Employment of Thin Thorium Films in Fission Track Neutron Dosimetry

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Received in February 24, 1995

In tliis work, a neutron inonitor based on the 232Thinduced fission was developed. lts iinportance on mineral dating by tlie Fission Track Method is described. The monitor is constitutecl by a thin film of tlioriuin juxtaposed to a muscovite mica sheet. Its calibration was accomplished by measuring its a-activity using a nuclear emulsion. By measuring track lengthis in tliis cletector, tlie 232Tliand 212Pocan be clistinguislied from the otlier a-emitters of the tliorium series and tlie effects of tlie emanation of 220Rn from tlie film could be observed. Such effects make difficult the calibration of the film by its total a-activity, making preferable to perforin it by measuring only the 232T11a-activity. As tlie chemical separation age of tlie thorium employecl to malie the filrn is important in the study of the 220Ri1emanation, a method for obtaining this parameter was also developed.

I. Introduction

Mineral dating by tlie Fission Track Method[1] (FTM) has been often employed, mainly due to its practicality. The possibility of its application in studies concerning oil exploration[2] lias attracted the attention of researchers all over the world. However, problems related to the value of the ^{238}U spontaneous fission clecay constant[3] aiid to tlie so-called neutron dosimetry[4] led tlie specially designatecl subcommission of the Interiiational Union of Geological Sciences[5] to recorninend that the dating by using the FTM should be performed by adopting one of the two suggested procedures. One of them[6] introduces a connection between tlie FTM and otlier dating methods, wliile the otlier one is an absolute calibration[7]. In both cases, however, the neutron irradiatioris, which are needed in order to obtain the mineral age by tlie FTM, must be made in well tliermalized positions, wliich are not commonly found in nuclear reactors, specially in the third world. In this way, tlie application of the FTM is restricted to researchers having access to reactor facilities containing well thermalized positions.

In previous works[8,9], a neutron monitor was studied (thin film of natural uranium), which minimise the problem of tlie absolute calibration relatecl to the employment of positions with poor neutron thermalization. In this work, another monitor, wliich is a complement of the previous one, is studied. This monitor is constituted by a sheet of a Solid State Nuclear Track Detector (SSNTD) for fission fragments juxtaposed to a thin film of thorium. The term "thin" means a film presenting a negligible self-absorption for fission fragments (and a-particles). Muscovite mica.is a.SSNTD that can be usecl as a cletector of fission fragments. In this case, a recommended chemical etching is: 48% HF, 15°C, for 90 min. These conditions are very suitable for track

observation in a transmitted-light optical microscope under a $\sim$ 500x nominal magnification, producing, for instance, the maximum number of developed tracks[10].

The experimental procedures employed in the manufacture of the thin films of thorium (thorium oxide deposited on a flat basis, which presents a low concentration of thorium and uranium) are similar to those adopted for the manufacture of thin films of uranium[8,9].

In the following sections, the procedures concerning the employment of thorium films in the FTM and the calibration of these monitors are described.

II. Theoretical considerations

When a mineral is irradiated with neutrons in a nuclear reactor, the track density of induced fissions in the mineral, ρ_I, can be written as[11]:

$$\rho_I = N_U \epsilon_{235} R_M \qquad (1a)$$

where

$$R_M = C_{235}A_{235} + C_{238}A_{238} + \left(\frac{N_{Th}}{N_U}\right) A_{232} \; ; \qquad (1b)$$

where N_U and N_{Th} are the number of uranium and ^{232}Th atoms, respectively, per unit volume, in the mineral; ϵ_{235} is tlie ratio between the number of induced fission traclis, per unit area, observable in the surface of the mineral after the chemical etching and the number of fissions per unit volume; C_{235} and C_{238} are the isotopic concentrations of ^{235}U and ^{238}U in natural uranium, respectively; and A_{235}, A_{238} and A_{232} are the number of fissions per target nucleus of ^{235}U, ^{238}U and ^{232}Th, respectively, occurred during the irradiation.

The main purpose of the neutron dosimetry in the FTM is the determination of R_M.

