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Determination of thorium and uranium contents in soil samples using SSNTD's passive method

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Abstract. Thorium-to-uranium ratios have been determined in different soil samples using CR-39 and LR-115-II solid-state nuclear track detectors (SSNTDs). A calibration method based on determination of SSNTD registration sensitivity ratio for α-particles of thorium and uranium series has been developed. Thorium and uranium contents of the standard soil samples have been determined and compared with its known values. There is a good agreement between the results of this method and the values of standard samples. The method is simple, inexpensive, non-destructive and has a wide range of applications in environment, building materials and petroleum fields.

Keywords. Thorium and uranium contents; CR-39; LR-115-II; soil samples.

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1. Introduction

Solid-state nuclear track detection technique is widely used as an effective tool to measure α-tracks activity [1], where SSNTDs have been used in geology [2–6] and environment [7,8]. Both thorium and uranium contents in various natural materials have been determined using SSNTD's active techniques [9]. Several authors [10–12] reported a method for analyzing uranium and thorium contents based on the measurement of α-tracks by α-autoradiography.

Thorium in natural samples is usually more abundant than uranium by a factor of about 4. It is necessary to know the ratio Th/U in order to perform a quantitative analysis of uranium and thorium in natural samples.

The present work investigates and demonstrates a method based on the use of SSNTDs to determine thorium and uranium and their rate in thorferous and uraniferous soil samples. Calibration curve of known uranium and thorium contents was established and utilized. Thorium-to-uranium ratio is determined by registration

Table 1. Prepared sample composites of PVC, RGU-1, RGTh-1, uranium and thorium concentrations in ppm and activities of both uranium and thorium.

Sample number	Quantity of the dilute (PVC)	RGU-1 (g)	RGTh-1 (g)	Uranium content (μg/g)	Thorium content (μg/g)	Thorium to uranium	Uranium activity (Bq/g)	Thorium activity (Bq/g)
1	10	2	1	61.51	61.50	1	0.757	0.252
2	10	2	2	57.14	114.30	2	0.703	0.469
3	10	2	3	53.33	160.00	3	0.656	0.656
4	10	2	4	50.00	200.00	4	0.615	0.820
5	10	2	5	47.05	235.30	5	0.579	0.965
6	10	2	6	44.44	266.66	6	0.547	1.093
7	10	2	7	42.10	294.73	7	0.518	1.208
8	10	2	8	40.00	320.00	8	0.492	1.312
9	10	2	9	38.09	342.85	9	0.469	1.406
10	10	2	10	36.36	363.63	10	0.447	1.491

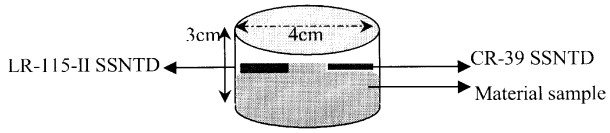

Figure 1. Arrangement of CR-39 and LR-115-II nuclear track detector films with the material sample inside the container.

sensitivities and track densities are detected by CR-39 and LR-115-II nuclear track detectors.

2. Experimental

Ten samples of different concentrations of uranium and thorium were prepared, where the thorium-to-uranium ratio rated throughout these samples. The dilute which is used to carry uranium and thorium is PVC powder approximately 2 μm diameter while the uranium and thorium samples that are mixed in PVC are RGU-1 and RGTh-1 respectively which are supplied by the IAEA. Table 1 shows the specification of these samples. Each sample together with three standard soil samples are placed in a cylindrical plastic container of 3 cm depth and 4 cm diameter. Two different nuclear track detectors (CR-39 and LR-115-II) are located in close contact over the material sample and hermitically sealed as shown in figure 1.

During a period of one month the α-particles emitted from uranium and thorium series will reach and register in the exposed solid-state nuclear track detectors. After this period the SSNTDs are developed in NaOH solution (2.5 N at 60°C during 120 min for LR-115-II film and 6.25 N at 70°C during 7 h for CR-39) and after this chemical treatment the CR-39 and LR-115-II track densities are detected by ordinary optical microscope according to the arranged protocol.

