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On energy absorption effects in uranium and thorium thin films

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Abstract

The use of U and Th thin films, attached to detectors of fission tracks, allows determining the neutron fluence in nuclear reactors. In principle, the parameters involved in neutron fluence determinations through U and Th thin films that might be influenced by effects related with fission fragments and α particle energy absorption are: (i) the detection efficiency of nuclear emulsions for α particles, (ii) the detection efficiency of muscovite mica for fission fragments from thin films, ε^F and (iii) the detection efficiency of nuclear emulsions for fission fragments, used to estimate ε^F. Fission fragments are more ionizing than α particles and are more influenced by energy absorption. We show that fission fragment energy absorption effects are negligible in case of U and Th thin films employed in neutron fluence determinations for fission-track (FT) dating.

Keywords: Thin film; Uranium; Thorium; Neutron fluence; Fission-track dating

1. Introduction

The use of U and Th thin films, attached to detectors of fission tracks, allows determining the neutron fluence in nuclear reactors (Bigazzi et al., 1999). It has been shown that neutron fluence determinations based on these films are adequate for fission-track (FT) dating (Iunes et al., 2002, 2005; Osório et al., 2003).

In principle, the parameters involved in neutron fluence determinations through U and Th thin films that might be influenced by effects related with fission fragments and α particle energy absorption are: (i) the detection efficiency of nuclear emulsions for α particles (nuclear emulsions are used to determine the U or Th content of thin films) (Iunes et al., 2004); (ii) the detection efficiency of muscovite mica for fission fragments originated from thin films, ε^F (muscovite is used as detector, attached to thin films during neutron irradiations used for FT dating) (Bigazzi et al., 1991) and (iii) the detection efficiency of nuclear emulsions for fission fragments, used to estimate ε^F (Hadler et al., 1996).

Fission fragments are more ionizing than α particles and thus more susceptible to absorption. The aim of this work is to prove that fission fragment absorption is negligible in U and Th thin films employed in neutron fluence determinations for FT dating.

2. Theoretical considerations

The number N of fission fragments which escape from a unit of area of surface of a film with thickness d, smaller than the fragment mean range, R (Fig. 1), is given by

$$N = \int_{V_1} A\frac{\cos\theta}{4\pi r^2}\mathrm{d}V + \int_{V_2} A\frac{\cos\theta}{4\pi r^2}\mathrm{d}V \tag{1}$$

where A is the number of fissions occurred in unit volume of the film, $\cos\theta/4\pi r^2$ is the probability that, in case of a fission occurred at a distance r and with an angle θ, one of the fragments intercepts the unit of area. V_1 and V_2 are the volumes produced by rotating the areas S_1 and S_2 around the z-axis over an angle 2π.

Eq. (1) can be written in spherical co-ordinates:

$$N = \frac{A}{4\pi}\left\{\int_{\phi=0}^{2\pi}\int_{r=0}^{(d/\cos\theta)}\int_{\theta=0}^{(\mathrm{ar}\cos(d/R))}\cos\theta\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi + \int_{\phi=0}^{2\pi}\int_{r=0}^{R}\int_{\theta=\mathrm{ar}\cos(d/R)}^{\pi/2}\cos\theta\sin\theta\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi\right\} \tag{2}$$

where A was assumed to be constant through the film.

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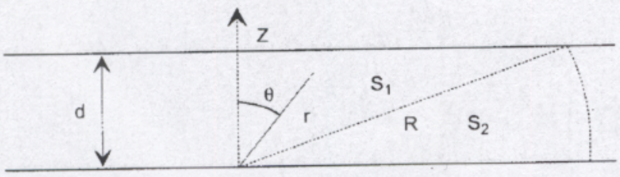

Fig. 1. Transversal section of a thin film of thickness d. R is the mean range of fission fragments.

