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Standardless fission-track dating of the Durango apatite age standard

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ABSTRACT

Five dating strategies were used for determining the standardless fission-track age of the Durango apatite. These use the same fossil-track densities but differ in the manner in which the induced-track densities are determined. A conventional age calculation, without correction for experimental factors, gives inconsistent ages with method-related differences >15%. Correcting for these factors brings the ages in line with each other and with the reference age but leaves no room for a partial-annealing correction based on the confined-track lengths. Three further reasons suggest that a length correction is not appropriate. (1) The evidence for length-based corrections is inconclusive. (2) The plateau age of the Durango apatite is consistent with its apparent fission-track age to within 1%. (3) The calculated effective etchable length of the fossil tracks agrees within error with that of the induced tracks; both are further consistent with the measured mean length of confined induced tracks. The circumstance that the (U,Th)/He ages of the accepted and proposed apatite age standards are consistent with their reference ages leaves no margin for a lowered fission-track age resulting from partial annealing, although the case of the Durango apatite itself is inconclusive because of its exceptional crystal size. It is conjectured that the shortening of the fossil tacks in the Durango apatite is due to a lowering of the track etch rate over time. In this case, annealing equations fitted to induced-track data underestimate the extent of confined-track-length reduction in geological samples.

1. Introduction: premises

The progress of the fission-track dating method can be divided into a time before and an almost equal time since the recommendation of the ζ-calibration (Hurford, 1990a, b). During the first period, it was dogged by problems associated with the ^{238}U-fission constant, neutron-fluence measurement and experimental factors related to the dating techniques (Bigazzi, 1981). Since the acceptance of the ζ-calibration and the development of apatite (T,t)-path modelling (Crowley, 1985; Green et al., 1989), it has met with great practical success. A decade before the ζ-recommendation, Bigazzi (1981) proposed two strategies for overcoming the problems of the fission-track method. One comes down to what became the ζ-calibration. The other involves solving the methodological problems in three steps: (1) achieving accurate neutron-fluence measurements, (2) eliminating errors associated with the dating techniques, and (3) establishing a reliable correction for partial annealing of the fossil tracks. The question of the ^{238}U-fission constant would then resolve itself.

Our contribution sets out from the premise that neutron-fluence measurements using the recommended metal monitors (Au, Co) and well-thermalized irradiation facilities are no longer an issue (Bigazzi et al., 1990; Curvo et al., 2013; De Corte et al., 1991, 1995; Van den haute et al., 1988, 1998). Our second assumption is that the matter of the fission constant has been settled following recent redeterminations (Eikenberg et al., 1993; Guedes et al., 2000, 2003; Suzuki, 2005; Yoshioka et al., 2005) and reassessments (Holden, 1989; Holden and Hoffman, 2000). Our work concentrates instead on steps (2) and (3) of Bigazzi's (1981) research program by using five dating methods, requiring method-specific corrections for experimental factors, for determining the standardless fission-track age of the Durango apatite.

2. Adjusted age equation

Natural apatite contains trace amounts of uranium. A fraction of the isotope ^{238}U undergoes spontaneous fission over geological time. The nuclear fragments, moving in opposite directions through the lattice, produce a single ca. 20-μm long (Bhandari et al., 1971; Jonckheere, 2003a, b), <10-nm diameter (Afra et al., 2011; Jaskierowicz et al., 2004; Lang et al., 2015; Li et al., 2014; Miro et al., 2005; Paul and Fitzgerald, 1992; Schauries et al., 2014) fossil fission track. The number of tracks per unit volume (N_S) is proportional to the age (t) and ^{238}U content (U_{238}) of the sample and the spontaneous-fission rate (λ_F) of ^{238}U (Eq. 1).

$$N_S = \lambda_F \, t \, U_{238} \quad (1)$$

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Fission tracks also form when apatite is irradiated in a reactor. The thermal neutrons in the reactor spectrum cause a fraction of the ^{235}U isotope to fission, producing induced fission tracks. The number of induced tracks per unit volume (N_I) is proportional to the concentration of ^{235}U (U_{235}) and the fission rate $R_F = \sigma\phi$, wherein ϕ is the thermal-neutron fluence and σ the effective cross-section for ^{235}U-fission by thermal-neutron capture (Eq. 2).

