

Radiation Measurements 35 (2002) 339-345

**Radiation Measurements** 

www.elsevier.com/locate/radmeas

# Dating of a quaternary limestone cave by combining the SSNTD technique with paleodose measurements: application to the stalagmite and stalactite growth

M.A. Misdaq<sup>a, \*</sup>, L. Oufni<sup>a, b</sup>, H. Erramli<sup>a</sup>, L. Boudad<sup>c</sup>, L. Kabiri<sup>c</sup>

<sup>a</sup>Nuclear Physics and Techniques Laboratory, Faculty of Sciences Semlalia, University Cadi Ayyad, B.P. 2390, Marakech, Morocco <sup>b</sup>Department of Physics (LOTEA-PN), Faculty of Sciences and Techniques, University My Ismaîl, B.P. 509 Boutalamine,

Errachidia, Morocco

<sup>e</sup>Department of Geology (LFSEC), Faculty of Sciences and Techniques, University My Ismaîl, B.P. 509 Boutalamine, Errachidia, Morocco

Received 15 June 2001; received in revised form 28 December 2001; accepted 11 February 2002

## Abstract

Uranium (<sup>238</sup>U) and thorium (<sup>232</sup>Th) contents were evaluated in different stalagmite and stalactite samples belonging to a quaternary limestone cave by using a method based on determining mean critical angles of etching of the CR-39 and LR-115 II solid-state nuclear track detectors (SSNTDs). Annual absorbed  $\alpha$ -,  $\beta$ - and  $\gamma$ -dose rates were determined in the stalagmite and stalactite materials. The stalagmite and stalactite samples were dated by exploiting data obtained for the total annual absorbed dose rates and measuring the corresponding paleodoses. Results obtained were compared with those obtained by classic thermoluminescence and U/Th disequilibrium methods. The studied speleothem began from about 121 kyr and continued to about 69 kyr, coinciding almost with the last interglaciation corresponding to the fourth and fifth isotope stages. The formation rates of the stalagmite and stalactite samples were found equal to 0.88 and 0.94 cm kyr<sup>-1</sup>, respectively. © 2002 Elsevier Science Ltd. All rights reserved.

# 1. Introduction

Measuring the annual  $\alpha$ -,  $\beta$ - and  $\gamma$ -dose rates is of great interest in dating objects by means of the thermoluminescence (TL) technique (Bell, 1979; Aitken, 1985; Singhvi and Mejdahl, 1985). Ousmoï (1989) has measured  $\alpha$ - and  $\beta$ -dose rates due to the uranium and thorium series as well as <sup>40</sup>K in different archaeological samples by performing ZnS scintillation counting measurements and using Bell's method (Bell, 1979) based on the use of a standard pottery for calibration. The annual absorbed  $\gamma$ -dose rates of soil samples of an archaeological deposit have been determined by using TL dosimetry (Erramli, 1986; Misdaq et al., 1998). However, due to the absorption of the incident gamma-rays in the protective walls of the capsule enclosing the dosimeter and the TL material, results obtained from TL dosimetry should be corrected (Fahde et al., 1996; Fahde, 1997).

Uranium and thorium contents have been evaluated in various material samples using alpha spectroscopy (Jurado Vargas et al., 1997), radiochemical separations (Herranz et al., 1997) and extraction chromatography with anion exchange and alpha spectroscopy (Goldstein et al., 1997).

Solid-state nuclear track detectors (SSNTDs) have been intensively used in various fields during the last three decades (Fleischer et al., 1975; Durrani and Bull, 1987; Khan and Quereshi, 1994). The SSNTD technique was utilized for uranium and thorium contents determination in different material samples (Samad Baig et al., 1983; Jojo et al., 1994; Misdaq and Satif, 1995; Misdaq et al., 1999; Misdaq et al., 2000).

In the present work, we determine ages of stalagmite and stalactite materials in a quaternary cave by combining the

<sup>\*</sup> Corresponding author. Tel.: +212-4-4434649; fax: +212-4-436769.

E-mail address: misdaq@ucam.ac.ma (M.A. Misdaq).



Fig. 1. Map showing the localization of the studied cave in the Aoufous region (Errachidia, Morocco).

SSNTD technique with paleodose measurements. The relevant stopping powers of the materials studied as well as ranges of the  $\alpha$ - and  $\beta$ -particles emitted by the uranium and thorium series and <sup>40</sup>K isotope in the considered stalagmite and stalactite samples were calculated by using a TRIM programme (Biersack and Ziegler, 1992).

#### 2. Geological setting and samples

The studied cave is located in the Aoufous area, Province of Errachidia (High-Atlas, Morocco, Fig. 1). This karstic cavity, opened in Turonian continental limestone. The cave is a vast cavity of some 48 m long and 2–12 m wide, and has a ceiling height of 8 m.