U and Th films employed in FT dating have a thickness $< 0.1\,\mu m$. In the present work the experimental results related with energy absorption for fission fragments was determined for a UO_3 loaded film with a thickness of 0.098 μm. The thickness was measured through the estimate of the U content via α activity, considering that UO_3 has a density of $7.29\,g/cm^3$ and a molecular weight of 286.07.

Measurements of the mean range of fission fragments in U_3O_8 (Segrè and Wiegand, 1946) indicate that for UO_3, which has a density very similar to U_3O_8, $R \sim 14\,\mu m$. In a U loaded nuclear emulsion, where the U atoms are in suspension in the emulsion and absorption does not exist, the mean range of fission fragments, $\overline{R}_T$, is $(12.07 \pm 0.09)\,\mu m$ (Hadler, 1979). Assuming that the range of fission fragments in a nuclear emulsion is approximately the same as in UO_3, the mean range of fission fragments in an emulsion that was attached to a UO_3 film during irradiation with neutrons, $\langle R_E \rangle$, is given by:

$$\langle R_E \rangle = \overline{R}_T - \langle r \rangle \tag{3}$$

where $\langle r \rangle$ is the mean length of the path run by fission fragments through film before penetrating into emulsion.

Using Eq. (2), $\langle r \rangle$ can be written in the following way:

$$\langle r \rangle = \left\{ \int_{\phi=0}^{2\pi} \int_{r=0}^{(d/\cos\theta)} \int_{\theta=0}^{(ar\cos(d/\overline{R}_T))} r\cos\theta\sin\theta\,dr\,d\theta\,d\phi + \int_{\phi=0}^{2\pi} \int_{r=0}^{\overline{R}_T} \int_{\theta=ar\cos(d/\overline{R}_T)}^{\pi/2} r\cos\theta\sin\theta\,dr\,d\theta\,d\phi \right\} \Bigg/ \left\{ \int_{\phi=0}^{2\pi} \int_{r=0}^{(d/\cos\theta)} \int_{\theta=0}^{(ar\cos(d/\overline{R}_T))} r\cos\theta\sin\theta\,dr\,d\theta\,d\phi + \int_{\phi=0}^{2\pi} \int_{r=0}^{\overline{R}_T} \int_{\theta=ar\cos(d/\overline{R}_T)}^{\pi/2} \cos\theta\sin\theta\,dr\,d\theta\,d\phi \right\}$$

therefore

$$\langle r \rangle = \frac{\ln(d/\overline{R}_T) + 1/2}{2/d[1 - (d/\overline{R}_T)] + 1/\overline{R}_T} \tag{4}$$

In case of a film with a thickness of 0.098 μm, considering $\overline{R}_T = 12.07\,\mu m$, using Eq. (4) one obtains $\langle r \rangle = 0.26\,\mu m$. This value, considering Eq. (3), is equivalent to $\langle R_E \rangle = 11.81\,\mu m$. One can conclude that theoretically the energy absorption causes a reduction of about 2% in the mean length of fission tracks originated from the UO_3 film with a thickness of $\sim 0.1\,\mu m$.

3. Experimental results and discussions

The determination of the length of fission fragment tracks was made through the measurement under a DIALUX 20EB Leitz/Wetzlar microscope at 1250× in oil. This microscope is equipped with micrometric devices of the parameters x, z' and T shown in Fig. 2. After chemical revelation the thickness of a nuclear emulsion suffers a reduction by a factor ~ 2 of its original value. The original thickness of emulsions used in this work (Ilford KO emulsions) certified by the manufacturer was 50 μm. The range of a fission fragment can be expressed as

$$R_E = (x^2 + z^2)^{1/2} \quad \text{where } z = \frac{50\,\mu m}{T} z'$$

The mean length of fission tracks determined in this work for the nuclear emulsion attached to the 0.098 μm UO_3 film, $\langle R_E \rangle = (11.58 \pm 0.21)\,\mu m$, is in agreement with the estimate made in the above section.