Thorium and uranium were assumed to be in secular equilibrium with their corresponding daughters. If C_{U} and C_{Th} are the uranium and thorium concentrations (ppm) respectively in a sample, the etched track densities per unit exposure time (ρ) registered in CR-39 and etched hole density per unit exposure time in LR-115-II due to thorium and uranium series emitted α-particles are respectively given by [10]

$$\rho_{\mathrm{U}}^{\mathrm{CR}} = K_{\mathrm{U}} C_{\mathrm{U}}, \tag{1}$$

$$\rho_{\mathrm{Th}}^{\mathrm{CR}} = K_{\mathrm{Th}} C_{\mathrm{Th}}, \tag{2}$$

$$\rho_{\mathrm{U}}^{\mathrm{LR}} = K_{\mathrm{U}}^{*} C_{\mathrm{U}}, \tag{3}$$

$$\rho_{\mathrm{Th}}^{\mathrm{LR}} = K_{\mathrm{Th}}^{*} C_{\mathrm{Th}}, \tag{4}$$

where K_{U} and K_{Th} are the CR-39 and LR-115-II registration sensitivities for α-particles from uranium and thorium series respectively (expressed in terms of track cm^{-2} day^{-1} per ppm parent nuclei). K_{U}^{*} and K_{Th}^{*} have the same meaning for LR-115-II detector. From eqs (1) and (2) the total track density rate on CR-39 surface is given by

$$\rho_{\mathrm{t}}(\mathrm{CR}) = K_{\mathrm{U}} C_{\mathrm{U}} \left[1 + \frac{K_{\mathrm{Th}}}{K_{\mathrm{U}}} \frac{C_{\mathrm{Th}}}{C_{\mathrm{U}}}\right] \tag{5}$$

and the total etched holes density rate on LR-115-II is given by

$$\rho_{\mathrm{t}}(\mathrm{LR}) = K_{\mathrm{U}}^{*} C_{\mathrm{U}} \left[1 + \frac{K_{\mathrm{Th}}^{*}}{K_{\mathrm{U}}^{*}} \frac{C_{\mathrm{Th}}}{C_{\mathrm{U}}}\right]. \tag{6}$$

From the last two equations, we get the following relation between the total track densities and thorium-to-uranium ratio.

$$\rho' = \frac{\rho_{\mathrm{t}}(\mathrm{LR})}{\rho_{\mathrm{t}}(\mathrm{CR})} = \frac{K_{\mathrm{U}}^{*}\left[1 + \frac{K_{\mathrm{Th}}^{*}}{K_{\mathrm{U}}^{*}} \frac{C_{\mathrm{Th}}}{C_{\mathrm{U}}}\right]}{K_{\mathrm{U}}\left[1 + \frac{K_{\mathrm{Th}}}{K_{\mathrm{U}}} \frac{C_{\mathrm{Th}}}{C_{\mathrm{U}}}\right]}. \tag{7}$$

The sensitivity is given by [13].
(a) For CR-39:

$$K_{\mathrm{U}} = A_{\mathrm{U}}(Bq/g) \cos\theta_{\mathrm{c}} C_{\mathrm{U}} \sum_{i=1}^{8} \varepsilon_i R_i, \tag{8}$$

$$K_{\mathrm{Th}} = A_{\mathrm{Th}}(Bq/g) \cos\theta_{\mathrm{c}} C_{\mathrm{Th}} \sum_{i=1}^{7} \varepsilon_i R_i. \tag{9}$$

Table 2. Values of the LR-115-II to CR-39 sensitivity ratio $K^*_{\mathrm{U}}/K_{\mathrm{U}}$ for uranium series α-particles at different residual thicknesses of LR-115-II.

Residual thickness (μm)	$(K^*_{\mathrm{U}}/K_{\mathrm{U}})$
3	0.707
4	0.539
5	0.378
6	0.275
7	0.178
8	0.104
9	0.052
10	0.010

(b) For LR-115-II:

$$K^*_{\mathrm{U}} = A_{\mathrm{U}}(Bq/g)\cos\theta'_{\mathrm{c}} C_{\mathrm{U}} \times 8 \times \Delta R_s, \tag{10}$$

$$K^*_{\mathrm{Th}} = A_{\mathrm{Th}}(Bq/g)\cos\theta'_{\mathrm{c}} C_{\mathrm{Th}} \times 6 \times \Delta R_s, \tag{11}$$

where $A_{\mathrm{U}}(Bq/g)$ is the activity concentration of 1 ppm ^{238}U, $A_{\mathrm{Th}}(Bq/g)$ is the activity concentration of 1 ppm ^{232}Th, ε_i is the branching ratio in %. The critical angles of etching θ_{c}, θ'_{c} for CR-39 and LR-115-II (the critical angle for a detector is defined as the angle between the direction of the projectile and the normal to the detector surface under which no track can be revealed by etching) are respectively given by [14].

