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### Standardless fission-track ages of the IUGS age standards

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ABSTRACT

Standardless fission-track ages for four zircon, two titanite and three apatite age standards, most of them recommended by the Fission Track Working Group of the IUGS Subcommission on Geochronology, were redetermined. The age calculations used the decay constant for spontaneous fission of  $^{238}$ U ( $\lambda_{\rm f}$  = (8.5 ± 0.1)  $10^{-17}$  a<sup>-1</sup>) recommended by the IUPAC, the ratio of etchable to latent track length in both the dated minerals (zircon, titanite and apatite) and mica detectors and the 187 track counts with the external-detector and population methods. We found the resulting standardless fission-track ages are in line with results from more recent high precision geochronology including zircon U-Pb dating. These results confirm that standardless fission-track dating of zircon, titanite and apatite with the external-detector method is feasible, as well as dating of apatite with the population method, and that  $\zeta$ -calibration can be based on the zircon U-Pb ages of the current standard samples. On the other hand, there is no room for an age correction for partial annealing of the fossil tracks based on their mean confined-track lengths of the apatite, titanite and zircon age standards.

#### 1. Introduction

Almost 30 years have passed since the 1988 Besançon conference, at which the fission-track community adopted an age-standard-based calibration approach (Hurford, 1986, 1990a). After the international recommendation of the "zeta age calibration" by the Fission Track Working Group of the International Union of Geological Sciences (IUGS) Subcommission on Geochronology (Hurford, 1990a, 1990b), the fission-track method has played a role as a tool of low-temperature thermochronometry (Van den haute and De Corte, 1998; Reiners and Ehlers, 2005; Lisker et al., 2009; Iwano et al., 2013). On the other hand, three challenging problems were identified for re-establishing absolute chronometry: 1) the fission-decay constant ( $\lambda_{cl}$ , 2) absolute measurement of thermal neutron fluence and 3) track counting factors (Hurford, 1990a).

In 2000, the International Union of Pure and Applied Chemistry (IUPAC) recommended an evaluated  $\lambda_f$  value of (8.5 ± 0.1)  $10^{-17}$  a<sup>-1</sup> based on determinations performed since 1950, but excluding measurements using solid-state nuclear track detectors (Holden and Hoffman, 2000). Recent redeterminations using track detectors (Guedes et al., 2000, 2003; Suzuki, 2005; Yoshioka et al., 2005, 2006) have however supported the IUPAC value, which has since been reaffirmed by Browne and Tuli (2015). The problem of neutron-fluence ( $\phi$ ) measurements was settled earlier using dilute metal activation monitors (Au, Co) and well-thermalized irradiation facilities (De Corte et al.,

1991, 1995; Van den haute et al., 1988, 1998; Curvo et al., 2013). In addition, the IRMM-540 dosimeter glass has been developed for relative  $\phi$  measurements against a certified absolute neutron fluence (Bellemans et al., 1995; De Corte et al., 1998). It appears therefore that  $\lambda_r$  and  $\phi$  never were the major problems, emphasizing the importance of track revelation and registration as the main sources of systematic error affecting the accuracy of absolute fission-track ages (Van den haute et al., 1998), especially using the external-detector method (EDM: Gleadow, 1981).

A fission track is a damaged zone in a mineral or glass along the paths of the heavy charged nuclear fragments produced by spontaneous fission of <sup>238</sup>U or induced fission of <sup>235</sup>U. Chemical etching is used to enlarge tracks enough to be visible under an optical microscope (Price and Walker, 1962; Fleischer et al., 1975). However, the etchant is generally not capable of revealing the entire range of a fission fragment, which means that the maximum etchable length of a confined track is shorter than the length of the latent track (Fleischer et al., 1975). Fig. 1 shows a relationship between latent and etched track lengths in a mineral and co-irradiated external muscovite mica detector. Moreover, not only the etchable length but also the unetchable range varies among minerals (Jonckheere, 1995, 2003; Iwano and Danhara, 1998; Danhara and Iwano, 2013). By accounting for these factors, standardless fissiontrack dating has been re-established for apatite (Jonckheere, 2003; Enkelmann et al., 2005c; Jonckheere et al., 2015; Wauschkuhn et al., 2015) and zircon (Danhara et al., 2010; Danhara and Iwano, 2013). In

