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TÍTULO/**TITLE**/: **On the use of thin Natural Uranium Film Dosimetry in Mineral Dating by the Fission Track Method**

Thematic Session : Nuclear Geophysics- Oral (20 min)

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Cordialmente
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ABSTRACT

One important step of the experimental procedure used in the dating of minerals by the Fission Track Metod (FTM) is the thermal neutron irradiation of the mineral to be dated. Since the beginning of FTM, a well - accepted neutron dosimetry has not been found, being this matter intensively discussed in many papers. The employment of thin natural uranium films as neutron dosimeter is an opportune alternative because, in this case, only the U-235 (n,fission) reaction takes place both inside the dosimeter and the mineral to be dated.

We observed that the thin uranium films made by us present uranium content (N'_u) that is constant for $N'_u \geq 2 \times 10^{13}$ cm^{-2} and decreases with time for $N'_u \leq 10^{14}$ cm^{-2}. This fact imposes some restrictions to the direct employment of our films as neutron dosimeters in FTM; however, they can be used indirectly, i e. to calibrate other natural uranium loaded dosimeters, like CN1 and CN2 Corning glasses, which are being crescently employed in FTM in the last years.

INTRODUCTION

In order to measure the age of a mineral, T, by FTM, a suitable amount of the mineral is divided in two fractions. In one of them, after a proper etching, one measures the spontaneous track density, ρ_s, which is related to T by the well-known radioactive decay law. The other one is irradiated in a nuclear reactor with a suitable fluence of thermal neutrons, ϕ, and, after etching, is used to one measures the induced track density, ρ_i. Combining spontaneous and induced equations, the usual equation for fission track age calculation is obtained (Price and Walker, 1963):

$$T = \frac{1}{\lambda_a} \log\left(1 + \frac{\rho_s \lambda_a \sigma \phi I}{\rho_I \lambda_f}\right) \quad (1)$$

where:

λ_a is the decay constant for the alfa emission of U-238;

σ is the cross section of U-235 fission induced by thermal neutrons;

λ_f is the decay constant for the spontaneous fission of U-238, and

I is the ratio U-235/U-238.

Neutron dosimetry is the main source of systematic errors of the FTM. This matter has been intensively discussed since the beginning of the last decade (for instance, Green and Hurford, 1984; Crowley, 1986; Van der Haute, 1988; Tagami and Nishimura, 1989).

Recently, the I.U.G.S. Subcommission on Geocronology standardized the experimental procedures to be followed when mineral dating is performed through FTM (Hurford, 1990). An important recommendation of this Subcommission was that only well thermalized neutron facilities should be employed. However, such facilities are not always found (with a flux of the necessary magnitude) in the nuclear reactors the FTM researchers can access, specially for those who live in third world countries.

The incertainties in dating results that can be caused by the use of not so well thermalized neutrons facilities are greatly reduced if thin films of natural uranium[1] are employed as neutron dosimeter (Iunes, 1990). This ocurrs because the neutron dosimeter and mineral to be dated work based on the same nuclear reaction U-235(n, fission) (Hadler et al., 1981; Hadler, 1982; Green and Hurford, 1984).

In this way, thin uranium films can be very useful in FTM. However, we observed that films with very low uranium content aren't stable. They lose uranium with time. In this work, this caracteristic of the uranium films we manufacture is discussed.

EXPERIMENTAL

The thin uranium film neutron dosimeter is made up of a uranium thin film coupled to a

[1] Thin here means a thickness of natural uranium where fission fragments lose a negligible amount of energy when they traverse it.

suitable fission fragment detector (usualy a muscovita mica sheet with low uranium content). When such assembly and the mineral to be dated are irradiated together, the number of induced fission per U-235 target nucleus of the mineral, $\phi\sigma$ from equation (1), is described by (Iunes, 1990):

$$\sigma\phi = \frac{\rho_I^F}{N_U^F \epsilon_F C_{235}} \qquad (2)$$

where:

ρ'_i is the induced track density in the muscovite sheet coupled to the thin uranium film (Bigazzi, 1991).

ϵ_f is the muscovite detecting efficiency for fission fragments originated from a thin uranium film coupled to it(Bigazzi, 1991).

C_{235} is the U-235 isotopic abundance in natural uranium.

Equation (1), with $\sigma\phi$ given by equation (2), was used to one obtains the age of three obsidians samples originated from Mullumica and Callejones (Ecuador) and Monte Arci (Italy). For each sample three complete age measurements where performed. For each dating, the experimental procedures adopted to one measures ρ_s and ρ_i (sample mount in resin, polishing, etching, etc.) are described elsewhere (Osorio, 1990).

