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Recalibration of U-doped standard glasses through uranium thin film for neutron-fluence measurements

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Abstract This work presents a recalibration of U-doped standard glasses using natural uranium thin films to be used as most dedicated neutron monitor for fission-track dating. The recalibrated U-doped glasses were used to determine fission-track ages in apatite samples from Brazilian alkaline formations (Alto Paranaíba) and standard Durango apatite. Samples were irradiated in two nuclear reactors with different characteristics and the results were compared. For well-thermalized neutron facility, metal activation monitor was also used. The ages of Alto Paranaíba arch and Durango apatite agree with those determined by other radiometric dating methods and metal activation monitors. These results suggest that the presented recalibration is suitable to be used routinely for fission-track dating studies even in a non-well thermalized neutron facility.

Keywords Neutron-fluence monitor · Dosimeter glasses · Uranium thin film · ϕ-Method · Heavy ion irradiation

Introduction

Fission-Track Thermochronology, FTT, is based on the spontaneous fission of ^{238}U, present as a trace element in natural minerals, over the geological time scales. The most applied method for calibrating is the ζ-calibration, a calibration factor that embraces the physical constants, irradiation parameters and efficiency factors and is obtained by irradiating the U-doped glasses with an age standard sample, whose age was known a priori. The ζ-calibration had been shown to be a very practical method and made possible the standardization of the fission-track dating in a time when there was a dispute on the correct value of λ_f and neutron fluence determinations were not so accurate [1]. However, the drawback of the ζ-calibration is its dependence on the age of a standard sample obtained by other dating methods. In case of apatite, the sample from Durango has been shown to yield very consistent results.

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As the dispute on the value of λ_f has been solved [2–6], the neutron fluence determinations had been shown to be accurate [7, 8] and the efficiency factors have been broadly studied [7–9], the absolute calibration yielded independent fission-track ages possessing geological meaning.

The accumulated spontaneous fission-tracks are quantified through the measurement of their superficial density, ρ_S. The ^{238}U uranium nuclei are represented by $C_{238}N_U$, where N_U is the volumetric concentration of uranium atoms in the mineral and C_{238} is the ^{238}U isotopic concentration. Taking into account that ^{238}U can decay by both alpha emission and spontaneous fission, only a fraction λ_f/λ of the ^{238}U will undergo fission and the age equation becomes:

$$t = \frac{1}{\lambda}\ln\left[1 + g\frac{(\rho_S)}{(\rho_I)}\frac{\lambda}{C_{238}\lambda_f}R_U\right] \quad (1)$$

where $\rho_{S(I)}$ is the spontaneous (induced) fission-track density; g is the factor, which embraces geometric, etching and observation efficiencies; λ is the alpha decay constant; λ_f is the ^{238}U spontaneous fission decay constant; C_{238} is the ^{238}U isotopic abundance in the natural uranium and $R_U = \Sigma R_i$ (R_i is the number of reactions per target nuclei of the ^{i}U, $i = 235, 238$). R_U is given by:

$$R_U = \sum_i C_i \int_0^\infty \sigma_i(E)\phi(E)\mathrm{d}E \quad (2)$$

In the Eq. 2, C_i is the isotopic abundance of the ^{i}U in the natural uranium; $\sigma_i(E)$ is the cross section for the nuclear reaction ^{i}U(n,f), as a function of neutron energy, E; $\phi(E)$ is the neutron fluence with energy E per unit energy.

Certainly, one of the most discussed problems within FTT is the difficulty to determine the R_U-value in a neutron facility when the traditional dosimetry is applied [10–12]. In this case, the neutron fluence is determined through metal monitor as Au and Co [7, 13–15]. For thermal energies (neutron energy less than 0.5 eV) their cross section curves are represented by $1/v$ relationship, where v is the speed of neutrons. In this energy interval, cross sections for the metal monitors reactions ^{197}Au(n,γ)^{198}Au and ^{59}Co(n,γ)^{60}Co are approximately parallel to the one of ^{235}U(n,f) reaction and the correction must be applied for the non-ideal $1/v$-dependence of the conventional (2,200 m/s) thermal neutron fission cross-section of ^{235}U [16–19]. Furthermore, in this energy interval, the cross sections for ^{238}U and ^{232}Th induced fission are negligible (Table 1).

However, for the energy range of the epithermal neutrons, resonance peaks are observed for both metal monitors and the comparison between uranium fission rates with neutron fluences measured with metal monitors become more complicated and includes other factors that must be known, such as the neutron spectrum in a specific nuclear reactor facility. Furthermore, the resonance integral of Au is so high that, even in quite well thermalized reactor, the correction for epithermal fission will be necessary because there is a significant difference in the response between the reactions ^{235}U(n,f) inside the mineral and (n,γ) metal monitors reactions [4]. In addition, epithermal neutrons also induce the ^{235}U fission with different response (diminished cross section compared to the thermal region) and the cross section for the ^{238}U induced fission is no longer negligible and can lead to error in the age (Table 1).

Table 1 The cross section values for neutron reaction considering energy interval of thermal, epithermal and fast neutrons

Isotope	Nuclear reaction	$\sigma_{thermal}$	$\sigma_{epithermal}$	σ_{fast}
^{235}U	(n,f)	584.33[a]	275[b]	1.203[c]
^{238}U	(n,f)	Negl.	0.015[b]	0.305[c]
^{232}Th	(n,f)	Negl.	Negl.	0.081[c]
^{59}Co	(n,γ)	37.13[d]	74[b]	Negl.
^{197}Au	(n,γ)	98.65[d]	1550[e]	Negl.