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Fig. 1. Relationship between latent and etched fission tracks in a mineral and a co-irradiated muscovite-mica external detector. (a) A correlated pair of etched induced tracks: the unetchable sections at either track termination are a different length due to the different etching thresholds of the mineral and external detector; (b) an uncorrelated track etched only in the external detector due to its lower etching threshold; (c) the whole latent track in the mineral without etching process and (d) the latent track partly etched as a confined track. The upper C-sections are longer in (a) and (b) than in (c) and (d) based on facts that not only the etchable length but also the unetchable range varies among minerals and that mica detectors have longer unetchable track range C than that in zircon.

this paper, we review a bias in fission-track counts based on the relationship between etchable and unetchable track ranges in the mineral and external detector, and demonstrate the feasibility of absolute fission-track dating of apatite, zircon and titanite by determining the standardless ages of the IUGS age standards. It is noticed that "absolute" and "standardless" have the same meaning in this paper, and that both refer to dating without recourse to age standards (except for verification purposes as in this work) but resting entirely on physical constants and measurements.

#### 2. Correction of unetchable-track-range bias

We developed a simple method for estimating unetchable ranges (range deficits; Fleischer et al., 1975) at the ends of fission tracks in minerals by using external detectors with a low threshold of track-revelation (Iwano et al., 1992, 1993; Iwano and Danhara, 1998). After etching induced fission tracks in both mineral and co-irradiated external detector and track-to-track matching (Fig. 1a), we identify uncorrelated tracks in the external detector, i.e. tracks without etched counterparts in the mineral (Fig. 1b). The observation that tracks can be etched in the external detector but not in the mineral has been explained by Jonckheere (1995, 2003). The higher induced-track densities are recorded in the material with the lower etching threshold. From the ratio of the etched track densities we can estimate the unetchable track range for each mineral (Iwano et al., 1992, 1993; Table 1). The DAP, (diallyl phthalate) plastic detector has a higher track registration efficiency than mica (Yoshioka et al., 2003), showing that fission-track densities in DAP are 1.04-1.05 times higher than in mica. This is explained by existence of a range deficit of  $\sim 0.5 \,\mu\text{m}$  at the ends of etched tracks in mica as an invisible track range (Danhara and Iwano, 2013; Fig. 1a, b). The latent track lengths were estimated by summing the unetchable track ranges and the measured mean confined-track lengths. The results for example for zircon (15.9 µm) and mica (21.5 µm), are in reasonable agreement with the calculated values

(Jonckheere, 2003) (Table 1). Chemical etching thus introduces a significant bias in measurements of the track densities of spontaneous and induced tracks. Therefore, a correction factor R (range deficit factor: Jonckheere, 1995, 2003) in the age equation is required.

Jonckheere (2003) argues that absolute fission-track dating based on the thermal neutron fluence, requires, in addition to a geometry factor (G), an experimental factor (Q) and a procedure (R). Of these factors, G is known, and Q and R can be evaluated by calculation and experiment. It is however easier to determine the combined correction factor [GQR] for a specific sample-surface/detector combination by induced-track density measurements (Jonckheere, 2003; Enkelmann et al., 2005a). In particular, Danhara and Iwano (2013) illustrated that [GQR]<sub>ED/ES</sub>, which is defined as the ratio of the measured induced track densities in an external mineral surface  $[\rho_i]_{ES}$  and co-irradiated external detector  $[\rho_i]_{ED}$ :

$$[GQR]_{ED/ES} = [\rho_i]_{ED} / [\rho_i]_{ES}$$
(1)

can be obtained most accurately by track-to-track matching between in an external surface (ES) and the attached external detector (ED) (Fig 1a, b). Our published [GQR]<sub>ED</sub>/<sub>ES</sub> values for zircon, titanite and apatite and a muscovite mica external detector are shown in Table 2. The correction factor [GQR]<sub>ED/IS</sub> for internal mineral surfaces ( $4\pi$  geometry) irradiated in contact with the same external detector ( $2\pi$  geometry), can be determined using the relationships (Jonckheere, 2003):

$$[GQR]_{ED/IS} = [GQR]_{ED/ES} [GQR]_{ES/IS}$$
(2)

$$[GQR]_{ED/IS} = [GQR]_{ED/ES} (1/2 [\eta q]_{ES} / [\eta q]_{IS})$$
(3)

where the  $[\eta q]$ -values are the combined track etching and observation efficiencies for the external (ES) and internal (IS) mineral surface. In this study, the  $[\eta q]$ -values are derived from the full etchable (3D) distributions of the surface tracks (Iwano, 1997; Iwano and Danhara, 1998) and the assumption of a minimum observable track length of 0.5 µm for zircon and mica and 1 µm for apatite and titanite. This gives

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