$$N_I = \sigma\,\phi\,U_{235} \tag{2}$$

It follows from Eqs. (1) and (2) that:

$$t = (\lambda_F)^{-1}\,(N_S/N_I)\,(U_{235}/U_{238})\,\sigma\,\phi \tag{3}$$

Eq. (1) neglects the depletion of ^{238}U by spontaneous fission and α-disintegration. An exact equation derived by Price and Walker (1963) implies that Eq. (1) is accurate to within 1% for $t < 10^{10}$ a. Eq. (2) does not account for burn-up of ^{235}U or for the production of induced tracks by epithermal- and fast-neutron fission of ^{235}U. Eqs. (1) and (2) furthermore do not consider spontaneous or induced fission of other isotopes. In practice, these are negligible for the neutron fluences required for dating apatite and for irradiations in well-thermalized reactor channels (Tagami and Nishimura, 1992; Wagner and Van den haute, 1992). Eq. (3) is adequate for dating samples up to 100 Ma.

There is a complication related to the ^{235}U-fission rate $R_F = \sigma\phi$ in Eq. (3), when ϕ is calculated from the γ-activities of co-irradiated metal-activation monitors (Au, Co). This calculation is in practice based on the "simple" Høgdahl formalism, which can cope with non-ideal epithermal-neutron spectra, and gives the conventional sub-cadmium neutron fluence ϕ_S. The Høgdahl formalism is however not suited for non-(1/v) reactions such as thermal-neutron induced ^{235}U fission. The appropriate formalism for non-(1/v) reactions is the more sophisticated Westcott convention, which includes the $g(T_n)$ correction factor (Westcott g-factor; T_n: Maxwellian neutron temperature) and uses the conventional total (Westcott) neutron fluence ϕ_W. It can be shown that in well-thermalized irradiation channels $\phi_W \approx \phi_S$ ($\phi_W = \beta\,\phi_S$). In channel 8 of the Thetis nuclear reactor (Universiteit Gent; thermal to epithermal fluence ratio: $f = 155$; epithermal spectrum parameter: $\alpha = 0.10$), used for the irradiations in this work, $\beta = 1.0025$ (Jonckheere, 1995; Verheijke, 1994) and $g(T_n) = 0.9832$ (Holden, 1999; $T_n = 27$ °C: Wagemans et al., 1988). Another consideration concerns the rare-earth-element content of the Durango apatite, in particular the Gd concentration of up to 200 ppm. Its high neutron-absorption cross-section (ca. 49,000 b) can cause fluence depression in the samples of up to ca. 4% (Naeser and Fleischer, 1975). The calculated correction factor for thermal-neutron shielding (G_{th}; De Corte et al., 1991) in the Durango sections dated here is $G_{th} = 0.986 \pm 0.001$ (Bellemans, pers. com.; Jonckheere, 2003b). Combining these factors, the fission rate $R_F = \sigma\phi$ in Eq. (3) can be rewritten as:

$$R_F = (g(T_n)\,\sigma_0)\,(G_{th}\,\beta\,\phi_S) = 0.972\,(\sigma_0\,\phi_S). \tag{4}$$

σ_0 is the conventional (2200 m/s) fission cross-section of ^{235}U ($\sigma_0 = 586 \pm 3$ b; Holden and Holden, 1989) and ϕ_S the conventional subcadmium neutron fluence calculated from the measured γ-activities of the co-irradiated metal-activation monitors using the Høgdahl formalism.

The natural isotopic ratio $I = \theta_{235}/\theta_{238}$ ($7.253\ 10^{-3}$; Cowan and Adler, 1976) is substituted for (U_{235}/U_{238}) in Eq. (3) for calculating the age of a sample. This is accurate if U_{238} and U_{235} in Eqs. (1)–(3) refer to the same elemental uranium concentration, i.e. if the fossil and induced fission-track densities, N_S and N_I, have been produced by the same concentration of uranium atoms. Natural apatite minerals often exhibit within-grain and between-grain uranium inhomogeneities. For this reason, the estimates of N_S and N_I are either each averaged over representative grain samples (multi-grain or population methods) or both measured in the same grains (single-grain or matched-areas methods; Gleadow, 1981; Galbraith, 1981).