σ is the cross section for thermal ($\sigma_{thermal}$), epithermal ($\sigma_{epithermal}$) and fast (σ_{fast}) neutrons. The values are in barns

[a] Carlson [32]

[b] Mughabghab et al. [41]

[c] Ziip and Baard [42]

[d] Holden and Holden [43]

[e] Mughabghab et al. [44]

In the case of fast neutrons, the cross section for ^{235}U induced fission is considerably lower than thermal and epithermal neutron energies whereas the ^{232}Th and ^{238}U fission events also have to be taken into account, mainly in samples where the Th/U ratio of the mineral is relatively high (for instance, Durango apatite where Th/U $\approx$ 25).

More directly dedicated neutron dosimeters for fission-track dating are the U-doped glasses calibrated against U-thin films [20–23]. These neutron dosimeters have an advantage because in both, dosimeter and minerals, the reactions are based on uranium fission, ^{235}U(n,f), and all integrated reactions during neutron irradiation are identical, once uranium is present in natural concentrations in both dosimeter and mineral. The calibration is, therefore, independent of the particular energy spectrum of the neutron flux and the number of induced fissions per target nucleus, R_U occurring during the irradiation is directly measured. When uranium thin films are irradiated coupled with muscovite external-detector, induced fission-track density is proportional to R_U-value [22]:

$$R_U = \frac{\rho_F}{N_U^F \varepsilon^F} \quad (3)$$

In the Eq. 3, N_U^F is the number of uranium atoms per unit area of the thin film [20–23] and ε^F is the efficiency detection for muscovite attached to it. Bigazzi et al. [20]

and Hadler et al. [24] have shown through comparison with nuclear emulsion that the efficiency detection of muscovite for fission fragment sources in thin film geometry is 1.000 ± 0.025. Thus, if N_U^F is determined by electronic alpha-particle spectroscopy, the R_U-value is easily determined.

When geological samples are irradiated, neutron fluence should be around $\approx 10^{14}$–10^{15} cm^{-2} and thin films cannot be directly used to monitor the neutron fluences due to the high uranium content deposited when thin films are manufactured. Thus, under the mentioned neutron fluences, there is superposition of etched fission tracks in the muscovite external-detector attached to the thin films, which makes the counting impossible under ordinary optical microscopy. Lower uranium content thin films could not be manufactured [20]. Fortunately, U-doped glass dosimeters with suitable uranium content can be calibrated against U-thin films and then employed to monitor neutron fluences for fission track dating. Surface fission-track densities in muscovite sheets coupled with the U-doped glasses during irradiation, ρ_D, can be related with R_U through the following equation [22]:

$$R_U = \frac{\rho_D}{N_U^V \varepsilon^V} \tag{4}$$

In the Eq. 4, N_U^V is the number of uranium atoms per unit volume in the glass dosimeter; ε^V is the detection efficiency of the muscovite external-detector attached to it [22, 25]. The calibration of a U-doped glass against a uranium thin film can be done if they are irradiated together and juxtaposed. With the determination of R_U-value through Eq. 3 by using uranium thin films and measuring ρ_D, the calibration factor, $\varepsilon^V N_U^V$, can be determined through Eq. 4. The R_U-value for fission-track dating (Eq. 4) can be determined for subsequent irradiations using U-doped glasses. Thus, Eq. 1 can be re-written in terms of induced fission-track density in the muscovite external detector attached to the standard glasses (ρ_D) and the parameter of calibration ($\varepsilon^V N_U^V$) determined for each glass:

$$t = \frac{1}{\lambda} \ln \left[1 + g \frac{(\rho_S)}{(\rho_I)} \frac{\lambda}{C_{238} \lambda_f} \left(\frac{\rho_D}{N_U^V \varepsilon^V} \right) \right] \tag{5}$$

For the case of non-well thermalized neutron facilities, Th thin film can be irradiated together with apatite samples. In order to consider the Th fission, the following equation was used [22]:

$$R_M = R_U + \left(\frac{N_{Th}}{N_U} \right) R_{Th} \tag{6}$$

The R_{Th} is the event by target nuclei of the ^{232}Th, which is calculated similarly to R_U by using the Eq. 3. The (N_{Th}/N_U) is the thorium and uranium ratio in the mineral to be dated. Thus, R_M is the fraction of fission events per uranium target nucleus, considering the thorium fissions that occurred during irradiation. This parameter replaces the R_U-value in the Eq. 5.

Despite Iunes et al. [22] have already carried out such calibration, the fission-track age results are always lower than values determined, e.g., metal activation monitor [7, 8] and reference ages even when fission-track ages are corrected. The observed differences could come from the thin film calibration, from the glass calibration procedure or from efficiency calibrations necessary for dating. Curvo et al. [26] advanced in the solution of this problem firstly recalibrating the thin films using a more reliable α-spectrometry calibration instead of alpha activity detected in polyallyldiglycol carbonate (PADC) alpha-track detectors calibrated via nuclear emulsion [20, 22, 23] because this procedure makes U-thin films calibration very ingenious and etching/observation efficiencies have to be taken into account. With electronic alpha particle spectrometry, this laborious procedure can be avoided and U-thin films can be calibrated directly. In a second experiment, Curvo et al. [26] carried out joint irradiations of standard glasses, metallic monitors (Au and Co) and standard age samples. The recalibration of the thin films confirmed the previous calibration by Iunes et al. [22]. However, neutron fluences determined with the standard glasses resulted around 10 % less than metal activation monitors, implying the same differences in the ages determined for the age standard samples. Curvo et al. [26] did not recalibrate the glasses. Instead, they used the calibration carried out by Iunes et al. [22]. In this way, Curvo et al. [26] apparently isolated the problem: the standard glass calibration. Thus, the purpose of this work is to present a new calibration of the glass-dosimeter via uranium thin films calibrated through electronic alpha-particle spectroscopy. U-doped glass calibrations were carried out in a nuclear reactor position with low neutron fluence. Besides, geometry factor was determined applying exactly the same experimental conditions in which unknown samples were dated [7–9]. As it will be seen in the next sections, this new calibration, together with a well determined geometry factor [9] resulted in much more consistent results when U-doped glasses and metal activation monitor are compared. An additional test for neutron dosimetry techniques is to date samples and compare the obtained ages with ages determined by independent radiometric ages. In this work, this procedure has been applied to apatite samples from Brazilian alkaline-carbonatites (Alto Paranaíba arch) and Durango apatite (Table 2). Furthermore, Durango, Fish Canyon Tuff and Limberg samples dated by Enkelmann et al. [8] using metal activation monitor were recalculated using the calibration presented in this work. These age determinations were possible because U-doped glasses were irradiated together with such samples. The obtained results suggest that it is