The cave developed in limestone bedrock due to the dissolution of calcium carbonate by carbonic acid. High concentrations of  $CO_2$  are frequently found in the soil atmosphere due to biological processes and partially dissolved in percolation water to yield carbonic acid thus greatly increasing the rate of dissolution of the underlying limestone. When seepage waters subsequently come in contact with the cave atmosphere, where  $CO_2$  concentrations are usually much lower than in soils, they quickly become highly supersaturated due to outgassing of  $CO_2$  giving rise to deposition of calcite (Buhmann and Dreybrodt, 1985). The depositional process can be characterised by the following equation:

$$\operatorname{Ca}^{2+} + 2\operatorname{HCO}_3^- \to \operatorname{CO}_2 + \operatorname{H}_2\operatorname{O} + \operatorname{CaCO}_3. \tag{1}$$

Precipitation of calcium carbonate from thin water films is controlled by four main mechanisms that act independently but any one of these can determine the overall rate:

- Kinetics of precipitation at the phase boundary between the CaCO<sub>3</sub>-H<sub>2</sub>O-CO<sub>2</sub> system and the limestone.
- (2) Kinetics of conversion of carbonic acid (H<sub>2</sub>CO<sub>3</sub>) into CO<sub>2</sub>.
- (3) Mass transport of dissolved ions by diffusion to phase boundaries.
- (4) Rate of outgassing of CO<sub>2</sub> from solution into the cave atmosphere.

The samples used for the present study are stalagmite and stalactite collected from the studied cave. They have been divided into several parts (Fig. 2). The speleothems studied in this work have been analysed by X-ray diffractometry. Calcite mineralogy is present.

# 3. Method of study

Disc-shaped Pershore Mouldings CR-39 (500  $\mu$ m thickness) and Kodak LR-115 type II (12  $\mu$ m cellulose nitrate on 100  $\mu$ m polyester base) SSNTD films of 4 cm



Fig. 2. Different edges of the stalagmite (a) and stalactite (b) samples.

diameter were separately placed on each geological material sample in a hermetically sealed cylindrical plastic container for 1 and half months. During this time  $\alpha$ -particles emitted by uranium (<sup>238</sup>U), thorium (<sup>232</sup>Th) and their corresponding decay products bombarded the SSNTD films. After the irradiation, the bombarded films were developed in an NaOH solution (2.5 M at 60°C during 120 min for LR-115 films and 6.25 M at 70°C during 7 h for the CR-39 films). After this chemical treatment, the CR-39 and LR-115 α-particles track densities were determined by means of an ordinary microscope with magnification 40×. Backgrounds of the CR-39 and LR-115 type II SSNTDs have been evaluated by placing these films in the same empty hermetically sealed plastic container used for analysing geological samples for 1 and half months and by counting the resulting track density rates. This operation was repeated 10 times: track density rates registered on the CR-39 and LR-115 type II detectors were found similar within the statistical uncertainty interval.

By calculating first the mean critical angles of etching of the CR-39 ( $\langle \theta_c^{CR} \rangle$  and LR-115 type II ( $\langle \theta_c^{LR} \rangle$ ) SSNTDs, and secondly by measuring the  $\alpha$ -particle track density rates  $\rho_G^{CR}$  (in tracks cm<sup>-2</sup> s<sup>-1</sup>) and  $\rho_G^{LR}$  (in tracks cm<sup>-2</sup> s<sup>-1</sup>), after subtracting the corresponding backgrounds, one can evaluate the thorium-to-uranium ratios and consequently the thorium [*C*(Th)] and uranium [*C*(U)] contents in the studied geological samples by using a method described in detail by Misdaq et al. (1999). Indeed, we have

 $\frac{C(\mathrm{Th})}{C(\mathrm{U})}$ 

$$= \frac{A_{\rm U}}{A_{\rm Th}} \frac{\sum_{i=1}^{8} k_i (\sin\langle\theta_{c_i}^{\rm CR}\rangle)^2 R_i - 8(\sin\langle\theta_c^{\rm LR}\rangle)^2 \Delta R(\rho_{\rm G}^{\rm CR}/\rho_{\rm G}^{\rm LR})}{6(\sin\langle\theta_c^{\rm LR}\rangle)^2 \Delta R(\rho_{\rm G}^{\rm CR}/\rho_{\rm G}^{\rm LR}) - \sum_{i=1}^{7} k_i (\sin\langle\theta_{c_i}^{\rm CR}\rangle)^2 R_i}$$
(2)

$$=\frac{\rho_{\rm G}^{\rm LR}}{2d_{\rm s}\Delta R[4A_{\rm U}(\sin\langle\theta_{\rm c}^{\rm LR}\rangle)^2+3A_{\rm Th}C({\rm Th})/C({\rm U})(\sin\langle\theta_{\rm c}^{\rm LR}\rangle)^2]},$$
(3)