In well thermalized positions, the two last terms of the right side of Eq. 1b vanish and the first one can be written as $\phi_0 \quad \sigma_0 \quad g_{235}$, where ϕ_0 is the conventional fiuence of thermal neutrons, σ_0 is the conventional cross section of the ^{235}U for fission by thermal neutrons and g_{235} is the Westcott's parameter for ^{235}U. σ_0 is known and ϕ_0 can be determined by using activation monitors[7]. g_{235} is given in tables as a function of the neutron teniperature in the irradiation position[12].

In poorly thermalized positions, the employment of thin films of natural uranium leads to the determination of the sum of the two first terms of the right side of equation 1b[8]. When this monitor is irradiated with neutrons in a nuclear reactor, the track density of induced fissions, ρ_U, observed in a detector juxtaposed to it during the irradiation, can be given by:

$$\rho_U = N_U^F \epsilon^F (C_{235}A_{235} + C_{238}A_{238}) \; , \qquad (2)$$

where N_U^F is the number of uranium atoms per unit area in the film; and ϵ^F is the ratio between the superficial track density observed on the detector after the chemical etching and the number of fissions per unit volume in the film.

In this way, by knowing N_U^F and ϵ^F, one can determine $(C_{235}A_{235} + C_{238}A_{238})$ if ρ_U i~measured. The procedures concerning the determinations of N_U^F and ϵ^F are described in Ref. [8].

The ratio thorium/uranium in the mineral (N_{Th}/N_U) can be obtained by different methods[13]. Thus, if N_{Th}/N_U is known, from equations 1b and 2 it can be seen that R_M can be determined by using, besides the dosimetry by thin films of natural uranium, other one that allows the quantification of the number of fissions per target nucleus of ^{232}Th, that has taken place during the irradiation.

When a thin film of thorium, juxtaposed to a SSNTD appropriate to detect fission fragments, is irradiated with neutrons in a nuclear reactor, the track density of induced fissions, ρ_{Th}, which is observed on the detector surface after a suitable chemical etching, can be written as

$$\rho_{Th} = N_{Th}^F \epsilon^F A_{232} \; , \qquad (3)$$

where N_F is the number of ^{232}Th atoms per unit area in the film.

In this equation, the value of the detection efficiency of the SSNTD for fission fragments originated from the thin film of thorium was assumed to be the same as in the case of a thin film of uranium. This can be justified based on the similar characteristics (charge, mass and energy of the fission fragments) of both fission processes. This assumption is also reinforced by the following experimental result: in a previous work[14], it was observed that the efficiencies for the detection of

fission fragments from ^{235}U and ^{238}U in artificial glass are equal.

By Eq. **3,** it can be seen that it is possible to determinate A_{232}, by knowing N_{Th}^{F} and ϵ^{Th}, if ρ_{232} is measured. The determination of N_{Th}^{F} is described in the next section.

When the ratio thorium/uranium in the mineral is unknown, its determination can be accomplished by the employment of two sets of thin films of natural uranium and thorium. However, an irradiation inside a cadmium box is needed.

Cadmium has a very high cross section for the absorption of thermal neutrons. Thus, if the mineral to be dated and the thin films of natural uranium and thorium are placed inside a cadmium box and irradiated together, the superficial fission track densities, in the mineral and in the detectors juxtaposed to the thin films, can be written, respectively, as:

$$\rho_{I}^{Cd} = N_{U}\epsilon_{235}\left[C_{235}A_{235}^{Cd} + C_{238}A_{238}^{Cd} + \left(\frac{N_{Th}}{N_{U}}\right)A_{232}^{Cd}\right], \tag{4}$$

$$\rho_{U}^{Cd} = N_{U}^{FCd}\epsilon^{F}\left[C_{235}A_{235}^{Cd} + C_{238}A_{238}^{Cd}\right], \tag{5}$$

$$\rho_{Th}^{Cd} = N_{Th}^{FCd}\epsilon^{F}A_{232}^{Cd}, \tag{6}$$

where the superscript Cd indicates that the irradiation was made inside a cadmium box. Combining these equations with the equivalent ones that are obtained when another set of mineral plus thin films is irradiated without Cd, equations 1 to **3,** the following expression can be written:

$$\frac{N_{U}}{N_{Th}} = \frac{\left(\frac{\rho_{U}}{N_{U}^{F}}\right) - \left(\frac{\rho_{I}}{\rho_{I}^{Cd}}\right)\left(\frac{\rho_{U}^{Cd}}{N_{U}^{FCd}}\right)}{\left(\frac{\rho_{I}}{\rho_{I}^{Cd}}\right)\left(\frac{\rho_{Th}^{Cd}}{N_{Th}^{FCd}}\right) - \left(\frac{\rho_{I}}{N_{I}^{F}}\right)} \tag{7}$$

