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Autoradiographic Study of Uranium and Thorium in Quartzite of the Kullu Area, India

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Abstract—The relation connecting the concentration of uranium and thorium atoms in geological samples with α-track production rates in a plastic track detector, LR-115 type 2, has been used for the analysis of quartzite samples collected from the Kullu area, Himachal Pradesh, India. The α-track measurements from the samples under secular equilibrium define the total uranium and thorium concentrations, while fission track measurements yield uranium concentration alone, thus by combining the results of both measurements, uranium, thorium and isotopic equilibrium/disequilibrium is determined. Distribution of uranium and thorium in radioactive quartzites was studied by taking α-particle track and fission track autoradiographs. Autoradiographic patterns reveal the highest track densities in the radioactively darkened zones of the samples.

1. INTRODUCTION

Uranium and thorium are natural α-emitters and an emulsion detector when placed against the surface of a geological sample should record the α-disintegrations. Coppens[1] has given the uranium and thorium content of a sample as a function of α-track production rate (N) as:

$$10^6\,\mathrm{N} = 9.20\,k\mathrm{U}, \tag{1}$$

if there is only uranium without thorium or

$$10^6\,\mathrm{N} = 2.80\,k\mathrm{Th}, \tag{2}$$

if there is only thorium without uranium and

$$10^6\,\mathrm{N} = (9.20\,\mathrm{U} + 2.80\mathrm{Th})k, \tag{3}$$

if both uranium and thorium are present.

Equations (1)–(3) have been modified[2] for use with solid state nuclear track detectors which can record α-particle tracks more precisely. We used cellulose nitrate plastic (LR-115 type 2) and modified equations (1)–(3) for this plastic track recorder respectively as:

$$10^6\,\mathrm{N} = 6.77\,k\mathrm{U}, \tag{4}$$

$$10^6\,\mathrm{N} = 1.60\,k\mathrm{Th}, \tag{5}$$

$$10^6\,\mathrm{N} = (6.77\,\mathrm{U} + 1.60\,\mathrm{Th})k, \tag{6}$$

where

$$k = \frac{0.85}{\sum \frac{CS}{A}}. \tag{7}$$

In equation (7), C is the concentration of an element of atomic weight A and S is the stopping power of an α-particle for that element. Knowing the value of k and estimating uranium by fission track technique, the thorium concentration can be easily determined from equation (6). A comparison between the uranium concentrations determined by α-autoradiography and those by

other methods (viz. fission track, fluorimetry) allows the existence of radioactive equilibrium or disequilibrium to be established.

In the present investigations the modified relation was employed for uranium, thorium and isotopic equilibrium/disequilibrium studies of some quartzite samples collected from the Kullu area of Himachal Pradesh (India).

2. EXPERIMENTAL TECHNIQUE

Polished sections of the samples are prepared by grinding with emery powder of mesh size varying from 100 to 600 μm and polishing with cerium oxide. To obtain α-autoradiographs, a sheet of LR-115 type 2 plastic is placed against the surface of the samples, which is stored undisturbed for a period of time to allow the α-tracks to accumulate. A surface area of <1 cm^2 is convenient, both for preparation and, later, for track scanning. After the accumulation period the plastic sheet is removed and etched for 2 h in 2.5 N NaOH at 50°C. The α-tracks are then counted using an Olympus binocular microscope at a magnification of 600X. The background of α-tracks in LR-115 type 2 plastic is checked and is found to be zero.

To obtain a fission track radiograph, a sheet of lexan plastic is placed against the surface of the same samples and is sent to the reactor for thermal neutron irradiation along with suitable standards. After irradiation, the plastic detectors are removed and etched in 6.25 N NaOH at 70°C for 25 min to obtain the fission track radiographs of the samples and these are then scanned under a microscope to determine the track densities.

3. RESULTS AND DISCUSSION

For the determination of uranium and thorium concentrations in quartzite samples it is sufficient to determine the number (N) of the optically visible α-tracks emitted per cm^2/s and apply to equations (4)–(6). However, the value of k, which depends upon the composition of the sample and is given by:

$$k = \frac{0.85}{\sum \frac{CS}{A}},$$

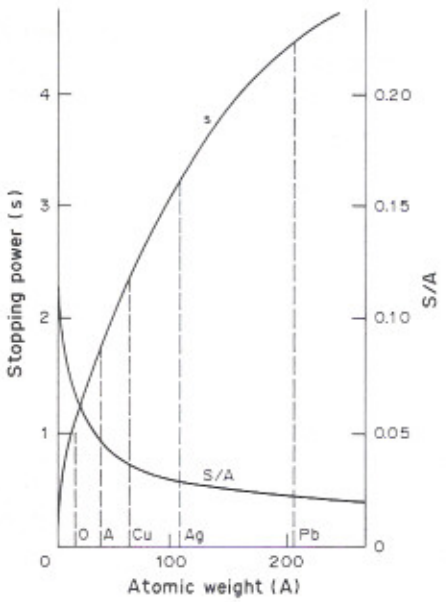

Fig. 1. Plot of S/A vs A.