$$\cos\theta_{\mathrm{c}} = \frac{1}{11.6R_D^{-0.464}} \tag{12}$$

and

$$\cos\theta'_{\mathrm{c}} = \frac{1}{1+\mathrm{e}^{-0.27R'_D+3}}, \tag{13}$$

where R_D, R'_D are the ranges of α-particles in CR-39 and LR-115-II SSNTD in μm, R_i the range of α-particles in the sample material and $\Delta R_S = R_{\max}(E_{\max}) - R_{\min}(E_{\min}), R_{\max}$ and $R_{\min}$ are the ranges of α-particles in the sample which correspond to the energy windows $E_{\max}$ and $E_{\min}$ at the upper and lower ends, both of which are functions of the residual thickness of the detector and critical angle [12].

The part $(K^*_{\mathrm{U}}/K_{\mathrm{U}})$ was determined by Hafiz and Naim [13] and they established that this ratio does not depend on the material of the sample but it does depend on the residual thickness of LR-115-II as shown in table 2. The present etching conditions used are that correspond to a residual thickness of 6 μm.

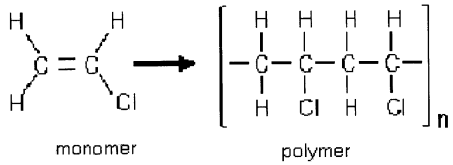

Figure 2. The molecular structure of polyvinyl chloride.

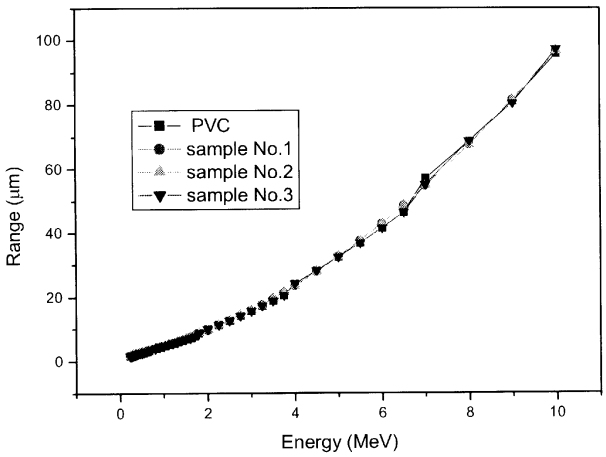

Figure 3. A comparison between α-particles range in PVC and the three soil samples.

3. Results and discussion

First we supposed that the range of α-particles in polyvinyl chloride (PVC) with chemical formula C_2H_3Cl (see figure 2) is almost equal to its range in the examined samples, then we calculated the range of α-particles in both material sample and PVC using TRIM-2003 program [15]. We noted a rather good evidence for the reality of our assumption as seen in figure 3.

For the ten samples prepared, first we studied the relation between the total activity and the track density on CR-39 and LR-115-II. As seen there is a linear relation for the two detectors (figure 4). It is obvious that as the activity increases the track density increases but it gives an impression that the present data are good. Also, we can approach the reality which declare that the spectrum of α-particles which can be detected by CR-39 is broader than that detected by LR-115-II.

Thorium-to-uranium ratio could be determined using calibration curve illustrated in figure 5, where it displays thorium-to-uranium ratio vs. track density ratio $\rho' = \rho(\mathrm{LR})/\rho(\mathrm{CR})$. The relation fits an exponential function of the form

$$\frac{\mathrm{Th}}{\mathrm{U}} = a\mathrm{e}^{\rho'/q}, \tag{14}$$

where $a = 0.3$ and $q = 0.205$.