Table 2 Radiometric ages of alkaline samples used in the present work

Sample	Radiometric age(Ma)	Method
Catalão	85 ± 6.9	K-Ar[a]
Tapira	87.2 ± 1.2	K-Ar[b]
Durango	31.0 ± 1.0	U-Th/He[c]

[a] Data from Gomes et al. [33]

[b] Biondi. [46]

[c] McDowell et al. [35]

Table 3 Uranium content in thin films determined by nuclear emulsion and electronic α-spectrometry

Thin film	U content ($\times 10^{16}$ atoms/cm^2)	
	Emulsion[a]	α-Spectroscopy[b]
XXVIII-2	0.24 ± 0.01	0.23 ± 0.01
XVIII-3	0.22 ± 0.01	0.21 ± 0.01

[a] Data from Iunes et al. [23]

[b] Data from Curvo et al. [26]

safe to use U-doped glasses calibrated against U-thin films and metal activation monitor for FTT applications in a geological contexts.

Methods

Calibration of the standard glasses

The uranium thin films were prepared following the procedure of Yagoda [27], starting from a solution of uranyl nitrate and parlodium dissolved in alcohol and ether. The solution is deposited on a muscovite plate. After evaporation of the volatiles, a thin film of uranium remains. The muscovite plate is then heated at 400 °C resulting in a uranium oxide film adhering to the muscovite support. The film is thin enough for fission-fragment stopping to be negligible [23, 25].

Iunes et al. [22, 23] calibrated uranium thin films using CR-39 alpha track detectors calibrated against nuclear emulsion [24]. This procedure requires determining the etching and observation efficiencies of alpha tracks in CR-39 through laborious procedures, which makes this calibration difficult to reproduce. Thus, we applied a calibration based on electronic alpha-particle spectroscopy just as Curvo et al. [26]. This laboratorial procedure is more precise and has better reproducibility once it does not depend on the etching and observer efficiencies. A 51 mm diameter detector at 16.7 mm from the sample was used. Vacuum was made in the sample/detector chamber to reduce alpha particle stopping in air. The instrumental detection efficiency for alpha particles is ca. 100 %. The geometric efficiency for each thin film was calculated through Monte Carlo simulations. The detector energy calibration was carried out using ^{241}Am (5.50 MeV) and ^{239}Pu (5.18 MeV) standard sources from the *Institute for Research, Production and Application of Radio-isotopes* of the former Czechoslovakia. Their activities in December 1980 were 0.669 kBq (±3 %) for Am and 0.816 kBq (±3 %) for Pu. The uranium concentrations determined in thin films used for calibration are shown in Table 3.

The dosimeter glasses calibrated in this work were [28]: CN-1 [U] = 39.9 μg g^{-1} ± 0.8 and [Th] = 0.42 μg g^{-1} ± 0.02; CN-2, [U] = 36.7 μg g^{-1} ± 0.7 and [Th] = 0.29 μg g^{-1} ± 0.02; CN-5 [U] = 12.2 μg g^{-1} ± 0.6 and [Th] = <0.019; and IRMM540 [U] = 15.0 μg g^{-1} ± 0.3 and [Th] is negligible (certified by Institute for Reference Materials and Measurements). For all those dosimeter glasses presented above, the uranium is in natural concentration and Th content is, in worst case, less than 2 %, which is an advantage if compared with older dosimeter glasses as, for instance, SRM 610–617, in which the U and Th concentrations are 1:1 in proportion [28].

The calibration of the dosimeter glasses was carried out in the IEA-R1 nuclear reactor, São Paulo state, Brazil. To avoid errors from fluence gradients, the standard glasses and uranium thin films were attached on either side of the muscovite external-detectors (<1 mm thickness). The nominal neutron flux was ca. 10^8 cm^{-2} s^{-1}. A low neutron flux was necessary, given the high uranium content of the thin film. Thus, after 60 h irradiation, the neutron fluence was (2.50 ± 0.08) × 10^{11} cm^{-2}, which gives convenient track densities in the external detectors attached to the thin films and the glasses. The muscovite external-detectors were etched with HF 48 %, for 90 min at 15 °C. Their track densities, ρ_D (dosimeter glass) and ρ_F (thin film) were measured with a Leitz Dialux 20 EB microscope at 500× magnification.

Preparation of the apatite samples

Fission-track dating was carried out in Durango apatite standard age and apatite samples collected from the Brazilian alkaline-carbonatite complexes associated with uplift on the boundary of the Paraná basin. The ages determined by other radiometric dating methods can be seen in Table 2, in which the applied method is specified. For Durango apatite, a large crystal was crushed and the resulting small pieces (between 250 and 350 μm) were analysed. Those grains that present basal and texturized section [29] were avoided, i.e., only surface in prismatic section or close to were used for dating. In the case of samples from Brazilian alkaline-carbonatite complexes, the

grains with size between 150 and 250 μm were used for dating and the same orientation criteria was considered for dating.