Irradiations were performed at the 14B-5 neutron facility of the IPEN/CNEN nuclear reactor, São Paulo, Brazil, where the neutron flux presents a $4{,}87\times10^{11}$ cm^{-2} s^{-1} magnitude and the Cd ratio (for Au) is 6,1.

RESULTS AND DISCUSSION

The obsidian samples studied in this work had been previously dated by FTM (Bigazzi et al., 1992., Arias et al., 1986 and Osorio, 1987). In these works, conventional neutron dosimetry (Au foils and/or NIST standard glasses) was employed. The resulting apparent ages are shown in Table I.

In Table II, one can observe the data concerning the three age mesurements performed for the three obsidian samples analysed by us. From the left to right, the columns contain irradiation date (month/year), ρ_i', ρ_i, apparent age using $\lambda_f = 7{,}03\times10^{-17}$ a^{-1} and apparent age using $\lambda_f = 8{,}46\times10^{-17}$ a^{-1}. Concerning these two most accepted λ_f values (Bigazzi, 1981; Hadler, 1982), being this work essentially dosimetric, we prefer to make no choice and used both. Other constants used to calculate the apparent ages were the folowing: $C_{235} = 7.2004\times10^{-3}$ (Browne et al., 1978); $I=7.2527\times10^{-3}$ (Cowan and Adler, 1976) $\lambda_\alpha = 1.55125\times10^{-10}$ a^{-1} (Jaffey et al., 1971) and $\epsilon_f = 1.00 \pm 0.02$ (Bigazzi et al., 1991). Callejones, Mullumica and Monte Arci obsidians were irradiated together with the films Z, R and T, respectively. Their uranium content, $N'_g(Z)$, $N'_g(R)$ and $N'_g(T)$, are shown beside the name of the related obsidian sample. These values were measured in February, 1991. In this table, it is assumed that the tracks present Poissonian distribution, being σ the mean standard deviation. σ of the apparent ages was obtained by usual error propagation.

Comparing the apparent ages shown in Table I and II it is clear that lesser ages were systematically obtained in this work and, for each obsidian sample, successive age measurements presented decreasing age results. The reason for this fact is shown in Table III. As each pair thin film-obsidian sample was irradiated together, its track density ratio, ρ'_i / ρ_i, should give a constant value. However, in Table III it is observed that that ratio decreases with time, for the three studied obsidians, for irradiations performed successively at time intervals of one month. Then, it is very probable that the thin uranium films are losing their uranium content with time, and the reason for this fact can be related with the very low uranium content of Z, R and T films.

These films present thicknesses of approximately one uranium atom diameter, if the uranium content is homogeneously distributed over all the area of each thin film. However, having some experience in manufacturing thin uranium films of different thicknesses, we know uranium atoms present a tendency to form small clusters (Iunes, 1990). In a cluster only few uranium atoms making up the botton of the cluster are chemically bound to the supporting muscovite sheet. As consequence, an uranium cluster, as a whole, is weakly by bound to the muscovite. Then, if the clusters contain a ponderable fraction of the total uranium content of the film, it is possible a measurable decrease in its uranium content with time, as observed.

It was experimentally known by us that films much thicker ($\approx 0{,}02$ µm) than R, Z and T ones present constant uranium content. Then, our plans to use thin uranium film dosimetry in FTM are meaningful if the uranium thickness range, where our films work reliably, permits mineral

dating by FTM.

To study this, the uranium content of films presenting different thicknesses was calibrated again. These results are shown in Table IV, where there are, also, the results of the first calibration of each film, performed at the time of the film manufacturing. The results shown in Table IV indicate that the technique our group have used to make uranium films (Iunes, 1990) is suitable for films that present $N'_u \geq 2x10^{11}$ cm^{-2} . However, for $N'_u \leq 10^{11}$ cm^{-2} , the uranium content seems to decrease with time.

For $N'_u \approx 2x10^{11}$ cm^{-2}, ρ'_i (see equation 2) can be measured without difficulties for fluences up to $\approx 10^{14}$n/cm^2 (for greater fluences, it is more difficult to measure ρ'_i because of the high superficial density of tracks). However, $\Phi \approx 10^{14}$ n/cm^2 is usually not enough to perform mineral dating by FTM (Fleischer et. al., 1975).