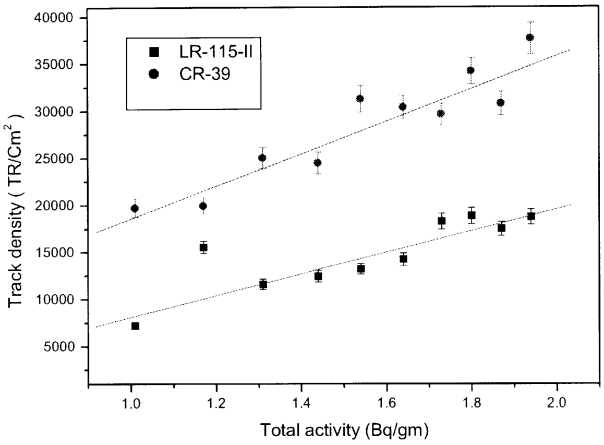

Figure 4. The track density in CR-39 and LR-115-II vs. total activity of uranium and thorium.

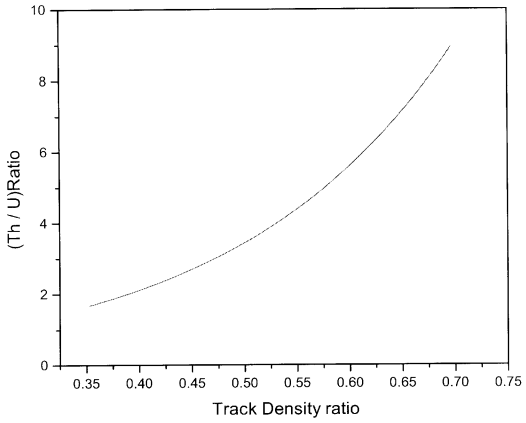

Figure 5. A calibration curve, where thorium-to-uranium ratio vs. track density ratio is displayed.

Substituting ρ' values for the soil samples we obtain data which agree very well with the real values for Th/U (see table 3). To determine the thorium and uranium contents, eq. (5) is used. Hafiz and Naim [13] established that the ratios $K_{\mathrm{Th}}/K_{\mathrm{U}}$ and $K^*_{\mathrm{Th}}/K^*_{\mathrm{U}}$ do not differ for different samples and their values are 0.3 and 0.25 respectively. All parameters are already known except K_{U} and K_{Th} if eq. (6) is used. It is enough to calculate K_{U} to obtain C_{U} and consequently C_{Th}. Table 4 gives the K_{U} values for the three soil samples and their uranium and thorium contents. It is clear that there is a good agreement between the present data obtained from this simple method and the real data. So following the same procedures for a sample of nearly the same density of PVC, it is necessary to know the track density rate ratio ρ' and substitute in eq. (14) to obtain Th/U value then calculate K_{U} to obtain thorium and uranium contents.

Table 3. Values of the track density ratio $\rho' = \rho(\mathrm{LR})/\rho(\mathrm{CR})$ obtained from the CR-39 and LR-115-II surface track densities and the calculated thorium-to-uranium ratio contents in the three soil samples.

	CR-39		LR-115		Th/U	
ρ'	Error	TR/cm^2	Error	TR/cm^2	Determined	Real
0.51	134	2732	99	1393	3.91	3.676
0.49	108	1809	71	886	3.40	3.307
0.40	129	2438	74	975	2.52	2.051

Table 4. Calculated registration sensitivities of CR-39 in close contact with soil samples containing uranium and thorium in secular equilibrium. The sensitivity is expressed in track/30 days for 1 ppm parent nuclide concentration in the sample at 10 μm thickness removed from the detector, obtained uranium and thorium content in the soil samples compared with the real values.

	Registration	U (ppm)		Th (ppm)	
Soil sample	sensitivity K_{U}	Our method	Real	Our method	Real
1	437	2.31	2.40	9.03	9.40
2	437	2.04	2.70	7.00	9.20
3	470	2.9	2.00	5.04	5.00

4. Conclusion

A new method using solid-state nuclear track detector for determining Th/U ratio in different soil samples has been developed. This method consists of a calibration curve of known thorium to uranium contents, and determination of the registration sensitivity of one of the two detectors as well as the track density of α-particles emitted from uranium and thorium series. This method has many advantages: it is not necessary to calculate the registration sensitivity for both detectors K_{U} or K^*_{U}, it's value for only one detector is quite enough to obtain thorium and uranium contents, it is simple, inexpensive and non-destructive and it may be applied to a wide variety of samples such as building materials, environmental samples and naturally occurring radioactive materials (NORM) in petroleum fields.

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