Each sample was divided in an aliquot for confined-track-length measurements and for determining the fission-track age. For external detector method, EDM, the grains were mounted in Struers Epofix resin and cured at 45 °C for ca. 24 h. The grain side of the mounts was ground with #1200 SiC paper to expose internal grain surface and polished with 6, 3, 1 and 0.25 μm diamond suspensions (Struers DiaDuo). All apatite samples were etched with 25 % HNO_3 at 25 °C during 15 s in order to reveal the spontaneous fission-tracks. After etching, the samples were covered by muscovite external-detector and irradiated in the well-thermalized "Angelschnur" position ($\phi th/\phi$ epi $\approx$ 600) of the FRM-II research reactor of the Technische Universität München at Garching. The muscovite external-detectors were etched with HF 48 %, for 90 min at 15 °C. For population method, PM, samples were divided in two aliquots, one was used to determine spontaneous fission-track density (ρ_S) and another aliquot was pre-annealed (450 °C during 24 h to reach total annealing) and irradiated with thermal neutrons in order to generate induced fission-tracks (ρ_I). The ρ_S/ρ_I ratio gives the fission-track age by population method. Apatite samples were mounted, polished and etched as described above.

The calibrated uranium glasses and metal activation monitors (IRMM-530: Al–0.1 %Au; IR MM-528: Al–1.0 %Co) were used for measuring the neutron fluence. Given the well-thermalized neutron spectrum, fission-tracks from epithermal and fast fission of ^{235}U, ^{238}U and ^{232}Th can be neglected.

The repositioning technique of Jonckheere [7] was used for performing the track counts in the apatite grains and matching areas of the external detectors. In this technique, the muscovite is replaced on the mount in the same position as during the irradiation, so that spontaneous (ρ_S) and induced (ρ_I) tracks can be counted by adjusting the focus of the microscope.

For comparison, different samples from Catalão, Tapira, as well as the Durango apatite age standard were also irradiated in the IEA-R1 nuclear reactor, São Paulo, Brazil, which has a less well-thermalized neutron spectrum than the FRM-II reactor. Due to the low uranium content, a low number of induced tracks are observed in the muscovite external-detector, making difficult the repositioning of the detectors. For this reason, these samples were dated with the population method. It is important to mention that, although the samples dated with external detector method, irradiated in the FRM-II reactor, are not the same samples dated with population method, irradiated in IEA-R1 nuclear reactor, they were collected from the same alkaline complex and must not give different fission-track ages. The track density measurements were carried out with a Zeiss Axioplan II microscope at a nominal magnification of 1000×, using dry objectives.

Heavy ion irradiation

Horizontal confined fission-tracks have been used to quantify the degree of annealing experienced by geological apatite samples. As these tracks are totally inside the volume of the mineral, it is difficult to access them by etching unless they cross superficial tracks or cleavages. However, it can be difficult to achieve satisfactory statistics when samples present a low fission-track density. A suitable way to improve the amount of horizontal confined fission tracks is to artificially produce open channels through the mineral surface. For this, the minerals can be irradiated with ^{252}Cf fission fragments or with heavy ions produced in a particle accelerator [30]. In the present work, samples were exposed to ^{78}Kr ions of 866 MeV kinetic energy. The irradiations were performed at the linear accelerator UNILAC at the GSI Helmholtz Centre (Darmstadt, Germany). The initial beam energy was reduced by mounting an aluminum degrader foil, $\sim$64.3 μm thick, in front of the samples. According to the SRIM2003 code [31], this Al degrader reduces the energy to 250 MeV and ion track ranges in apatite is $\approx$29 μm. At this stage, the maximum energy loss in the mineral surface is achieved and ion tracks are slightly larger, producing a better revelation of the confined fission-tracks, which are crossed by these opened (^{78}Kr) channels.

Horizontal confined fission track measurements in samples dated by EDM were carried out using an optical microscopy Zeiss Axioplan II, with aid of a digitalizing table and the CCD camera, using a nominal magnification of 1500×, dry. For samples dated by PM, a nominal magnification of 1500×, dry.

The geometric factor *g*

The geometric factor *g* has an important role in Eq. 5 when EDM is applied. This occurs because spontaneous fission-track density in apatite is determined under 4π-geometry, whereas induced fission-track density is determined under 2π-geometry. However, intrinsic differences between revelation by etching and observation under optical microscopy makes the *g*-value $\neq$ 0.5, thus, this parameter should be measured in the same conditions samples are dated [7, 9].

To determine *g*, two prismatic sections of Durango apatite were heated for 48 h at 450 °C to erase the spontaneous fission-tracks. After this, the sections were mounted and polished. Muscovite external-detectors were attached and the assemblies were irradiated in the Thetis nuclear reactor of Institute for Nuclear Sciences, Ghent University (Belgium).

After irradiation, the apatite samples were re-polished so that a 4π-geometry was obtained. The tracks in the apatite and muscovite external-detector were etched as described above. The experimental procedures used for determination of g are exactly the same as used with the external dating detector method. In practice, g is calculated as the ratio of the track densities in the muscovite-external detector and internal apatite surface [7]. For population method, g is considered as 1 because geometry and efficiencies of revelation/observation are the same for both, spontaneous and induced fission-track counts.

Metal activation monitors

The γ-activities of the Au (Al–0.1 % Au; IRMM-530) and Co (Al–1.0 % Co; IRMM-528) monitors were measured on a Ge(Li) detector connected to a 4000 channel γ-spectrometer. At least 50,000 counts were collected. The thermal neutron fluences were calculated in the Høgdahl convention using the nuclear constants in De Corte et al. [14]. A correction was applied for the non-ideal $1/v$-dependence of the conventional (2,200 m/s) thermal neutron fission cross-section of ^{235}U [16–19] and for neutron self-absorption in the monitors.