where  $d_s$  is the density of the material sample (g cm<sup>-3</sup>),  $A_U$  (Bq g<sup>-1</sup>) is the specific activity in 1 ppm (10<sup>-6</sup> g g<sup>-1</sup>) of <sup>238</sup>U,  $A_{Th}$  (Bq g<sup>-1</sup>) is the specific activity in 1 ppm of <sup>232</sup>Th,  $R_i$  is the range of an  $\alpha$ -particle of initial energy  $E_{i,\alpha}$ and index *i* in the material sample,  $\Delta R = R_{max} - R_{min}$  where  $R_{min}$  and  $R_{max}$  are the  $\alpha$ -particle ranges in the sample which correspond to the lower ( $E_{min} = 1.6$  MeV) and upper ( $E_{max} =$ 4.7 MeV) ends of the energy window ( $\Delta E = E_{max} - E_{min}$ ) and  $k_i$  is the branching ratio.

Based on the contents of uranium, thorium and potassium in the studied samples, the annual dose rates are evaluated by using a method developed by Misdaq et al. (1997). Indeed, the total  $\alpha$ -dose rate ( $\mu$ Gy yr<sup>-1</sup>) in the considered sample, due to uranium and thorium series, is given by

$$D_{\rm G}^{\alpha} = C(\mathbf{U})D_{\rm sp}^{\alpha}(\mathbf{U}) + C(\mathrm{Th})D_{\rm sp}^{\alpha}(\mathrm{Th}),\tag{4}$$

where  $D_{sp}^{\alpha}(U)$  (µGy yr<sup>-1</sup>) and  $D_{sp}^{\alpha}(Th)$  (µGy yr<sup>-1</sup>) are the specific  $\alpha$ -dose rates deposited by 1 ppm of <sup>238</sup>U and 1 ppm of <sup>232</sup>Th inside a material sample.

Let us consider an  $\alpha$ -particle of index *i*, initial energy  $E_{i,\alpha}$ and range  $R_{i,\alpha}$  emitted by the uranium 238 series inside a material sample. We assume that each  $\alpha$ -particle is born, inside the considered sample, in a cubic box of dimension  $R_{i,\alpha}$ (volume  $V_{i,\alpha} = R_{i,\alpha}^3$ ). The dose deposited by the considered  $\alpha$ -particle inside the cubic box is

$$d_i(\mathbf{U}) = \frac{E_{i,\alpha}}{m},\tag{5}$$

where  $m = d_s V_{i,\alpha}$  is the mass of the cubic box and  $d_s$  is the density (g cm<sup>-3</sup>) of the material sample.

Indeed, we have

$$E_{i,\alpha} = R_{i,\alpha} S_{i,\alpha},\tag{6}$$

where  $S_{i,\alpha}$  (MeV cm<sup>-1</sup>) is the stopping power of the material for the emitted  $\alpha$ -particle.

The corresponding  $\alpha$ -dose rate is given by

$$D_i^{\alpha}(\mathbf{U}) = \frac{d_i(\mathbf{U})}{t_{\mathbf{e}}}.$$
(7)

Hence,

$$D_i^{\alpha}(\mathbf{U}) = \frac{R_{i,\alpha} S_{i,\alpha}}{d_{\rm s} V_{i,\alpha} t_{\rm e}},\tag{8}$$

where  $t_e$  is the exposure time of the considered sample.

For a number N of  $\alpha$ -particles of energy  $E_{i,\alpha}$ , Eq. (8) becomes

$$D_{\text{tot}}^{\alpha}(\mathbf{U}) = \frac{NR_{i,\alpha}S_{i,\alpha}}{d_{s}V_{i,\alpha}t_{e}}.$$
(9)

The last equation can be written as

$$D_{\text{tot}}^{\alpha}(\mathbf{U}) = \frac{A_{\text{c}}(\mathbf{U})R_{i,\alpha}S_{i,\alpha}V_{i,\alpha}}{d_{s}V_{i,\alpha}},$$
(10)

where  $A_c(U) = N/V_{i,\alpha}t_e$  expressed in Bq cm<sup>-3</sup> is the  $\alpha$  activity per unit volume corresponding to 1 ppm (10<sup>-6</sup> g g<sup>-1</sup>) of <sup>238</sup>U.

For all  $\alpha$ -particles emitted by the nuclei of the uranium series, the specific  $\alpha$ -dose rate is given by

$$D_{\rm sp}^{\alpha}(\mathbf{U}) = K \frac{A_{\rm c}(\mathbf{U})}{d_{\rm s} \sum_{i=1}^{8} V_{i,\alpha}} \sum_{i=1}^{8} k_i S_{i,\alpha} R_{i,\alpha} V_{i,\alpha}, \tag{11}$$

where  $k_i$  is the branching ratio and K is a conversion factor.