From Eqs. 1, 2, **3** and 7, it can be seen that the employment of thin films of natural uranium, in conjunction with natural thorium ones, makes possible the obtainment of RM, independently of the neutron thermalization at the irradiation facility used for the dating of minerals by the FTM.

It should be noted that the fissions of ^{232}Th are induced by fast neutrons. This means that the thin films of thorium can be useful in a not well explored field of reactor physics[15]. However, such an application is not the purposes of this work.

III. On the determination of N_{Th}^{F}

In order to determinate N_{Th}^{F}, the $K0$ Ilford nuclear emulsion was employed as an a-particle detector. At our conditions of storage and processing, this detector presents a detection efficiency, ϵ_{α}, equal to 1.00 ± 0.02, for a-particles with energies greater than 2.2 MeV[9,16]. In addition, as the track length in nuclear emulsion depends on the a-particle energy, this detector can be used as an a-spectrometer.

In Fig. **1,** one can see the track length distribution measured in a nuclear emulsion sample that was coupled to a thin film of thorium for a certain exposition time and, then, developed. It can be noted *a* separation between the a-tracks emitted by ^{232}Th (4.01 MeV), which are the tracks having lengths lower than 19 μm, and the α-tracks from the other α-emitters belonging to the thorium series (energies higher than 5.42 MeV). In this way, N_{Th}^{F} can be determined by measuring the track density, ρ_{Th}, corresponding to the lengths lower than 19 μm. Therefore, in the case of an emulsion exposed to a thin film of thorium during the period of time t, ρ_{Th} can be written as:

$$\rho_{Th} = \lambda_{232}N_{Th}^{F}\frac{\epsilon_{\alpha}}{2}t \tag{8}$$

where λ_{232} is the a-decay constant of ^{232}Th.

In this equation, the factor 1/2 represents the fact that only a half of the a-particles emitted by the film reaches the juxtaposed emulsion.

The measurement of N_{Th}^{F} by means of the activity of ^{232}Th is very laborious because it requires the measurement of the track length distribution. However, the determination of N_{Th}^{F} by means of the total α-activity can be influenced by a systematic error because the sixth radionuclide of the thorium series (^{220}Rn) is a noble gas that can emanate from the film.

As there are no results concerning the emanation of ^{220}Rn from thin thorium films in the literature, in this work this subject was also studied aiming at the determination of N_{Th}^{F}.

Taking into account the half-lives of the radionuclides belonging to the thorium series, it can be shown, by using the basic equations of the radioactive decay,

that the ^{228}Th activity reaches the ^{232}Th one when the age of the thorium (period of time elapsed since the chemical separation of the thorium from the rock) solution is approximately 50 years. By the other hand, considering a certain amount of ^{228}Th as an isolated system, it can be also shown that the radioactive equilibrium (between ^{228}Th and its daughters) is attained in approximately 25 days. Thus, if the ^{220}Rn emanation is negligible, when a nuclear emulsion is juxtaposed to a thin film of thorium (separated at least 25 days before) for a period of time t, the total α-track density, ρ_α, can be written as:

$$\rho_\alpha = N_{Th}^F \lambda_{232}(1+5K)\frac{\epsilon_\alpha}{2}t \ , \qquad (9a)$$

with

$$K = \frac{N_{228}^F \lambda_{228}}{N_{Th}^F \lambda_{232}} \ , \qquad (9b)$$

where N_{228}^F is the number of atoms of ^{228}Th per unit area in the film; and λ_{228} is the α-decay constant of ^{228}Th.

In this equation, it was taken into account that the thorium series is made up by seven α-emitters, being, however, the sum of the α-activities of the ^{212}Bi and ^{212}Po equal to the activity of the ^{228}Th.