Results and discussion

Calibration factors

Table 4 summarizes the results of the dosimeter glass calibration based on Eqs. 3 and 4. The final $N_U^V\varepsilon^V$ values are the averages of the values from two irradiations (Table 4): CN-1: $N_U^V\varepsilon^V = (1.43 \pm 0.05) \times 10^{14}$ cm^{-2}; CN-2: $N_U^V\varepsilon^V = (1.26 \pm 0.05) \times 10^{14}$ cm^{-2}; CN-5: $N_U^V\varepsilon^V = (0.40 \pm 0.03) \times 10^{14}$ cm^{-2}; IRMM540: $N_U^V\varepsilon^V = (0.52 \pm 0.02) \times 10^{14}$ cm^{-2}. The values for irradiations I-1 and I-2 are consistent but lower than the $N_U^V\varepsilon^V$ values presented by Iunes et al. [22]: 2.8 % for CN-1, 6.7 % for CN-2, 4.1 % for CN-5 and 3.0 % for IRMM-540. Table 5 shows the result of the measurements of the geometric factor. The mean ($g = 0.595 \pm 0.013$) for the two samples used to calculate fission-track ages.

Table 5 Measurements of geometry factor

Sample	N_{ap}	$\rho_{ap} \pm 1\sigma$ (10^5 cm^{-2})	N_{ed}	$\rho_{ed} \pm 1\sigma$ 10^5 cm^{-2})	$g \pm 1\sigma$ (N_{ap}/N_{ed})
1	3801	4.07 ± 0.07	2262	2.42 ± 0.05	0.595 ± 0.017
2	3822	4.09 ± 0.07	2275	2.43 ± 0.05	0.595 ± 0.020

$N_{ap(ed)}$ is the amount of counted fission-tracks in apatite (muscovite external-detector); $\rho_{ap(ed)}$ is the superficial fission-track density in apatite (muscovite external-detector)

Age determination

The fission-track ages (Table 6) were calculated using EDM (Eq. 5). The neutron fluences were determined with metal monitors and with U-doped glasses calibrated against uranium thin films. The ϕ-values were calculated with Eqs. 2 and 4 and using $\sigma = 584.33$b [32].

As can be seen in Table 6, the ϕ-values determined with U-doped glasses are in agreement within 1σ with those determined with metal activation monitor. This result

Table 4 Calibration of uranium doped standard glasses

Thin film	Irradiation	N_{IF}	$\rho_F \pm 1\sigma$ (10^5 cm^{-2})	$R_U \pm 1\sigma$ (10^{-10})	Glass	N_{IG}	$\rho_D \pm 1\sigma$ (10^4 cm^{-2})	$N_U^V\varepsilon^V \pm 1\sigma$ (10^{14} cm^{-2})
XXVIII-2	I-1	9545	3.05 ± 0.03	1.34 ± 0.04	CN-1	2,841	1.97 ± 0.04	1.47 ± 0.06
						1,248	1.92 ± 0.05	1.43 ± 0.06
					CN-5	572	0.56 ± 0.02	0.39 ± 0.02
	I-2	5980	3.21 ± 0.04	1.41 ± 0.05	CN-1	1,315	1.96 ± 0.05	1.39 ± 0.06
					CN-2	1,253	1.75 ± 0.02	1.24 ± 0.05
						917	1.68 ± 0.06	1.19 ± 0.06
XVIII-3	I-1	2377	2.98 ± 0.06	1.39 ± 0.05	CN-2	2,849	1.75 ± 0.03	1.26 ± 0.05
						1,829	1.79 ± 0.04	1.28 ± 0.06
					IRMM540	1,067	0.74 ± 0.02	0.53 ± 0.03
						1,332	0.76 ± 0.02	0.55 ± 0.03
	I-2	6153	3.28 ± 0.04	1.53 ± 0.05	CN-5	569	0.60 ± 0.03	0.41 ± 0.02
					IRMM540	949	0.73 ± 0.02	0.48 ± 0.02

I-1 and I-2 are different irradiations; $N_{IF(IG)}$ is the number of induced fission-tracks counted in muscovite external-detector attached to the thin film (dosimeter glass); $\rho_{F(D)}$ is the fission-track density in muscovite external-detector attached to the thin film (dosimeter glass); $N_U^V\varepsilon^V$ is the calibration factor for the standard uranium glass

Table 6 Fission-track age determined with U-doped glass dosimeter using external detector method, EDM (irradiated at the FRM-II reactor)

Sample	N_S	$\rho_S \pm 1\sigma$ ($\times 10^5$)	N_I	$\rho_I \pm 1\sigma$ ($\times 10^5$)	ϕ_{metal} (10^{15} cm^{-2})	t_{metal} (Ma)	$\phi_{U\text{-}glass}$ (10^{15} cm^{-2})	$T_{U\text{-}glass}$ (Ma)	L (μm) (N)
DUR2	1974	1.97 ± 0.05	4605	4.61 ± 0.07	2.63 ± 0.10	33.0 ± 1.9	2.58 ± 0.08	33.1 ± 2.0	14.6 ± 0.1 (68)
DUR3	9870	2.19 ± 0.02	1343	0.30 ± 0.08	0.129 ± 0.003	27.8 ± 1.6	0.127 ± 0.005	28.0 ± 1.7	14.6 ± 0.1 (68)
Catalão	1460	2.34 ± 0.06	1360	2.18 ± 0.06	2.63 ± 0.10	82.0 ± 6.0	2.58 ± 0.08	82.6 ± 5.3	14.3 ± 0.1 (139)
Tapira	623	0.87 ± 0.03	539	0.74 ± 0.03	2.63 ± 0.10	88.7 ± 6.2	2.58 ± 0.08	88.9 ± 7.4	14.2 ± 0.2 (64)

