

# U and Th thin film neutron dosimetry for fission-track dating: application to the age standard Moldavite

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## Abstract

Neutron dosimetry based on U and Th thin films was used for fission-track dating of the age standard Moldavite, the central European tektite, from the Middle Miocene deposit of Jankov (southern Bohemia, Czech Republic). Our fission-track age ( $13.98 \pm 0.58$  Ma) agrees with a recent  $^{40}\text{Ar}/^{39}\text{Ar}$  age,  $14.34 \pm 0.04$  Ma, based on several determinations on Moldavites from different sediments, including the Jankov deposit. This result indicates that the U and Th thin film neutron dosimetry represents a reliable alternative for an absolute approach in fission-track dating.

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## 1. Introduction

During the last 20 years most fission-track (FT) groups adopted the system calibration known as “zeta calibration” (Hurford and Green, 1983). The use of the zeta calibration, based upon age standards, is a way of overcoming difficulties related to uncertainties in the fundamentals (such as in the value of the  $^{238}\text{U}$  spontaneous fission decay constant or in the approach to be adopted for adequate neutron dosimetry) as well as in parameters related to some experimental procedures (such as the external detector method, Gleadow, 1981). However, the zeta calibration makes the FT method

dependent on other isotopic techniques employed for determining the independent reference age of the standards used for the determination of the zeta factor. For this reason, research on procedures devoted to enabling FT dating to be an independent system with an absolute calibration was encouraged (see for instance Hurford, 1998).

One of the crucial points for setting an absolute system calibration is neutron dosimetry. Bigazzi et al. (1999) have shown that the use of natural U and Th thin films can be an efficient alternative for accurate neutron dosimetry. In addition, it has been shown that U and Th thin films make possible to apply the FT method also when only weakly thermalized irradiation facilities are available, such as in many research nuclear reactors. The International Union of Geological Sciences (IUGS) had discouraged the use of such facilities (Hurford, 1990), because of the difficulties related

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to the determination of the contribution (to the induced track density) of the U and Th fissions induced by epithermal and fast neutrons.

Recently, neutron dosimetry based on U and Th thin films was adopted for dating Durango apatite (Iunes et al., 2002), one of the age standards recommended by the IUGS subcommission on geochronology for FT dating of apatite (Hurford, 1990). Our result,  $29.7 \pm 1.1$  Ma, was consistent with the recommended reference age,  $31.4 \pm 0.6$  Ma, average of K–Ar determinations of the Carpintero group (McDowell and Keizer, 1977).

In the present work, the neutron dosimetry based on U and Th thin films was used for dating Moldavite, the central European tektite, the unique glass recommended as the age standard for FT dating of natural glasses (Hurford, 1990).

## 2. Experimental

Neutron irradiations were performed in three nuclear reactors: IPEN/CNEN—São Paulo, Brazil (irradiation I27MD and I32MD), LENA—Pavia, Italy (irradiation P4MD) and IPEN—Lima, Peru (irradiation L19MD). The sample-set present in these irradiations was made up of thin Th films, natural U-doped glasses (Cornig glass CN1 in irradiations I32MD and P4MD, and Cornig glasses CN2 and CN5 in irradiations I27MD and L19MD, respectively) and two Moldavites (MJ1 and MJ2) from the Middle Miocene deposit of Jankov, southern Bohemia (Bouška, 1994; Balestrieri et al., 1998). Moldavites MJ1 and MJ2 were included in irradiation I32MD, but only MJ1 was included in the other irradiations. Only in case of irradiation I32MD, Moldavite samples were pre-heated for 10 h at  $560^\circ\text{C}$  in order to erase spontaneous tracks.

