

## Method for Estimation of Uranium, Thorium & Potassium in Rocks using Gamma Ray Spectrometry

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Gamma ray spectrometric method for rapid determination of uranium, thorium and potassium in rocks using well-type NaI (Tl) crystal coupled to multichannel analyzer, is described. Critical sample weight requirement is found to be 16 g for  $4\pi$  detector geometry. Combination of gamma ray peaks at 1.46, 1.76 and 2.62 MeV is found to be most suitable for K, U and Th analysis. The experimental set-up provides a good reproducibility of results.

### 1 Introduction

The method of estimating concentration of three radioactive elements, viz. U, Th and K by gamma ray spectrometry is well known<sup>1-3</sup>. In the U spectrum, the prominent gamma ray peaks at 0.61, 1.12 and 1.76 MeV are often employed for the analysis of uranium. In the Th spectrum, prominent peaks at 0.24, 0.94 and 2.62 MeV are used for analysis of thorium, while the prominent 1.46 MeV peak of  $^{40}\text{K}$  is used for potassium<sup>2</sup>. Based on these gamma energy peaks of U, Th and K spectra, the combinations of gamma ray peaks used for analysis in various energy regions are as follows:

High (MeV)	Medium (MeV)	Low (MeV)
1.46	1.46	1.46
1.76	1.12	0.61
2.62	0.94	0.24

Using  $10.16 \times 5.08$  cm NaI(Tl) detector coupled to a single channel analyzer, Rao<sup>1</sup> has described relative errors in analysis with 400 g samples in  $2\pi$  geometry using high energy peaks centred at 1.46, 1.76 and 2.62 MeV. In the present work, we have employed well-type  $7.62$  (dia)  $\times 7.62$  cm NaI(Tl) detector coupled to highly stabilized multichannel analyzer set-up. Using  $4\pi$  detector geometry, the following studies have been undertaken:

- The requirements for set-up (viz. the critical mass and background),
- The comparison of errors using gamma peaks in various energy regions, and
- The errors, reproducibility and comparison of gamma peaks in high energy region.

### 2 Experimental Details

**Description of spectrometric set-up**—The detector is an integral assembly consisting of a well-type  $7.62$

(dia)  $\times 7.62$  cm NaI (Tl) crystal coupled to a photomultiplier tube obtained from Bicron Ltd, USA. The detector offers provision for sample analysis in  $4\pi$  geometry using a well-type crystal (Fig. 1). Resolution of the detector is 7.5% for  $^{137}\text{Cs}$ . This detector is coupled to a multichannel analyzer (MCA) through preamplifier, amplifier and analog-to-digital converter (ADC). The spectrometer is calibrated using different gamma sources of known energy.

**Background reduction**—A basic requirement in low level counting is the reduction of background ra-

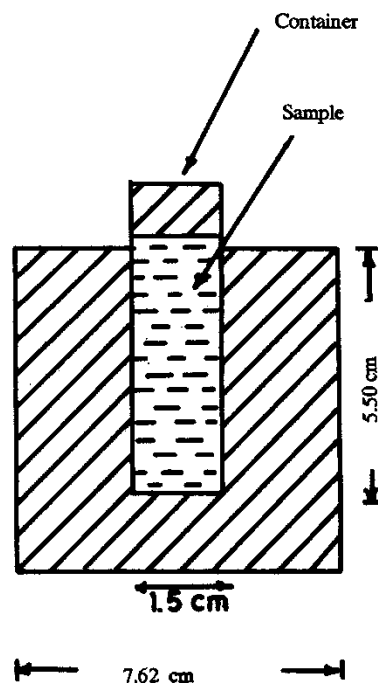


Fig. 1—Sample in  $4\pi$  detector geometry

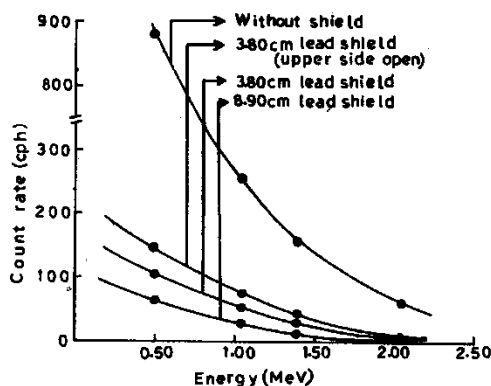


Fig. 2—Variation of count rate per hour (cph) with energy using different lead shields