The ratio between the activities of the ^{228}Th and the ^{232}Th can be given as a function of the age of the thorium, t_s, according to the following equation:

$$K = 1 - \frac{\lambda_{228}}{\lambda_{228}-\lambda_{Ra}}(e^{-\lambda_{Ra}t_s} - e^{-\lambda_{228}t_s}) \qquad (10)$$

where λ_{Ra} is the β-decay constant of ^{228}Ra.

From this equation, it can be seen that K can be obtained by knowing t_s. In this way, N_{Th}^F, can be determined by using the equation 9a, if ρ_α is measured. In the case of the thorium oxide employed in this work, t_s was unknown. However, analysing a nuclear emulsion loaded with this substance in 1983, it was possible to measure the value of the parameter K, and, by employing equation 10, to obtain the value of t_s corresponding to the loading made in 1983. By adding to this t_s the period of time elapsed between 1983 and the exposition of the emulsion to the thin film of thorium (equation 9), the present value of t_s was obtained, making possible the determination of the present value of K by using Eq. 10.

If a thorium loaded emulsion is developed after a suitable storage time (the age of the loaded emulsion, counted immediately after the end of the loading process), one can observe single tracks and stars, which are double, triple, etc. α-particle tracks starting at a common point inside the emulsion. Stars are originated by the α-decay of a radionuclide and its decay products during the storage time. If the development is performed after a storage time, t_d, varying between approximately 800 h and 1300 h, it can be shown[17] that:

i) All the single tracks can be attributed to the α-decay of ^{232}Th and ^{228}Th,

ii) All the quintuple stars are generated by the decay of ^{228}Th and its α-emitter daughters;

iii) Tracks from α-particles emitted by ^{232}Th are present only as the single tracks;

iv) Tracks from a-particles emitted by ^{228}Th are present only as the single tracks and belonging to quintuple stars.

Taking into account these considerations, the following equation can be deduced:

$$\rho_s + \rho_Q = N_{Th}^E \lambda_{232}(1+K)\epsilon_\alpha t_d \ , \qquad (11)$$

where, ρ_s and ρ_Q are the superficial density of single tracks and quintuple stars, respectively; and N_{Th} is the number of atoms of ^{232}Th per unit area in the emulsion.

Three pieces of the emulsion loaded in 1983 were developed when t_d was equal to 888, 1080 and 1248 h. As the mean result of these three developments, it was obtained:

$$\frac{\rho_s + \rho_Q}{t_d} = (1.194 \pm 0.024) \times 10^{-2} cm^{-2} s^{-1}$$

N_{th}^E was determined by gravimetry; the result was $(4.514 \pm 0.010) \times 10^{15}$ cm^{-2} and the value of λ_{232} is known[18] as $1.558 \times 10^{-18} s^{-1}$.

Thus, substituting these values into Eq. 11, it was obtained a value of K in 1983 of 0.698 ± 0.048.

By considering that the values of λ_{228} and λ_{Ra} are, respectively, 1.141×10^{-8} s^{-1} and 3.813×10^{-9} s^{-1}[18], and using, the value of K obtained above, in the Eq.(10), the age of the thorium solution, t_s, in 1983 was obtained. The result was 13.0 ± 1.0 years.

The α-activity of the thin film of thorium related to the present work was measured by coupling it to

an piece of unloaded emulsion for a suitable exposition time. This was performed in 1993. In this way, considering that $t_s = 23.0 \pm 1.0$ years, the present value of K could be found by using the equation 10, what resulted in $K = 0.906 \pm 0.015$.

From these results, if the ^{220}Rn emanation effect is negligible, N_{Th}^F can be obtained through Eq. 9a, by measuring ρ_α/t.

The piece of thin film of thorium employed in this work (Th-3) was juxtaposed to a nuclear emulsion in March, 1993, for 21.433 h. The measurement of ρ_{Th} in the emulsion (performed by using a Leitz microscope, with a 12.5 x 100 nominal magnification) resulted in $(8.82 \pm 0.51) \times 10^3$ cm^{-2}. Using these results in Eq. 8, yielded $N_{Th}^F = (1.468 \pm 0.090)$ x 10^{17} cm^{-2}. On the other hand, the result of the measurement of p, was (4.09 ± 0.11) x 10^3 cm^{-2}. Then, according to Eq. 9a, one would be expect $N_{Th}^F = (1.231 \pm 0.045) \times 10^{17}$ cm^{-2}. Therefore, the determinations of N_{Th}^F by using Eqs. 8 and 9a are not in agreement. This means that the ^{220}Rn emanation effect can not be neglected.