Similarly, the specific  $\alpha$ -dose rate due to  $\alpha$ -particles emitted by the nuclei of the thorium series is given by

$$D_{\rm sp}^{\alpha}({\rm Th}) = K \frac{A_{\rm c}({\rm Th})}{d_{\rm s} \sum_{i=1}^{7} V_{i,\alpha}} \sum_{i=1}^{i} k_i S_{i,\alpha} R_{i,\alpha} V_{i,\alpha}, \qquad (12)$$

where  $A_c(U)$  and  $A_c(Th)$  (Bq cm<sup>-3</sup>) are, respectively, the activities per unit volume corresponding to 1 ppm of <sup>238</sup>U and 1 ppm of <sup>232</sup>Th,  $V_{i,\alpha}$  is the volume of a cubic box of dimension  $R_{i,\alpha}$  ( $R_{i,\alpha}$  is the range of an  $\alpha$ -particle of initial energy  $E_{i,\alpha}$ ),  $S_{i,\alpha}$  (MeV cm<sup>-1</sup>) is the geological material stopping power for an  $\alpha$ -particle of index *i*,  $k_i$  is the branching ratio,  $d_s$  (g cm<sup>-3</sup>) is the sample density and *K* is a conversion factor.

The total  $\beta$ -dose rate ( $\mu$ Gy yr<sup>-1</sup>) in the considered sample, due to the nuclei of uranium and thorium series as well as <sup>40</sup>K isotope, is given by (Misdaq et al., 1997)

$$D_{G}^{\beta} = C(U)D_{sp}^{\beta}(U) + C(Th)D_{sp}^{\beta}(Th) + [\%K_{2}O]D_{sp}^{\beta}(^{40}K), \qquad (13)$$

where [%K<sub>2</sub>O] is the concentration of K<sub>2</sub>O (in %),  $D_{sp}^{\beta}(U)$  ( $\mu$ Gy yr<sup>-1</sup>) and  $D_{sp}^{\beta}(Th)$  ( $\mu$ Gy yr<sup>-1</sup>) are the specific  $\beta$ -dose rates deposited by 1 ppm of <sup>238</sup>U and 1 ppm of <sup>232</sup>Th inside the material sample.

Similarly, the  $D_{sp}^{\beta}(U)$  and  $\overline{D}_{sp}^{\beta}(Th)$  specific  $\beta$ -dose rates are given by

$$D_{\rm sp}^{\beta}(\mathbf{U}) = K \frac{A_{\rm c}(\mathbf{U})}{d_{\rm s} \sum_{i=1}^{8} V_{i,\beta}} \sum_{i=1}^{8} k_i S_{i,\beta} R_{i,\beta} V_{i,\beta}$$
(14)

and

$$D_{\rm sp}^{\beta}({\rm Th}) = K \frac{A_{\rm c}({\rm Th})}{d_{\rm s} \sum_{i=1}^{5} V_{i,\beta}} \sum_{i=1}^{5} k_i S_{i,\beta} R_{i,\beta} V_{i,\beta}, \qquad (15)$$

where  $V_{i,\beta}$  is the volume of a cubic box of dimension  $R_{i,\beta}$  ( $R_{i,\beta}$  is the range of a  $\beta$ -particle of index *i* and energy

 $E_{i,\beta}$ ),  $S_{i,\beta}$  (MeV cm<sup>-1</sup>) is the sample material stopping power for a  $\beta$ -particle and  $k_i$  is the branching ratio.

Knowing the concentration of  $K_2O$  (in %), one can find the  $\beta$ -dose rates due to  ${}^{40}K$  in the studied materials. Indeed, we have

$$D_{\rm sp}^{\beta}({}^{40}{\rm K}) = K \, \frac{A_{\rm c}(1\% \text{ of } {\rm K}_{2}{\rm O})}{d_{\rm s}} \, k.\bar{S}.\bar{R},\tag{16}$$

where  $A_c(1\% \text{ of } K_2\text{O})$  is the  ${}^{40}\text{K}$   $\beta$  activity (Bq cm<sup>-3</sup>) in 1% of K<sub>2</sub>O, k = 89.33% is the branching ratio,  $\bar{S}$  (MeV cm<sup>-1</sup>) is the average stopping power of the stalagmite and stalactite samples for the  $\beta$ -particles emitted by  ${}^{40}\text{K}$ ,  $\bar{R}$  is the average range of the  $\beta$ -particles emitted by  ${}^{40}\text{K}$  ( $\bar{E}_{\beta} = 1.33$  MeV) inside the sample.