To overcome this restriction to high fluences, three alternatives can be followed:

i) Thin uranium films with $N'_u \leq 10^{11}$ cm^{-2} can be used if their uranium content is calibrated before and after each time they are employed. Of course, this procedure very time consuming, being, therefore, not recommendaded for routine use in FTM;

ii) Thin uranium films with $N'_u \geq 2 \times 10^{11}$ cm^{-2} can also be employed as primary neutron dosimeters, i e., to calibrate other natural uranium loaded dosimeters, like CN1 and CN2 Corning glasses (Iunes, 1990). These glasses, by their turn, can be used to measure neutron flounces greater than 10^{11} cm^{-2} (Hurford and Green, 1983) , which is enough to perform the dating of most minerals that can be dated by FTM. Other interesting characteristic of this use of thin films as primary dosimeters is that neutron irradiations will produce the same number of induced fission per U-235 target nucleus both in the secondary dosimeters (glasses) as in the primary ones (thin films).

iii) To improve the procedure of manufacture of our thin uranium films so that constant uranium content films can be obtained for all the uranium range that is necessary to perform FTM dating.

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Sample	(Apparent age ± σ) x (10^3a)	Reference
Mullumica Abrigo 2	1,58 ± 0,09	Bigazzi et al.,1992
Callejones Sample 3	1,70 ± 0,09	Bigazzi et al.,1992
Monte Arci (no 6)	26,5 ± 2,0	Arias et al., 1986
	26,7 ± 2,6	Osorio, 1987

Table I – Age of the obsidian samples obtained in other works.

Irradiation date	(ρ_i' ± σ) x($10^3 cm^{-2}$)	(ρ_i ± σ) x($10^3 cm^{-2}$)	(ρ_s ± σ) x($10^3 cm^{-2}$)	(App. age ± σ) x(10^3 a) λ_1	λ_2
a) Callejones – N'_s (Z) = (3,32 ± 0,19) x($10^{13} cm^{-2}$)					
08/91	1,436 ± 0,030	1,950 ± 0,047	4,39 ± 0,39	1,40 ± 0,14	1,17 ± 0,12
09/91	1,523 ± 0,078	2,544 ± 0,080	4,73 ± 0,40	1,21 ± 0,13	1,01 ± 0,11
10/91	1,189 ± 0,040	1,887 ± 0,048	4,57 ± 0,37	1,25 ± 0,12	1,04 ± 0,10
b) Mullumica – N'_s (R) = (7,52 ± 0,19) x ($10^{13} cm^{-2}$)					
08/91	3,922 ± 0,082	2,295 ± 0,059	4,29 ± 0,36	1,40 ± 0,13	1,16 ± 0,11
09/91	3,084 ± 0,073	1,944 ± 0,046	4,17 ± 0,35	1,24 ± 0,12	1,03 ± 0,10
10/91	6,720 ± 0,220	4,740 ± 0,013	4,21 ± 0,38	1,13 ± 0,12	0,94 ± 0,10
c) Monte Arci – N'_s (T) = (3,74 ± 0,11) x ($10^{13} cm^{-2}$)					
08/91	1,303 ± 0,033	1,328 ± 0,033	45,8 ± 1,90	17,20 ± 1,00	14,27 ± 0,87
09/91	1,360 ± 0,027	1,673 ± 0,046	43,9 ± 1,80	13,67 ± 0,81	11,36 ± 0,67
10/91	1,821 ± 0,067	3,508 ± 0,099	43,5 ± 2.70	8,64 ± 0,72	7,18 ± 0.60

Table II– Data concerning three successive apparent age determinations performed in the studied obsidians. The symbols are specified in the text.

Irradiation date	$(\rho'_1/\rho_1) \pm \sigma$		
	Z film/ Callejones	R film/ Mullumica	T film/ Monte Arci
08/91	0,736 ± 0,024	1,709 ± 0,057	0,981 ± 0,035
09/91	0,598 ± 0,036	1,547 ± 0,051	0,813 ± 0,035
10/91	0,630 ± 0,026	1,418 ± 0,061	0,519 ± 0,024

Table III- Ratio between ρ_1' and ρ_1, for the studied samples.

Thin film	Irradiation date (month/year) and $(N_\alpha' \pm \sigma) \times (10^{13}\ cm^{-2})$	Irradiation date (month/year) and $N'_\alpha \pm \sigma\ (\times 10^{13}\ cm^{-2})$
XXIII-3	(09/88) 70,5 ± 1,9	(03/93) 74,7 ± 2,8
XVII-4	(09/88) 16,27 ± 0,47	(04/93) 15,91 ± 0,91
XVII-1	(09/88) 2,270 ± 0,048	(10/92) 2,224 ± 0,067
A	(02/91) 0,0812 ± 0,0026	(02/93) 0,0587 ± 0,0032

Table IV- Calibration results of thin films presenting different uranium contents (N_α').