Table 1—Results of Total/Background Ratio for Gamma Peaks of Different Energies in  $4\pi$  Detector Geometry

Source	Around gamma peak (MeV)	Total/background
Uranium (312 ppm)	0.61	1.17
	1.12	1.75
	1.76	4.83
Thorium (166 ppm)	0.24	4.07
	0.94	1.91
	2.62	3.25
Potassium (26.6%)	1.46	2.22

diations and the enhancement of total background (T/B) ratio. Lead shield is employed for background reduction. Background count rate variation in 0 to 2.5 MeV energy region is studied using lead shielding of different geometrical thicknesses (Fig. 2).

Values of T/B ratio achieved for different gamma peaks with U (312 ppm), Th (166 ppm) and K (26.6%) sources in  $4\pi$  detector geometry for a counting time of 1 hr, are reported in Table 1.

**Critical mass studies**—To find out the minimum limit for sample weight required for the analysis, experiments with varying sample weights are performed. The plot of count rate versus sample weight (Fig. 3) shows that the count rate becomes independent of weight beyond 16 g in  $4\pi$  detector geometry.

**Preparation of standard and the sample**—Uranium and thorium standards are prepared from samples obtained from the National Geophysical Research Institute, Hyderabad. These samples, 350 g each, are U and Th ores diluted in dunite. U standard contains 312 ppm and 53 ppm Th. Th standard contains 166

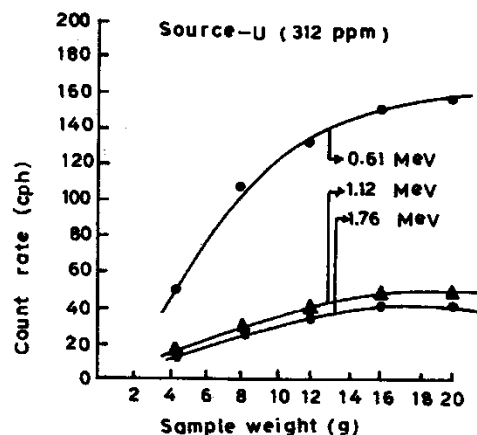


Fig. 3—Plot of count rate versus sample weight in  $4\pi$  detector geometry

ppm Th and 5 ppm U. Potassium dichromate salt (26.6% K) is used as K standard. The sample to be analyzed is ground to 100 mesh and is sealed in a container. The containers are air-tight cylindrical plastic jars.

**Counting procedure**—Sample is placed in the well and the spectrum is taken for sufficient time, depending on the activity of the sample. Spectra of U, Th and K standards (Fig. 4a, b, c) of comparable activity are recorded along with background spectrum for the same period using a teleprinter (Decwriter IV) coupled to MCA.

**Method of data analysis**—Matrix method developed by Stromswold and Kosanke<sup>4</sup> is used for data analysis as follows:

The observed count rate  $R$  due to a concentration  $C$  of gamma-emitting elements can be written as:

$$R = AC \quad \dots(1)$$

where  $A$  is proportionality constant.