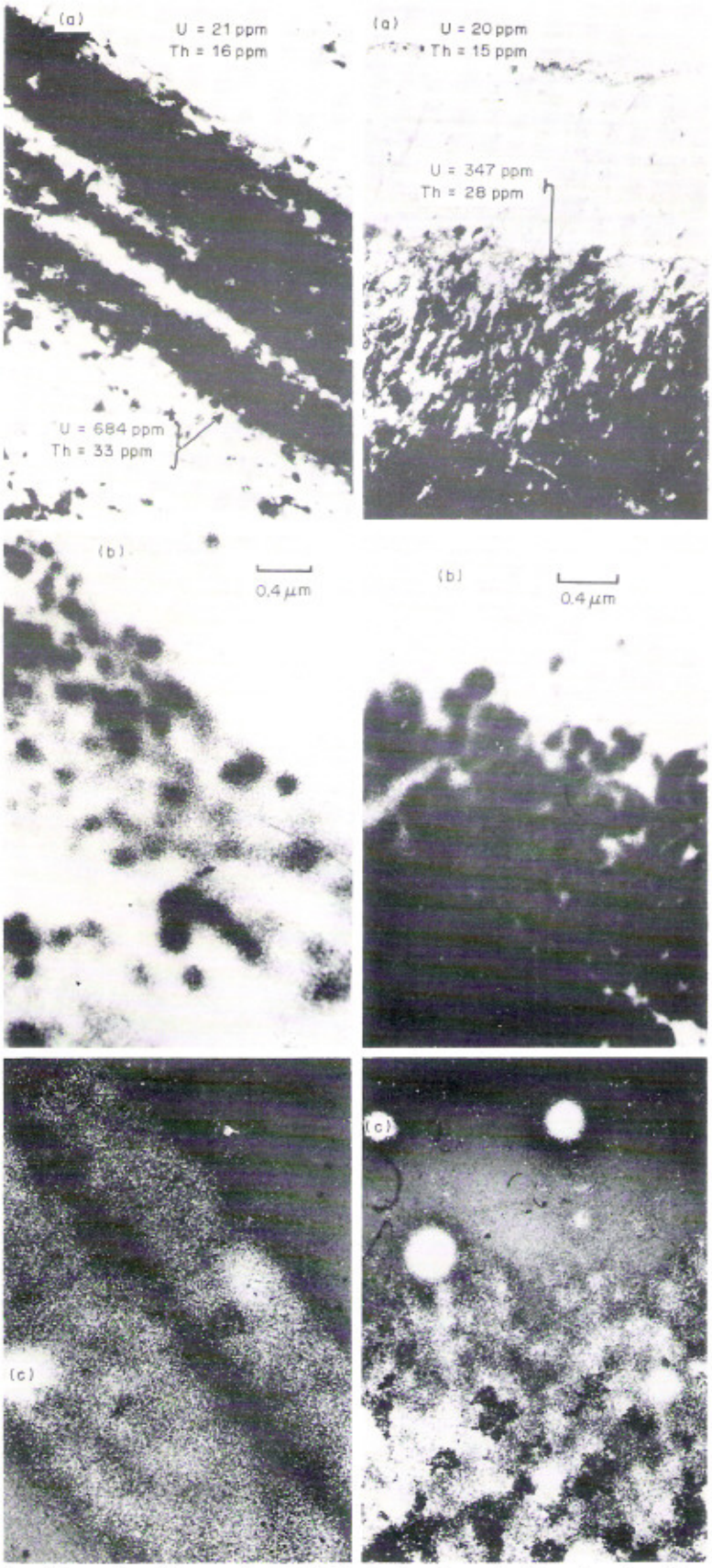

Fig. 2. Microphotographs of the samples, (a), fission track radiographs (b) and α-autoradiographs (c) of two different quartzite samples.