$N_{S(I)}$ is the amount of counted fission-track in apatite (muscovite external-detector); $\rho_{S(I)}$ is the spontaneous (induced) track density; ϕ is the neutron fluence determined by metal activation monitor (metal) the glass dosimeter (U-glass). L is the average of spontaneous fission-track length. t is the fission-track age in Ma

$\lambda = 1.55125 \times 10^{-10}$ a^{-1} Lederer and Shirley [45], $\lambda_f = 8.5 \times 10^{-17}$ a^{-1} Holden and Hoffman [2] and $C_{238} = 0.99275$

Standard U-doped glass used was CN-5 with the calibration showed in Table 4

differs from those presented by Curvo et al. [26] where the ϕ-values determined by U-doped glasses are always lower and reach up to 14 % of difference. This agreement is reflected in the fission-track ages. For the Catalão and Tapira samples from the Alto Paranaíba arch, the ages based on U-doped glass and metal activation monitors agree with each other and with the literature values for other radiometric methods (Table 2). These results are also supported by Gomes et al. [33] and Eby and Mariano [34] where fission-track ages are in good agreement with K–Ar determination within experimental error.

The Durango apatite ages for both neutron fluence values (U-thin film and metal monitor activation) (Table 6) are acceptable compared with the radiometric dating, e.g. McDowell et al. [35]; (U-Th)/He: 31.0 ± 1.0 Ma), Green [36] (K–Ar feldspar: 31.4 ± 0.5 Ma). This agreement is because Durango apatite and samples from Alto Paranaíba are characterized by a fast cooling. Thus, even ages determined with lower closure temperature methods (as the case of the fission-track thermochrology or (U-Th)/He) must give the same results of the methods that have higher closure temperature. Our values for Durango apatite also agree with those obtained by Van den haute et al. [13] and Jonckheere [7] with the fission-track ϕ-method (Au and Co monitors).

The Tapira, Catalão and Durango apatites were also dated after irradiation in the less well-thermalized IEA-R1 reactor, São Paulo (Brazil) using PM. The results are shown in Table 7. As discussed above, for the samples from Alto Paranaíba arch, the ages agree with literature values and also with data reported in Table 6. For Durango apatite, the fission-track ages determined in Tables 6 and 7 are also in agreement with each other. The average of fission-track ages determined in four Durango apatite samples (Tables 6, 7) is 30.4 ± 3.5 Ma, well in agreement with the reference value of 31.4 ± 0.5 Ma within one error bar. Comparing with the previous dating by Iunes et al. [22] and Curvo et al. [26], the ages obtained in this work is closer to the reference age. Part of this difference comes from the new calibration factors for the standard glasses, which are 2.8–6.7 % less than those presented by Iunes et al. [22]. The effect of these smaller calibration factors are older ages (see Eq. 5). The more careful determination of the geometry factor [9] do the rest of the work. Combined these two improvements make fission-track ages consistent with reported ages dated independently by other dating method and dated using metal activation monitors.

For irradiation in the nuclear reactor IEA-R1, Th thin film was irradiated together with Durango apatite samples taken into account their high Th/U ratio (Th/U = 25). In

Table 7 Fission-track age determined with U-doped glass dosimeter (irradiated at the IEA-R1 reactor)

Sample	N_S	$\rho_S \pm 1\sigma$ ($\times 10^5$)	N_I	$\rho_I \pm 1\sigma$ ($\times 10^5$)	$L \pm 1\sigma$ (n)	$L_0 \pm 1\sigma$ (n)	L/L_0	$t \pm 1\sigma$ (Ma)
DUR-4	2087	1.88 ± 0.04	3165	8.17 ± 0.14	14.8 ± 0.3 (38)	16.3 ± 0.12 (89)	0.90 ± 0.02	31.7 ± 6.0
DUR-5	2530	1.93 ± 0.04	3135	9.31 ± 0.17	14.8 ± 0.3 (42)	16.3 ± 0.12 (112)	0.90 ± 0.02	28.6 ± 5.4
Tapira	602	2.93 ± 0.11	490	2.72 ± 0.12	14.9 ± 0.3 (21)	16.6 ± 0.79 (19)	0.89 ± 0.05	86.9 ± 7.0
Catalão	761	1.31 ± 0.06	793	1.35 ± 0.05	13.9 ± 1.0 (13)	15.9 ± 0.95 (22)	0.87 ± 0.08	78.3 ± 4.2

$N_{S(I)}$ is the amount of spontaneous (induced) fission-track counts; $\rho_{S(I)}$ is the spontaneous (induced) track density. $L_{(0)}$ is the average of spontaneous (induced) fission-track length; n is the amount of horizontal confined fission-track. It is important to mention that L values presented in this work are in agreement with showed in Table 6. t is the fission-track age in Ma

Standard U-doped glass used was IRMM540 with the calibration showed in Table 4

order to consider the Th fission by fast neutrons, Eq. 6 was used [22].

For Durango apatite dated in Table 7, the R_U value = 1.17 ± (0.06) × 10^{-8} and R_{Th} determined by Eq. 3 is 1.05 ± (0.06) × 10^{-10}. Thus, the R_M value is 1.18 ± (0.09) × 10^{-8}. This result indicates that even the contribution of ^{232}Th fission by fast neutrons would be less than 1 % and the fission-track ages would change by the same amount and do not differ from the presented results. This probably occurs because, for epithermal neutrons, the cross section for ^{235}U fission is 275b whereas for ^{232}Th fission the cross section is negligible. For fast neutrons, the cross section for ^{232}Th fission is 0.081b, a lower value compared with ^{235}U cross section. As the case of Tapira and Catalão samples, the R_M-value is also negligible due to low Th/U ratio, between 0.09 and 0.10 μg g^{-1}.