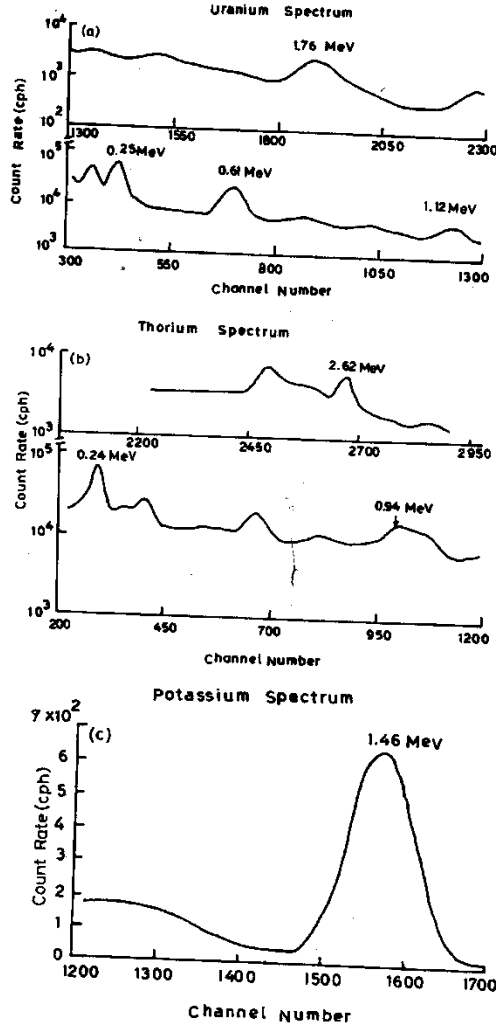
The right side of Eq. (1) can be expanded as follows to include contributions to the counts  $R$ , from all gamma rays

$$R = AC + \sum A' C' \quad \dots(2)$$

where the summation ( $\sum$ ) is over all gamma emitters that contribute to  $R$  by energy shifts into the  $R$  window. For K, U and Th analyses, there are three terms on the right side of Eq. (2):

$$R = A_1 C_1 + A_2 C_2 + A_3 C_3 \quad \dots(3)$$

where  $C_1$ ,  $C_2$ , and  $C_3$  represent the concentrations of potassium, uranium, and thorium, respectively.


 Fig. 4—Spectra of (a) uranium (b) thorium, and (c) potassium samples taken with  $7.62 \times 7.62$  cm well-type NaI(Tl) detector

Because  $R$  can represent the count rate in any energy window, it is convenient to adopt the following index notation:

$$R_i = \sum_{j=1}^3 A_{ij} C_j \quad \dots (4)$$

where  $R_i$  is the count rate in the  $i$ th window,  $C_j$ , concentration of  $j$ th radioactive element, and  $A_{ij}$  is the constant relating count rate in  $i$ th window to the concentration of  $j$ th element.

Eq. (4) can be written in a matrix notation as:

$$R = AC \quad \dots (5)$$

where  $R$  and  $C$  are  $3 \times 1$  matrices and  $A$  is a  $3 \times 3$ . In this approach, Eq. (4) specifies one element,  $R_i$ , of the matrix  $R$ .

Eq. (5) can be inverted to give:

$$C = A^{-1} R \quad \dots (6)$$

where  $A^{-1}$  is the matrix inverse of  $A$ .