The total annual absorbed  $\gamma$ -dose rate (in  $\mu$ Gy yr<sup>-1</sup>) in the considered material, due to the uranium and thorium series as well as <sup>40</sup>K isotope, is given by (Misdaq et al., 1998)

$$D_{G}^{\gamma} = C(U)D_{sp}^{\gamma}(U) + C(Th)D_{sp}^{\gamma}(Th) + [\%K_{2}O]D_{sp}^{\gamma}(^{40}K),$$
(17)

where  $D_{sp}^{\gamma}(U)$ ,  $D_{sp}^{\gamma}(Th)$  and  $D_{sp}^{\gamma}({}^{40}K)$  are the specific  $\gamma$ -dose rates deposited by 1 ppm of  ${}^{238}U$ , 1 ppm of  ${}^{232}Th$  and 1% of K<sub>2</sub>O ( ${}^{40}K$ ) inside a geological sample.

Let us consider a gamma photon of index *i* and energy  $E_{i,\gamma}$  emitted by the nuclei of the uranium series inside a material sample. The dose deposited by the considered  $\gamma$ -ray inside the material sample of volume V (cm<sup>3</sup>) and density  $d_s$  (g cm<sup>-3</sup>) is given by

$$d_i(\mathbf{U}) = \frac{E_{\mathrm{d}}^i}{d_{\mathrm{s}}V},\tag{18}$$

where  $E_{d}^{i} = \sum_{j=1}^{N} E_{i,\gamma}(1 - e^{-\mu l_{j}})$  is the energy deposited by the  $\gamma$  photon of index *i* and energy  $E_{i,\gamma}$  inside the considered sample,  $\mu$  is the total attenuation coefficient of the  $\gamma$  photons in the material sample,  $l_{j}$  is the path length of the considered  $\gamma$  photon in the sample.

The corresponding dose rate is

$$D_{i}(\mathbf{U}) = \frac{\sum_{j=1}^{N} E_{i,\gamma}(1 - e^{-\mu l_{j}})}{d_{s} V t_{e}},$$
(19)

where  $t_e$  is the exposure time of the considered sample.

For a large number N of  $\gamma$  photons of energy  $E_{i,\gamma}$  Eq. (19) can be written as

$$D_i^{\text{tot}}(\mathbf{U}) = \frac{A_c(\mathbf{U})}{d_s} E_{i,\gamma}(\text{SAC})_{i,\gamma},$$
(20)

where  $A_{\rm c}({\rm U})$  (Bq cm<sup>-3</sup>) is the activity perunit volume corresponding to 1 ppm of <sup>238</sup>U and (SAC)<sub>*i*, $\gamma = \sum_{j=1}^{N} (1 - e^{-\mu l_j})/N$  is the self-absorption coefficient of the  $\gamma$ -ray inside the material sample which is calculated by using a method developed by Misdaq et al. (1998).</sub>

The specific  $\gamma$ -dose rate  $D_{sp}^{\gamma}U$  ( $\mu$ Gy yr<sup>-1</sup>) deposited by 1 ppm of <sup>238</sup>U inside the material sample is given by

$$D_{\rm sp}^{\gamma}(\mathbf{U}) = K \frac{A_{\rm c}(\mathbf{U})}{d_{\rm s}} \sum_{i=1}^{10} I_{i,\gamma} E_{i,\gamma} (\rm SAC)_{i,\gamma}, \qquad (21)$$

where  $I_{i,\gamma}$  is the intensity of a  $\gamma$ -photon of index *i* and energy

 $E_{i,\gamma}$ . Similarly, the specific  $\gamma$ -dose rates deposited by 1 ppm of <sup>232</sup>Th  $D_{sp}^{\gamma}$ (Th) ( $\mu$ Gy yr<sup>-1</sup>) and 1% of K<sub>2</sub>O  $({}^{40}\text{K}) D_{sb}^{\gamma}({}^{40}\text{K}) (\mu\text{Gy yr}^{-1})$  are given by

$$D_{\rm sp}^{\gamma}(\rm Th) = K \, \frac{A_{\rm c}(\rm Th)}{d_{\rm s}} \sum_{i=1}^{17} I_{i,\gamma} E_{i,\gamma}(\rm SAC)_{i,\gamma} \tag{22}$$

and

$$D_{\rm sp}^{\gamma}({}^{40}{\rm K}) = K \, \frac{A_{\rm c}({}^{40}{\rm K})}{d_{\rm s}} I_{\gamma} E_{\gamma}({\rm SAC})_{\gamma}, \qquad (23)$$

where  $A_c(Th)$  and  $A_c(^{40}K)$  (Bq cm<sup>-3</sup>) are the activities corresponding to 1 ppm of <sup>232</sup>Th and 1% of K<sub>2</sub>O, respectively, and  $(SAC)_{\gamma}$  is the self-absorption coefficient of the  $\gamma$ -ray emitted by <sup>40</sup>K which has an energy  $E_{\gamma} = 1.461$  MeV and intensity  $I_{\gamma} = 10\%$ .