Work published by Enkelmann et al. [8], using the absolute approach was applied by directly measuring the thermal neutron fluence with metal activation monitors. In order to compare our results with theirs, data from Enkelmann et al. [8] were used to re-calculate FT age by U-doped glasses calibrated in this work. Figure 1 shows the relationship between FT dating determined with U-doped glasses and metal activation monitor [7, 8]. For Fish Canyon Tuff (FCT) the age distributions show (1:1) relationship between metal activation monitor and U-doped glasses. The same result is observed for Limberg (Lim), i.e., there is no any fluctuation in FT age that distinguishes results determined by metal activation monitor from U-doped glasses. For the case of Durango (Dur), it can be seen more dispersion in age distribution: FT age determined by metal activation monitor is slightly higher than U-doped glasses, however, it is easy to realize that this small portion of dispersion is related to only one observer (B.W), i.e., it is possible that this results is only due to criteria of analysis of the observer. Finally, considering the mean FT dating determined by U-thin films, there is no any significant difference of the reference age for these samples (FCT = 27.5 ± 0.24 Ma determined by K-^{39}Ar dating of biotite [37]; Lim = 16.8 ± 1.0 Ma for apatite and 16.5 ± 1.0 Ma for titanite, both determined by (U-Th)/He dating [38]; Dur = 31.44 ± 0.18 Ma, ^{40}Ar/^{39}Ar dating and 31.0 ± 1.0 Ma, (U-Th)/He dating [35]). For FCT the obtained FT age is 27.0 ± 0.9 Ma, for Lim the age is 17.1 ± 0.7 Ma and for Dur the age is 30.8 ± 1.0 Ma. By comparison, the ratio between U-doped glasses age ($\phi_{thin\text{-}film}$) and reference age ($\phi_{ref.}$) is $\phi_{thin\text{-}film}/\phi_{ref.}$ = 0.98 for FCT and Dur and $\phi_{thin\text{-}film}/\phi_{ref.}$ = 1.02 for Lim. This variation is around 2 %, significantly below all associated errors. Such results suggest that both neutron dosimeters (metal activation monitor and U-doped glasses calibrated this work) are suitable to be used routinely for fission-track dating studies.

Finally, the thermal histories of Tapira and Catalão were modeled with the program HeFTy (version 1.7.5) [39], using the spontaneous horizontal confined fission-track shown in Fig. 2. The fanning curvilinear annealing equation [40] was assigned. Only one comprehensive Monte Carlo box was initially chosen to search for general trends in thermal history model. For the segment parameter, values of two for "Halve segments" and "Episodic" for "Randomizer style" were used. Furthermore, the initial confined induced fission track (L_0) was determined for each sample and used for determining the thermal histories. This parameter is important to quantify with good confidence the degree of annealing of the spontaneous fission tracks in unknown-age samples. Therefore the determination of L_0 is essential for accurate thermal history modeling [9]. As it can be seen in Fig. 3a, the thermal history from Catalão, is the simplest cooling episode, characterized by the fast passage through the PAZ. On the other hand, the sample from Tapira (Fig. 3b) shows a linear cooling and reaches the upper limit of Partial Annealing Zone (PAZ) at about 70 Ma. These results agree with Eby and Mariano [34], where they concluded that these complexes were emplaced at shallow levels and with fast cooling to the low temperatures (characterized by total annealing zone).

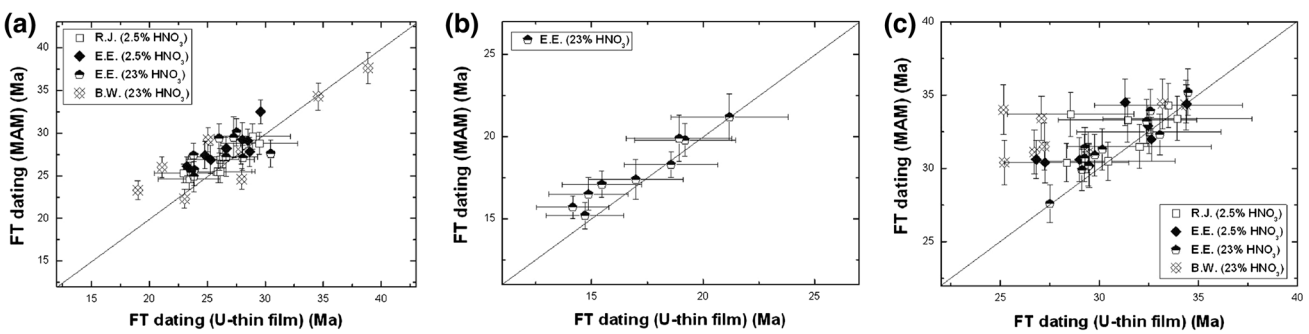

Fig. 1 FT age distribution determined by U-doped glasses calibrated against U-thin films and metal activation monitor using data from Enkelmann et al. [8]. The observer description is: R.J. (Raymond Jonckheere); E.E. (Eva Enkelmann) and B.W. (Bastian Wauschkunh). **a** Fish Canyon Tuff; **b** Limberg; **c** Durango