Ages were computed using the following equation:

$$T = \frac{1}{\lambda} \ln \left[ \frac{(\rho_S/\rho_I)\lambda R_M}{C_{238}\lambda_F} + 1 \right], \quad (1)$$

where

$$R_M = R_U + \left( \frac{N_{\text{Th}}}{N_U} \right) R_{\text{Th}} \quad (2)$$

$$R_U = C_{235}A_{235} + C_{238}A_{238}, \quad R_{\text{Th}} = A_{232}, \quad (3)$$

where  $\lambda$  is the  $^{238}\text{U}$   $\alpha$ -decay constant;  $\rho_S/\rho_I$  is the spontaneous to induced track areal density ratio;  $C_{235}$  ( $C_{238}$ ) is the  $^{235}\text{U}$  ( $^{238}\text{U}$ ) abundance in natural U;  $\lambda_F$  is the  $^{238}\text{U}$  spontaneous fission decay constant;  $N_{\text{Th}}/N_U$  is the sample Th/U ratio; and  $A_{235}$ ,  $A_{238}$  and  $A_{232}$  are the numbers of fissions per  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  target nucleus, respectively. The following values for the constants of Eq. (1) were adopted:  $\lambda = 1.55125 \times 10^{-10} \text{ a}^{-1}$ ,  $C_{238} = 0.99275$  and  $\lambda_F = 8.46 \times 10^{-17} \text{ a}^{-1}$  (Spadavecchia and Hahn, 1967). This  $\lambda_F$  “historical” value, widely adopted through the FT community by people working with an absolute approach, is in close agreement with the  $\lambda_F$  value of  $(8.5 \pm 0.1) \times 10^{-17} \text{ a}^{-1}$

recently recommended by Holden and Hoffman (2000) and is also confirmed by a new measurement which is the average of nine determinations carried out using neutron dosimetry based on natural U thin films,  $(8.37 \pm 0.17) \times 10^{-17} \text{ a}^{-1}$  (Guedes et al., 2003).

The  $R_M$  values used in the present work were computed from the determinations of the  $R_U$  and  $R_{\text{Th}}$  parameters, using in Eq. (2) a Th/U ratio of 4.41 for Jankov Moldavite (Bouška, 1994).

The  $R_U$  values were obtained through natural U-doped glasses present in the irradiations quoted above. The calibration of these glasses was carried out by irradiating them together with natural U thin films (Bigazzi et al., 1999), which played the role of primary calibrators.

The  $R_{\text{Th}}$  values were obtained through Th thin films, also present in the irradiations (Bigazzi et al., 1999).

The calibration parameters of the glasses and thorium thin films used in this work are shown in Iunes et al. (2002).

## 3. Results and discussion

Results of age determinations of Moldavites MJ1 and MJ2 from the Jankov deposit are shown in Table 1. It must be noted that all results shown in this work are quoted at  $1\sigma$ , where  $\sigma$  is the standard error of the mean. These samples showed a certain degree of reduction of the spontaneous track sizes in comparison with the induced ones ( $D_S/D_I < 1$ ). Reduced track sizes indicate that glass suffered a certain amount of partial annealing of spontaneous tracks, which are etched with reduced efficiency. Therefore, FT “raw” ages of MJ1 and MJ2 Moldavites are partially rejuvenated. In this work the plateau method introduced by Storzer and Poupeau (1973) was used for correcting thermally lowered ages. To apply this age correction technique, two groups from each sample—one for spontaneous track counting and the other for induced track counting (the irradiated one)—were heated for 24 h at  $350^\circ\text{C}$ . This heating step produced a significant reduction in track densities. The attainment of the plateau condition—an identical etching efficiency for spontaneous and induced tracks—is proved by the  $D_S/D_I$  ratio value that became close to 1.

Considering only the plateau age determinations of irradiation I32MD, the weighted mean of the  $\rho_S/\rho_I$  ratios is  $0.422 \pm 0.019$ . Using this value and the  $R_M$  value of Table 1, a mean plateau age of  $13.31 \pm 0.83$  Ma was computed using Eq. (1).

For irradiation P4MD, two thermal treatments were imposed for the plateau age determination—4 and 24 h at  $350^\circ\text{C}$ , respectively. In both cases, the plateau condition was attained ( $D_S/D_I \sim 1$ ). The weighted mean of the  $\rho_S/\rho_I$  ratios is  $0.688 \pm 0.043$ , and the corresponding mean plateau age is  $14.2 \pm 1.1$  Ma.

Considering the mean ages from irradiations I32MD and P4MD and the ages determined through irradiations L19MD and I27MD as independent determinations, the weighted

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