Table 1. Calculation of k for quartzite

Element	A	C	S/A	CS/A
Silicon	28	0.4667	0.052	0.2426840
Oxygen	16	0.5333	0.066	0.0351978
			$CS/A =$	0.0594662
			$k =$	14.29

is not known. The constant k is calculated taking the composition of the quartzite as SiO_2 and S/A values from the plot of S/A vs A (Fig. 1). The value of k is calculated as 14.29 for quartzite (Table 1).

3.1. *Uranium concentrations by α-autoradiography*

If U and Th represent the concentrations of uranium and thorium (in ppm), N the number of α-tracks per cm^2/s, and k the coefficient of absorption of α-particles, we have

$$10^6\,N = (6.77\,U + 1.60\,Th)k$$

if both uranium and thorium are present. With $k = 14.29$, the above equation becomes

$$10^6\,N = 96.74\,U + 22.86\,Th$$

and,

$$U = \frac{10^6 N - 22.86\,Th}{96.74} \tag{8}$$

Knowing N, the α-track rate, and Th, we can calculate the value of U. The α-autoradiographic study is carried out on the samples already analysed[3] by γ-ray spectrometry. With the thorium concentrations determined by γ-ray spectrometry the value of uranium is determined using expression (8). Uranium contents determined by α-autoradiography and γ-ray spectrometry are reported in Table 2. The values are found to agree which shows the validity of the α-autoradiography for uranium estimation.

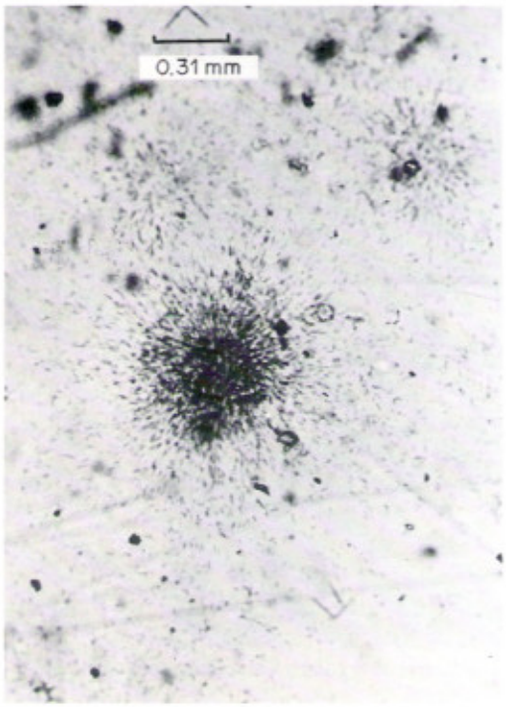

Fig. 3. Photograph of radiating cluster of fission tracks.

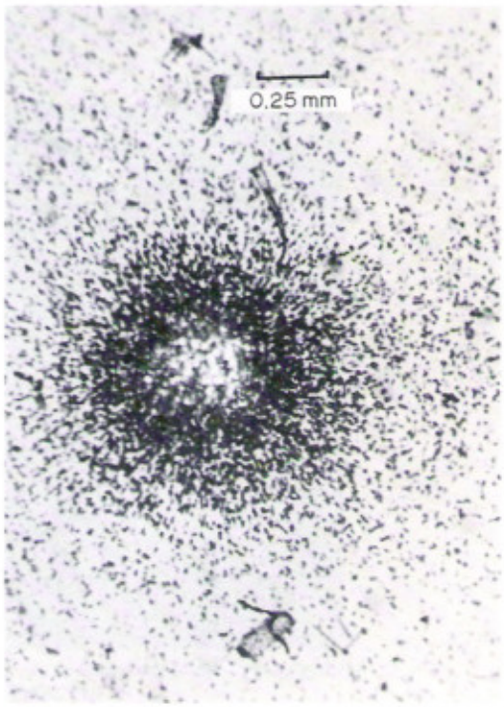

Fig. 4. Photograph of radiating cluster of α-tracks.