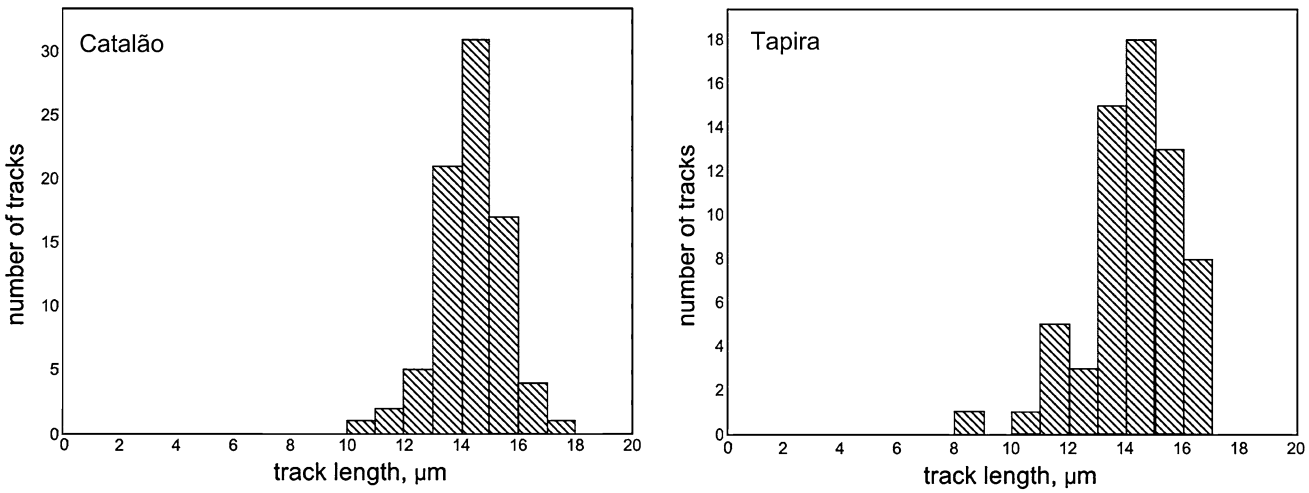

Fig. 2 Horizontal confined spontaneous fission-track distribution of the samples from Catalão and Tapira alkaline rocks

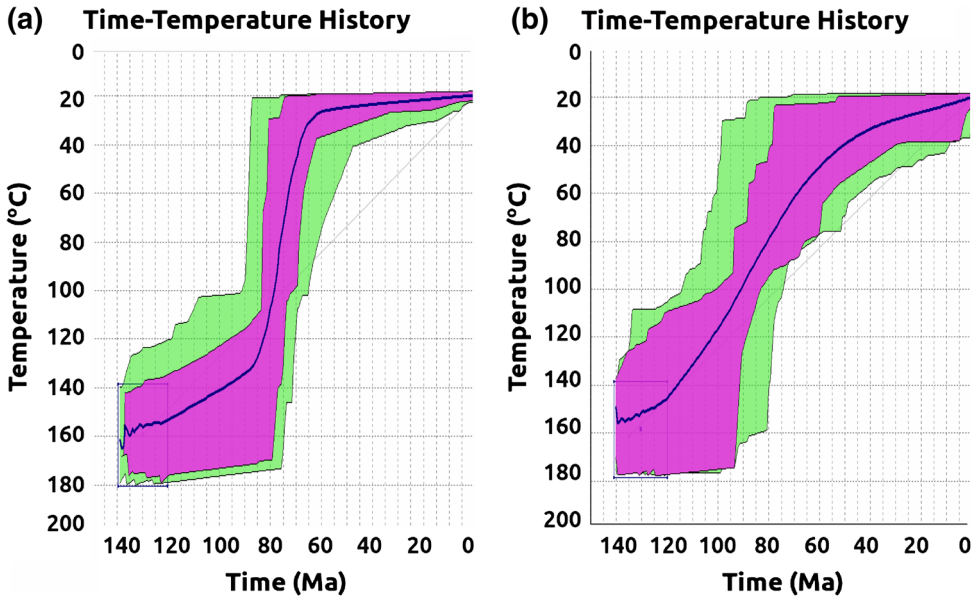

Fig. 3 Thermal history reconstruction: **a** Catalão; **b** Tapira

Conclusion

In this work, the U-doped glasses were recalibrated against U-thin films to be used as more dedicated neutron dosimeters for fission-track dating. The standard apatite Durango and Alto do Paranaíba arch samples (Catalão and Tapira) were dated in this work by the fission-track method using this dosimetry and metal activation monitors. The fission-track ages determined with recalibrated U-doped glasses differ from those presented in Iunes et al. [22] and Curvo et al. [26] and are in agreement with metal activation monitor and ages obtained by other radiometric methods. This result suggests that U-doped glasses calibrated against U-thin films and metal activation monitors can be applied for fission-track thermochrolonogy. Curvo et al. [26] have already shown that the thin film calibration carried out by Iunes et al. [22] was accurate. The main contribution of this work is to present a more accurate glass calibration, which, along with the improved determination of the geometry factor [9], yields more reliable fission-track ages.

Ages of Durango samples obtained using the ϕ-method (Au and Co dosimeters) in a well-thermalized nuclear reactor [7, 8, 13] are in agreement with the ones obtained through U-doped glass monitors (calibrated against uranium thin films). These fission-track ages are also in agreement when determined using the not-so-well thermalized neutron flux (Cd ratio for Au around 6) of the IPEN–CNEN nuclear reactor of São Paulo. Concerning induced fission in uranium, it should be noted that the employment of the neutron dosimetry by uranium thin

films implies that the same nuclear reaction, U(n,f), produces fission tracks inside the mineral to be dated and in the dosimeters. For this reason, this methodology can be also used in less well-thermalized neutron facilities.

Finally, fission-track dating of samples dated in Enkelmann et al. [8] were recalculated using the recalibration factors presented in this work. These results of Durango, Fish Canyon Tuff and Limberg are in very good agreement with metal activation monitor and referenced ages. This result suggest that metal activation monitor and U-doped glasses calibrated against U-thin films are suitable to be used routinely for fission-track dating studies. Despite this work present fission-track ages in apatite samples, the U-doped glass dosimeter can be applied in all minerals to be dated once this approach does not depend on standard samples. The only recommendation is that geometry factor must be determined for each mineral to be dated, mainly because this is a parameter that takes into account the efficiency of each observer.

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