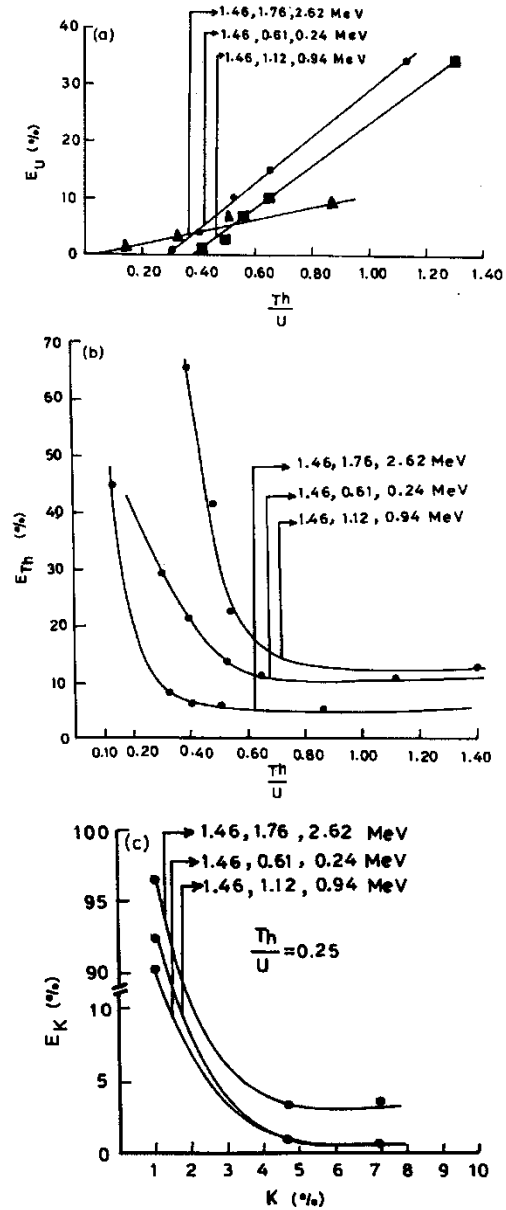


Fig. 5—Relative error of (a) uranium (b) thorium as a function of Th/U and (c) potassium as a function of K

The matrices  $A$  and  $A^{-1}$  are determined by detector system calibration using sources of known concentration.

### 3 Results and Discussion

**Set-up requirement of studies**—The set-up requirements as ascertained by initial studies, are summarized as follows:

(a) From the variation of count rate in 0-2.5 MeV energy range, using different lead shields, it is observed that 8.90 cm thickness of lead reduces the background sufficiently (Fig. 2). With our set-up, the background reduction is better as compared to the set-up reported by Rao<sup>1</sup> (using 15.24 cm thick mild steel + 7.62 cm thick lead shield).

(b) The variation of count rate with sample weight in  $4\pi$  detector geometry is shown in Fig. 3, and it is found to be independent of sample weight greater than 16 g. Thus the sample requirement for  $4\pi$  geometry is fixed at 20 g to avoid sample weight correction.

**Selection of gamma ray peaks**—Combinations of gamma ray peaks in low, medium and high energy regions are tried in sample analysis. T/B and relative errors (error in estimation of one element due to relative abundance of other elements) are studied for selec-

tion of gamma ray peaks (Table 1). From comparison it is evident that peaks under high energy region show better T/B ratio compared to peaks under low and medium energy regions. Using three combinations of gamma peaks, relative errors in analysis are also evaluated [Figs 5(a, b and c)]. Figs 5(a and b) show that high energy combination results in low relative error both in U and Th analysis. But relative error in K analysis is slightly more for high energy combination compared to low and medium energy combinations (Fig. 5c). This may be due to the fact that 1.46 MeV energy region lies in the back-scattered Compton contributions of high energy gamma rays<sup>5</sup> (viz. 1.76 and 2.62 MeV).

Overall, the high energy combination is found to be the most suitable due to (i) high T/B under gamma peaks (ii) low relative error and (iii) the ease in computation work<sup>2</sup>.

**Sample analysis**—Samples are analyzed using high energy combination of gamma peaks (1.46, 1.76, 2.62 MeV) and  $4\pi$  detector geometry. Using standard samples, relative errors encountered in the analysis are studied [Figs 5(a, b and c) and 6(a, b, c and d)]. Fig. 5(a and b) shows the variation of relative errors with Th/U. Errors depending upon the relative abundance of K are reported in Fig 6(a, b, c and d). These

