	1.87
Er	0.20
Dy	0.58
Yb	0.10
Th	8.01
U	0.50
O	26.4
Total	98.5