According to TL dating technique (Bell, 1979; Singhvi and Mejdahl, 1985), the age t (in year) of a sample is given by

$$t = \frac{\text{Paleodose}}{0.1D_{\text{G}}^{\alpha} + D_{\text{G}}^{\beta} + D_{\text{G}}^{\gamma} + D_{\cos}}.$$
(24)

In underground sites, the amount of cosmic radiation is low (in our case, the cosmic dose rate  $(D_{cos})$  is negligible).

By measuring the paleodose and evaluating the  $D_G^{\alpha}$ ,  $D_G^{\beta}$ and  $D_G^{\gamma}$  dose rates one can determine the age of a geological sample using Eq. (24).

#### 4. Results and discussion

The studied stalagmite (46 cm length) and stalactite (32 cm length) samples were collected from a limestone cave situated in the [1050-1150 m] altitude interval in the Aoufous region (High-Atlas, Morocco) (Fig. 1). Their uranium C(U) and thorium C(Th) contents are shown in Table 1. From the statistical error on track counting one can determine the error on track density rate and then evaluate the relative uncertainty of the uranium and thorium concentrations determination which is of 6%. Results obtained by this method are in good agreement with those obtained by isotope dilution mass spectrometry (IDMS) (Table 1). We notice from results shown in Table 1 that the C(U) and C(Th) contents decrease from the base to the top of the studied stalagmite and stalactite materials. This is due to the fact that the material's diameters decrease from the base to the top (see Fig. 2) favouring the concentration of  $^{238}$ U and <sup>232</sup>Th in the stalagmite and stalactite bases. We also notice that the uranium content of the studied samples does not exceed 1.12 ppm and C(Th)/C(U) ratio is lower than 30%.

The annual  $\alpha$ -,  $\beta$ - and  $\gamma$ -dose rates in the studied stalagmite and stalactite samples have been evaluated by using the SSNTD technique and Bell's method (Bell, 1979). The data Table 1

Data obtained for the uranium and thorium contents and %<sup>40</sup>K in the studied stalagmite and stalactite samples

Samples	SSNTD		IDMS $C(Th)$	% <sup>40</sup> K
	<i>C</i> (U)	C(Th)	(ppm)	
	(ppm)	(ppm)		
Stalagmite				
Sg <sub>1</sub>	$1.140\pm0.048$	0.390	$1.16\pm0.056$	0.09
$Sg_2$	$1.120\pm0.036$	0.260	$1.11\pm0.050$	0.080
Sg <sub>3</sub>	$1.080\pm0.045$	0.160	$1.06\pm0.052$	0.090
$Sg_4$	$0.890\pm0.026$	0.130		0.100
$Sg_{11}$	$0.720\pm0.025$	0.115	$0.71\pm0.02$	0.110
Sg <sub>13</sub>	$0.320\pm0.009$	0.085		0.095
Stalactite				
$Sc_1$	$0.630\pm0.021$	0.173	$0.620\pm0.018$	0.102
Sc <sub>2</sub>	$0.600\pm0.018$	0.130	$0.580\pm0.014$	0.086
Sc <sub>8</sub>	$0.180\pm0.005$	0.033		0.095

obtained are shown in Table 2. We notice that results obtained by the two methods are in good agreement with each other. We also notice that the annual dose rates  $(D_G^{\alpha}, D_G^{\beta})$ and  $D_G^{\gamma}$ ) decrease from the base to the top for the stalagmite and stalactite samples according to their uranium and thorium contents.

The formation ages of the studied stalagmite samples were determined by using this method and the thermoluminescence and U/Th dating methods. The data obtained are shown in Table 3. A good agreement has been found between the results obtained by the three methods (Table 3).

The thermoluminescence ages of the quaternary stalagmite ranges from 69 to 121 kyr. These ages are in good stratigraphical succession according to the growth of the studied stalagmite (Fig. 2). From these results, one can say that the stalagmite formation occurred in a time interval of 52 kyr, giving a stalagmite growth rate of 0.88 cm kyr<sup>-1</sup>. Three of the stalactite samples belonging to the base and the top have been dated by using our method and the U/Th dating method (Table 3). The data obtained by the two methods are in good agreement with each other (Table 3). This stalactite material was formed in a time interval of 34 kyr. Consequently, its formation rate is of  $0.94 \text{ cm kyr}^{-1}$ . Uncertainty of the stalagmite and stalactite ages determination by using our method is of 7%, whereas those corresponding to the thermoluminescence and U/Th dating methods are of 5% and 6%, respectively.