Table 2. Uranium concentration in quartzite samples by γ-ray spectrometry, α-autoradiography and fission track analysis

Sample location	Sample number	γ-Ray spectrometry Th (ppm)	U_g (ppm)	α-Autoradiography 10^6 N/cm^2	U_α (ppm)	Fission track analysis U_f (ppm)
Rohtang	RQ1	3.14	3.29	368	3.06	3.59
Pass	RQ2	11.00	3.13	537	2.95	3.31
	RQ3	6.12	3.24	478	3.49	4.19
Jari	JQ1	101.00	671.00	72200	702.00	667.00
	JQ2	149.00	366.00	39900	377.00	348.00
	JQ3	40.20	93.00	9155	83.00	86.80
Kasol	KQ1	25.60	61.00	6052	56.00	73.06
	KQ2	46.50	93.00	10403	96.00	86.00

U_g—Uranium content determined by γ-ray spectrometry;[13] U_α—U content determined by α-autoradiography; U_f—U content determined by fission track analysis.

3.2. *Study of radioactive equilibrium*

Uranium concentrations calculated by α-autoradiography and fission track analysis (which gives the real content of the uranium without taking into account its daughter products) are given in Table 2. A comparison of the results show that uranium determined by both techniques are in good agreement, indicating thereby the existence of radioactive equilibrium in these samples.

3.3. *Measurement of thorium content*

We know that:

$$10^6\,\mathrm{N} = (6.77\,\mathrm{U} + 1.60\,\mathrm{Th})k,$$

so that

$$\mathrm{Th} = \frac{10^6\,\mathrm{N} - 96.74\,\mathrm{U}}{22.86}, \quad \text{for} \quad k = 14.29. \tag{9}$$

Putting in the values of N and U, Th can be calculated.

The quartzite samples collected from three different locations of the Kullu area are analysed using the above relation. Uranium concentrations in these samples are determined using the fission track technique. Uranium and thorium concentrations are found to vary from 2.63 to 18.73 ppm and 1.55 to 86.70 ppm, respectively (Table 3).

3.4. *Study of uranium and thorium distribution*

Microphotographs of radioactive quartzite rock specimens, the fission track and α-track radiographs are shown in Fig. 2. Detailed inspection of the autoradiographs show that the α-track autoradiographs match exactly in size, shape and position to those of the fission track radiographs

Table 3. Thorium analysis of quartzite samples of Kulu area (Himachal Pradesh)

Sample location	Sample No.	10^6 N	Uranium content (ppm)	Thorium content (ppm)
Shat	QS1	1014	2.88 ± 0.07*	32.16 ± 0.60
	QS2	1998	18.73 ± 0.19	8.09 ± 0.26
	QS3	410	3.59 ± 0.09	6.91 ± 0.26
	QS4	714	2.63 ± 0.06	20.10 ± 0.44
	QS5	698	5.06 ± 0.08	9.12 ± 0.21
Rohtang	QR1	2808	8.54 ± 0.11	86.70 ± 1.00
	QR2	673	3.85 ± 0.07	13.14 ± 0.34
Bunter	QB1	350	3.25 ± 0.07	1.55 ± 0.05
	QB2	571	3.31 ± 0.07	10.97 ± 0.23
	QB3	1913	7.85 ± 0.10	50.46 ± 0.75

Value of k for quartzite samples = 14.29.
*Statistical counting error $(1\sigma) = 1/\sqrt{T}$.
T tracks per cm^{-2}.

of the specimens. The track rich areas of these autoradiographs in turn match exactly the dark areas of the specimens. The uranium and decay products in these specimens are, therefore, concentrated in dark zones. The presence of such uranium enriched zones in quartzites has already been reported[4] and found to be radioactively darkened.

Qualitative observations with a microscope show that the track rich areas of the fission track autoradiographs are largely made up of radiating clusters of tracks (Fig. 3), and in these areas the surface is almost completely covered with tracks. Track counting in these track rich areas of fission track radiographs is therefore difficult. To overcome this difficulty, the same samples were irradiated at a lower neutron dose. Radiating clusters of α-tracks (Fig. 4) are also present in the α-autoradiographs, but the exposure time is so adjusted that even in most of the track rich areas of these α-autoradiographs the tracks can be counted. In areas of uniform or smoothly varying track density the uranium concentrations measured by means of fission tracks are used in equation (6) to calculate the precise values of thorium concentration in different zones of these samples. Uranium and thorium concentrations estimated in different zones of these specimens are reported in Fig. 2. The results reveal the existence of high concentration of uranium only in the dark zones of the quartzite samples.

4. CONCLUSIONS

The present studies reveal that the modified formulation between uranium, thorium concentrations and α-track rate employing LR-115 type 2 plstic detector based on α-autoradiography can successfully be employed in the study of radioactive equilibrium/disequilibrium and for uranium/thorium estimation and distribution in geological samples.

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