The distribution of track lengths measured for computing $\langle R_E \rangle$ is shown in Fig. 3. All tracks are longer than 6.5 μm, indicating optimal observation conditions. It had been shown (Hadler et al., 1996) that in case of α particles from ^{147}Sm, which have a mean length of $(8.34 \pm 0.05)\,\mu m$, with a significant fraction of tracks lesser than 6.5 μm, the observation efficiency in nuclear emulsion could be considered equal to 1. Therefore, one can argue that the observation efficiency is not influenced by the small energy absorption of fission fragments originated from 0.098 μm thick film. This result can be extended to α particles from natural U and Th, which are less ionizing and have longer ranges than fission fragments.

The track lengths of Fig. 3 cluster into two peaks corresponding to heavy fragments (shorter tracks) and light fragments (longer tracks). This indicates that spectrometric characteristics of the nuclear emulsion are not substantially influenced by the energy absorption of fission fragments originating in the 0.098 μm thick film.

Bigazzi et al. (1991) submitted UO_3 thin films with thickness between 0.0015 and 0.1 μm, attached to muscovite mica detectors, to the same neutron irradiation. The ratio of FT density in mica to U content (determined through α particle tracks in nuclear emulsions) is the same for all films. Films with a thickness of 0.0015 μm in practice consist of a single layer of UO_3, so that energy absorption can be excluded. Consequently, considering the detection efficiency of the nuclear emulsion is not influenced by energy absorption for α particles originating from these films, one can conclude that the results reported by

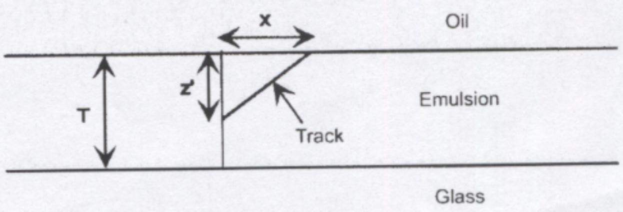

Fig. 2. Transversal section of a nuclear emulsion showing the parameters measured for track length determinations.

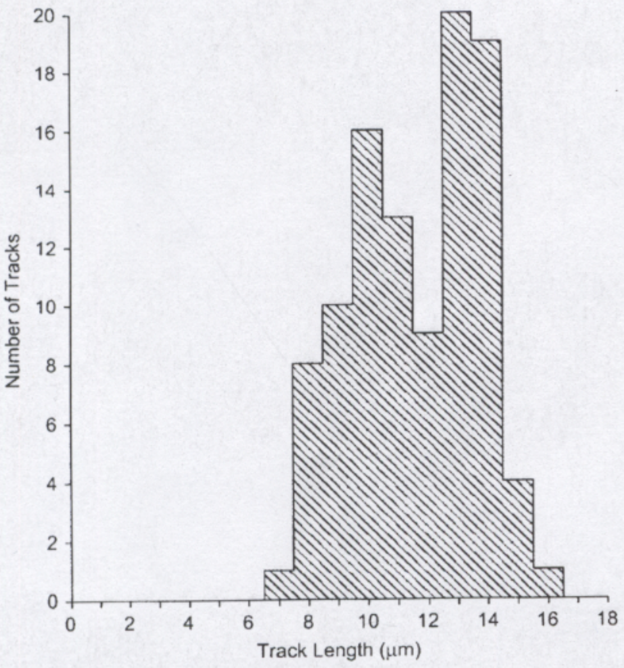

Fig. 3. Fission fragments track length distribution in a nuclear emulsion attached to a uranium 0.098 μm thick film.

these authors indicate that also the mica detection efficiency for fission fragments originating from < 0.1 μm thick films is not influenced by energy absorption.

The Th thin films used for neutron fluence determinations for FT dating are doped with ThO_2, which has a density $\sim 30\%$ higher than UO_3 (Iunes et al., 2004). However, considering that the maximum thickness of Th thin films used for FT dating was 0.07 μm, although the relationship between track length and track density of the material is not a fully linear relationship, one can reasonably extend the results obtained for U thin films to Th thin films.

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