On the basis of these results, the chronostratigraphy of the studied stalagmite and stalactite materials enables us to say that the formation of these, covered the last Pleistocene climatic cycles, and reconstructed the environmental changes which have occurred during the chronological isotopic stages 5 and 4.

Samples	This method ( $\mu$ Gy yr <sup>-1</sup> )			Bell's method ( $\mu$ Gy yr <sup>-1</sup> )		
	$D_{ m G}^{lpha}$	$D_{ m G}^{eta}$	$D_{ m G}^\gamma$	$D_{ m G}^{lpha}$	$D_{ m G}^{eta}$	$D_{ m G}^\gamma$
Stalagmite						
Sg <sub>1</sub>	$3560\pm260$	$250.0\pm19.5$	$180.0\pm12.7$	$3660\pm200$	$235.6 \pm 11.2$	$166.0\pm7.8$
Sg <sub>2</sub>	$3240\pm200$	$225.4 \pm 15.1$	$164.5 \pm 9.9$	$3200\pm150$	$220.0\pm8.0$	$160.0\pm8.2$
Sg <sub>3</sub>	$2800 \pm 170$	$213.2\pm13.8$	$138.4 \pm 9.4$	$2760\pm130$	$210.0\pm8.8$	$137.5\pm6.5$
Sg <sub>4</sub>	$2530\pm140$	$210.0\pm14.6$	$125.0\pm8.4$	$2580 \pm 110$	$210.0\pm8.3$	$130.0\pm4.7$
Sg <sub>11</sub>	$1520\pm110$	$160.0\pm9.7$	$90.5\pm5.4$	$1450\pm80$	$150.0\pm8.4$	$85.0\pm3.6$
Sg <sub>13</sub>	$980\pm70$	$102.1\pm 6.2$	$56.8\pm3.4$	$950\pm40$	$114.2\pm4.5$	$60.3\pm2.8$
Stalactite						
Sc <sub>1</sub>	$2190\pm120$	$168 \pm 11$	$97.6 \pm 5.3$			
Sc <sub>2</sub>	$1770\pm115$	$153 \pm 9$	$102.6\pm7.0$			
Sc <sub>8</sub>	$510\pm40$	$110\pm7$	$48.6\pm3.0$			

Table 2 Data obtained for  $\alpha$ -,  $\beta$ -, and  $\gamma$ -dose rates in the studied stalagmite and stalactite samples

Table 3 Padeodose, annual total dose rates  $(0.1D_G^{\alpha} + D_G^{\beta} + D_G^{\gamma})$  and ages of the stalagmite and stalactite material samples

Samples	Total dose rates ( $\mu$ Gy yr <sup>-1</sup> )		Paleodose (GY)	Age (kyr)		
	This method	TL method	(01)	This method	TL method	U/Th method
Stalagmite						
Sg <sub>1</sub>	$786.0\pm27.4$	$767.6\pm24.2$	$92.93 \pm 4.86$	$118.23\pm11.23$	$121.06\pm10$	$121 \pm 7$
Sg <sub>2</sub>	$713.9\pm25.8$	$700.0 \pm 18.8$	$81.20\pm3.95$	$113.74\pm8.23$	$116.00\pm7$	$119 \pm 8$
Sg <sub>3</sub>	$631.6 \pm 18.3$	$623.5\pm16.9$	$69.30\pm3.30$	$109.72\pm7.58$	$110.00\pm 6$	$113 \pm 6$
Sg <sub>4</sub>	$588.0\pm20.3$	$598.0 \pm 14.5$	$62.40\pm3.13$	$106.12\pm 6.66$	$104.00\pm 6$	$108 \pm 4$
Sg <sub>11</sub>	$402.5\pm19.8$	$380.0 \pm 12.1$	$30.78 \pm 1.46$	$76.47 \pm 6.33$	$81.00\pm7$	$84\pm3$
Sg <sub>13</sub>	$256.9\pm9.0$	$269.5\pm6.6$	$18.59\pm0.75$	$72.36 \pm 4.98$	$69.00\pm4$	$71\pm3$
Stalactite						
Sc <sub>1</sub>	$484.6\pm37.3$	$495.6\pm26.8$	$47.80 \pm 2.02$	$98.63 \pm 5.20$		$96 \pm 4$
Sc <sub>2</sub>	$432.6\pm29.8$	$443\pm29$	$41.00\pm1.51$	$94.77 \pm 4.12$		$92\pm3$
Sc <sub>8</sub>	$192.1\pm10.6$	$204\pm14$	$12.60\pm0.52$	$60.25\pm2.70$		$62\pm3$

### 5. Conclusion

It has been shown by this study that by combining the solid-state nuclear track detectors (SSNTDs) technique with paleodose measurements one can evaluate the formation ages of stalagmite and stalactite material samples belonging to a limestone quaternary cave. A good agreement has been found between the SSNTD method and the thermolumines-cence and U/Th dating methods. The growth rates of the stalagmite and stalactite materials studied have been found similar within the uncertainty interval. The SSNTD technique used has the advantage of being simple, inexpensive, accurate, non-destructive and does not need the use of any standard for its calibration and is a good tool for studying climatological changes of the speleothems (e.g. stalagmite and stalactite).