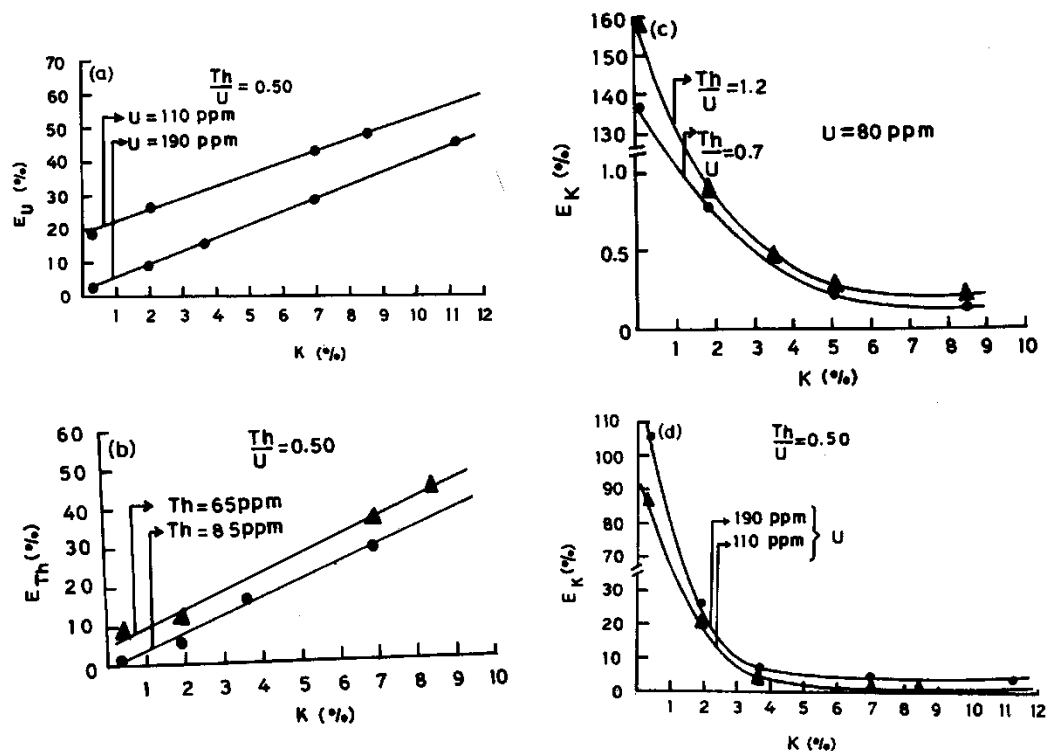


Fig. 6—Relative error of (a) uranium (b) thorium (c) potassium and (d) potassium as a function of K

Table 2—Comparison of Gamma Ray Spectrometric Results with Fission Track Analysis

Sample No	Gamma ray spectrometry			Fission track analysis
	U (ppm)	Th (ppm)	K (%)	U (ppm)
Quartzite from Jari in Himachal Pradesh				
Q1	671.00 ± 5.00	100.90 ± 0.70	3.14 ± 0.20	667.00 ± 5.00
Q2	367.00 ± 4.00	149.10 ± 0.80	24.20 ± 0.60	348.00 ± 3.00
Q3	168.00 ± 2.60	35.90 ± 0.40	5.30 ± 0.26	178.60 ± 1.30
Phosphorite from Maldeota in Uttar Pradesh				
P1	49.50 ± 1.40	7.95 ± 0.19	1.01 ± 0.11	44.10 ± 0.50

errors are incorporated to get the final results. Reproducibility of results is also checked by repeating the analysis of the same sample at three different times. The quartzite samples collected from Jari (Kulu) HP and phosphorite sample from Maldeota (Mussoorie Syncline) UP are analyzed using this set-up. The concentrations of U, Th and K in quartzite samples are found to vary from  $168.00 \pm 2.60$  to  $671.00 \pm 5.00$  ppm,  $35.90 \pm 0.40$  to  $149.10 \pm 0.80$  ppm and  $3.14 \pm 0.20$  to  $24.20 \pm 0.60\%$  respectively (Table 2). The concentrations of U, Th and K in phosphorite sample are found to be  $49.50 \pm 1.40$  ppm,  $7.95 \pm 0.19$  ppm and  $1.01 \pm 0.11\%$ , respectively. The concentration of U in these samples is pronouncedly higher than that reported by Menon *et al.*<sup>3</sup> This is due to the presence of uranium mineralization in these areas<sup>6,7</sup>. A comparison of the results for U content in these samples with fission track method<sup>8,9</sup> is also reported (Table 2). The results obtained by the two techniques are in good agreement.

#### 4 Conclusion

The gamma ray spectrometric technique using well-type detector provides the facility for analyzing samples where large amounts are not available. The technique is quite efficient for U, Th and K analyses and the results show a good reproducibility.

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