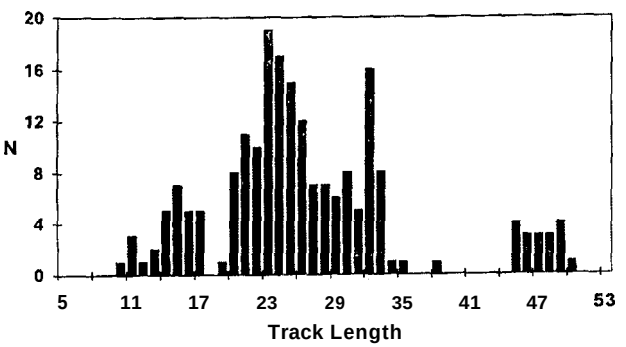

Figure 1. Figure 1: Track length distribution, in μm, measured in an unloaded nuclear emulsion sample coupled to a thin film of thorium for a suitable exposition time. N is the number of tracks.

In Fig. 1, it can be noted that the tracks due to ^{212}Po (8.78 MeV), with lengths higher than 41 μm, are separated from those due to the other a-emitters. This rneans that the track density due to ^{212}Po, ρ_{Po}, can be measured separately.

The relationship among p, ρ_{Th}, ρ_{Po} and the track density due to the α-particles emitted by ^{220}Rn, ρ_{Rn}, can be written as:

$$\rho_\alpha = \rho_{Th}(1 + 2K) + 2\rho_{Rn} + \frac{\rho_{Po}}{0.663} \qquad (12)$$

In this equation, due to the small half-life of ^{216}Po (0.16s), it was assumed that this radionuclide and its parent, ^{220}Rn, are in radioactive equilibrium. In addiction, it was considered that 66.3% of the ^{212}Bi atoms decay into ^{212}Po by emitting β-particles and 33.7% into ^{208}Tl by emitting α-particles. As the half-lifeof ^{212}Po is very short (0.30 μs), this radionuclide is in equilibrium with its parent, ^{212}Bi. Then, the last term of the right side of 12 gives the track density due to the a-particles emitted by ^{212}Bi and ^{212}Po.

The measurement of ρ_{Po} resulted in $(3.22 \pm 0.31) \times 10^3$ cm^{-2}. Substituting this result into 12, $\rho_{Rn} = (5.62 \pm 0.96)$ x 10^3 cm^{-2} was obtained, which is significantly lower than $K\rho_{Th} = (7.99 \pm 0.48)10^3$ cm^{-2}, the expected value without the ^{220}Rn emanation. This result confirms that the ^{220}Rn emanation constitutes an obstacle for the determination of N_{Th}^F by the measurement of ρ_α, using the equation 9a.

It should be noted that ρ_α could be employed in conjunction with ρ_{Po} in order to determinate N_{Th}^F, if it is admitted that the ^{220}Rn activity is equal to the suin of the a-activities of ^{212}Bi and ^{212}Po. However, it is not certain that the emanation rate of ^{220}Rn is the same before and during the period of time the emulsion is juxtaposed to the thin film of thorium. In the case of a decrease in the emanation rate of ^{220}Rn, the excess of this radionuclide retained in the film will be in equilibrium with ^{216}Po, however, the same doesn't occur from the beginning of the juxtaposition with its other decay products.

IV. Conclusion

The employment of thin films of thorium in the FTM neutron dosimetry seems to be very promising. This technique and the neutron dosimetry by thin film of uranium are complementary. If both techniques are employed together, R_M (Eq.1) is accurately determined, taking into account the uranium and thorium content of the mineral to be dated. This determination of R_M is independent of the neutron thermalization at the employed irradiation facility.

The calihration of a thorium film by means of its total a-activity is not suitable once the ^{220}Rn emanation effect can not be neglected. However, this calibration can be performed by measuring separately the activity of ^{232}Th, which can accomplished with the employment

of nuclear ernulsions

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