#### References

- Aitken, M.J., 1985. Thermoluminescence Dating. Academic Press, London.
- Bell, W.T., 1979. Thermoluminescence dating: revised dose rate data. Archaeometry 21, 243–245.
- Biersack, J.P., Ziegler, J.F., 1992. TRIM, Version 92.
- Buhmann, D., Dreybrodt, W., 1985. The kinetics of calcite dissolution and precipitation in geologically relevant situations of karst areas: 1 open system. Chem. Geol. 48, 189–211.
- Durrani, S.A., Bull, R.K., 1987. Solid State Nuclear Track Detection: Principles, Methods and Applications. Pergamon Press, Oxford.
- Erramli, H., 1986. Thesis. University Blaise Pascal of Clermont II. France.
- Fahde, K., 1997. Thesis. University Cadi Ayyad of Marrakech. Morocco.

- Fahde, K., Erramli, H., Misdaq, M.A., 1996. Study of the influence of correction parameters on gamma doses in different soils. Third Topical Meeting on Industrial Radiation and Radioisotope Measurements and Applications. NC, USA.
- Fleischer, R.I., Price, R.B., Walker, R.M., 1975. Nuclear Tracks in Solids: Principles and Applications. University of California Press, Berkeley, CA.
- Goldstein, S.J., Rodriguez, J.M., Lujan, N., 1997. Measurement and application of uranium isotopes for human and environmental monitoring. Health Phys. 72, 10–18.
- Herranz, M., Abelairas, A., Legarda, F., 1997. Uranium contents and associated effective doses in drinking water from Biscay (Spain). Appl. Radiat. Isot. 48, 857–861.
- Jojo, P.J., Rawat, A., Rajendra Prasad, 1994. Ehancement of trace uranium in fly ash. Nucl. Geophys. 8, 55–59.
- Jurado Vargas, M., Vera Tomé, F., Martin Sánchez, A., Crespo Vásquez, M.T., Gascón Murillo, J.L., 1997. Distribution of uranium and thorium in sediment and plants from a granitic fluvial area. Appl. Radiat. Isot. 48, 1137–1141.
- Khan, H.A., Quereshi, A.A., 1994. Solid state nuclear track detection: a useful geological/geophysical tool. Nucl. Geophys. 8, 1–37.
- Misdaq, M.A., Satif, C., 1995. A new method adapted to the experimental conditions for determining thorium and uranium contents in geological samples. J. Radioanal. Nucl. Chem. 198, 179–189.

- Misdaq, M.A., Fahde, K., Erramli, H., Mikdad, A., 1997. A new method for evaluating annual alpha and beta dose rates in different ceramic samples by using solid state nuclear track detectors. Radiat. Phys. Chem. 50, 293–297.
- Misdaq, M.A., Fahde, K., Erramli, H., Mikdad, A., Rzama, A., Yousfi Charif, M.L., 1998. A new method for evaluating annual absorbed gamma dose rates in an archaeological site by combining the SSNTD technique with Monte Carlo simulations. Radiat. Phys. Chem. 53, 377–384.
- Misdaq, M.A., Bakhchi, A., Ktata, A., Merzouki, A., Youbi, N., 1999. Determination of uranium and thorium contents inside different materials using track detectors and mean critical angles. Appl. Radiat. Isot. 51, 209–215.
- Misdaq, M.A., Khajmi, H., Aitnouh, F., Berrazzouk, S., Bourzik, W., 2000. A new method for evaluating uranium and thorium contents in different natural material samples by calculating the CR-39 and LR-115 type II SSNTD detection efficiencies for the emitted α-particles. Nucl. Instrum. and Methods B 171, 350–359.
- Ousmoï, M., 1989. Thesis. University Blaise Pascal of Clermont II. France.
- Samad Baig, M.A., Sit Mahmoud Sheikh, A., Hameed, A.K., Aslam Sial, A.S., Khalid, J., 1983. Use of alpha sensitive plastic films (ASPF) method for uranium exploration in Pakistan. Nucleus 20, 61–64.
- Singhvi, A.K., Mejdahl, V., 1985. Thermoluminescence dating of sediments. Nucl. Tracks Radiat. Meas. 10, 137–161.