The external-detector method (Gleadow, 1981) is the single-grain method most used for dating. The fossil tracks are counted in a polished internal apatite surface and the induced tracks in an external detector irradiated in contact with it. Thus, the fossil tracks result from uranium fission on both sides of the etched apatite surface (4π-geometry) whereas the induced tracks result from uranium fission on one side of the surface of the external detector (2π-geometry). The ratio of the fossil to induced-track densities (ρ_S/ρ_I) is therefore multiplied by a geometry factor G ($= 2\pi/4\pi$). This also applies to the re-etch method (Gleadow, 1981), where the induced tracks are counted in an external apatite surface.

The Durango apatite dated in this work consists of cm-sized crystals, which were cut into ca. ½-mm thick sections for the specific purpose of comparing the fission-track ages of basal and prism faces. Part of the analyses was carried out with a population method. In our case this did not involve different aliquots but different sections from a single crystal (Section 3). One section was annealed and irradiated for counting the induced tracks, while the adjoining section, which was neither annealed nor irradiated, was used for counting the fossil tracks. In the presence of uranium-concentration gradients, this presents a risk of systematic error. To avoid it, both sections were re-irradiated in contact with muscovite external detectors. A correction factor U can then be calculated from the induced-track densities ρ_{ED} in both external detectors and the respective neutron fluences ϕ_S:

$$U = (\rho_{ED}/\phi_S)_I/(\rho_{ED}/\phi_S)_S. \tag{5}$$

The subscripts outside the brackets refer to the irradiations of the sections with fossil (S) and induced (I) tracks. The single-grain dating methods present a similar problem. For the external-detector and re-etch methods (Gleadow, 1981), it is assumed that half the uranium that produced the fossil tracks is not available for producing induced tracks during neutron irradiation. In the presence of significant short-range uranium variation perpendicular to the apatite surface, this assumption is invalid. We have no means of accounting for it, and assume that it is negligible ($U = 1$). With the repolish method (Gleadow, 1981), there is a small offset between the sections used for counting the fossil and induced tracks due to the intervening repolish. It is again assumed that no correction is required.

N_S and N_I in Eq. (3) are volumetric track densities. In practice, the fossil and induced tracks intersecting the detector surface are enlarged by etching and their areal densities (ρ_S and ρ_I) are determined by counting under a microscope. Because of etching and observation effects, the track-counting efficiencies $[\eta q]_S$ and $[\eta q]_I$ (Jonckheere, 1995, 2003b; Wagner and Van den haute, 1992) are in general <1. For dating methods where ρ_S and ρ_I are determined in different track-registration geometries, it cannot be assumed that $[\eta q]_S = [\eta q]_I$ (Jonckheere and Van den haute, 1998, 1999, 2002). This is accounted for by introducing a procedure factor $Q = [\eta q]_I/[\eta q]_S$ and substituting $(N_S/N_I) = Q\,(\rho_S/\rho_I)$ in Eq. (3).

It is expedient to introduce an etch-time factor T for the re-etch method (Gleadow, 1981), in which the fossil tracks are etched before ($\rho_S(t_E)$; t_E: etch time) and after irradiation ($\rho_S(2t_E) + \rho_I(t_E)$). Because of the etch-time difference, it should not be assumed that the fossil tracks are revealed and identified the same in both counts ($[\eta q]_S(2t_E) \neq [\eta q]_S(t_E)$ or $\rho_S(2t_E) \neq \rho_S(t_E)$). T can be determined from step-etch experiments ($T = \rho_S(t_E)/\rho_S(2t_E)$). T is important for correcting for the effect of residual etch figures, i.e. etch pits that persist after the lower track end-points have been overtaken by surface etching (Jonckheere and Van den haute, 1996).

A fission fragment crossing from mineral to external detector leaves a damage trail in both. However, not every etched track in the mineral corresponds to one in the external detector (Iwano and Danhara,