URANIUM AND THORIUM MEASUREMENTS ON PRECAMBRIAN ROCKS OF SOURTHERN INDIA[1]

SURENDRA PAL*

RESUMEN

Se midieron los contenidos de Uranio y Torio en rocas precambricas del sur de la India, por espectrometría de rayos γ. Se discuten los resultados de medidas en unas 40 muestras en términos del comportamiento geoquímico conocido de estos elementos.

ABSTRACT

Uranium and thorium contents of Precambrian rocks of Southern India have been measured by γ - ray scintillation spectrometry. The results of measurements on some 40 specimens are discussed in the light of known geochemical behavior of these elements.

INTRODUCTION

Ever since the discovery of natural radioactivity, uranium and then thorium have been of increasing geologic and geochemical interest. There are many intimate and interesting relationships between the goechemistry of thorium and uranium and the problem of geologic dating and economic development. Another special aspect of their geochemistry is the heat produced in the earth by the radioactive disintegration of Th, U and their daughters.

A study was therefore undertaken to understand the behavior of these elements under different geological conditions.

EXPERIMENTAL DETAILS

The various analytical techniques of uranium and thorium measurements have been discussed by many authors, e.g., Hurley (1956), Eichholz et al (1958), Cherry (1963) and Hamilton (1965). The choice of a particular method is governed by the concentration of uranium and thorium present. Chemical, spectrophotometric and radioactivation procedures are more suitable when analyzing samples having high concentrations of uranium and thorium. At a few parts per million level of concentration, they are rather less accurate, time-consuming and subject to serious interference from other elements. The fluorimetric method of analysis though rapid, has low accuracy. The mass spectrometric isotope dilution technique is, of course, quite precise, sensitive and accurate. But the radiometric methods have been found to be very useful in uranium and thoruim assay. These can be broadly classified into two groups:

a) Direct methods —based on the properties of U and Th themselves and include $\alpha-$ detection by ionization chambers, scintillation counters and photographic plates.

b) Indirect methods —based on the properties of daughter elements, and the amount of uranium and thorium present is deduced indirec-

tly. They include the β-and γ -counting methods: the α-γ, β-γ and γ-γ methods, α-discrimination methods, emanation flow counters and photographic methods. Of these, the most common method of radiometric assay is the γ -scintillation spectrometry.

An important point about the γ-ray scintillation counting is that the method utilizes a large amount of rock sample making it easier to get a representative sample, while other methods make use of very small quantities of sample material which may hardly be a representative of the original rock-body.

γ -RAY SCINTILLATION SPECTROMETRY

Figure 1 gives the block diagram of the Na I(TI) scintillation spectrometer constructed in the laboratory. A photograph of this instrument is shown in Fig. 2. The different parts are described in the following.

SCINTILLATION HEAD

The Na I(TI) scintillation crystal has the dimensions 1 3/4" dia. x 1" height and is of the aluminium canned-type. This is the general purpose crystal supplied by Harshaw Chemical Company. The crystalunit is a single package with the crystal hermetically sealed and optically coupled to the transparent window of 2" diameter of DuMont 6292 photomultiplier. Good optical coupling is ensured with
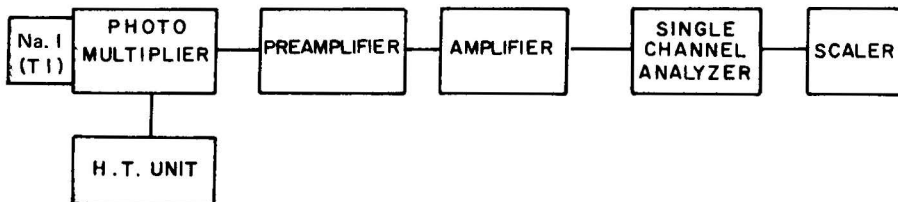

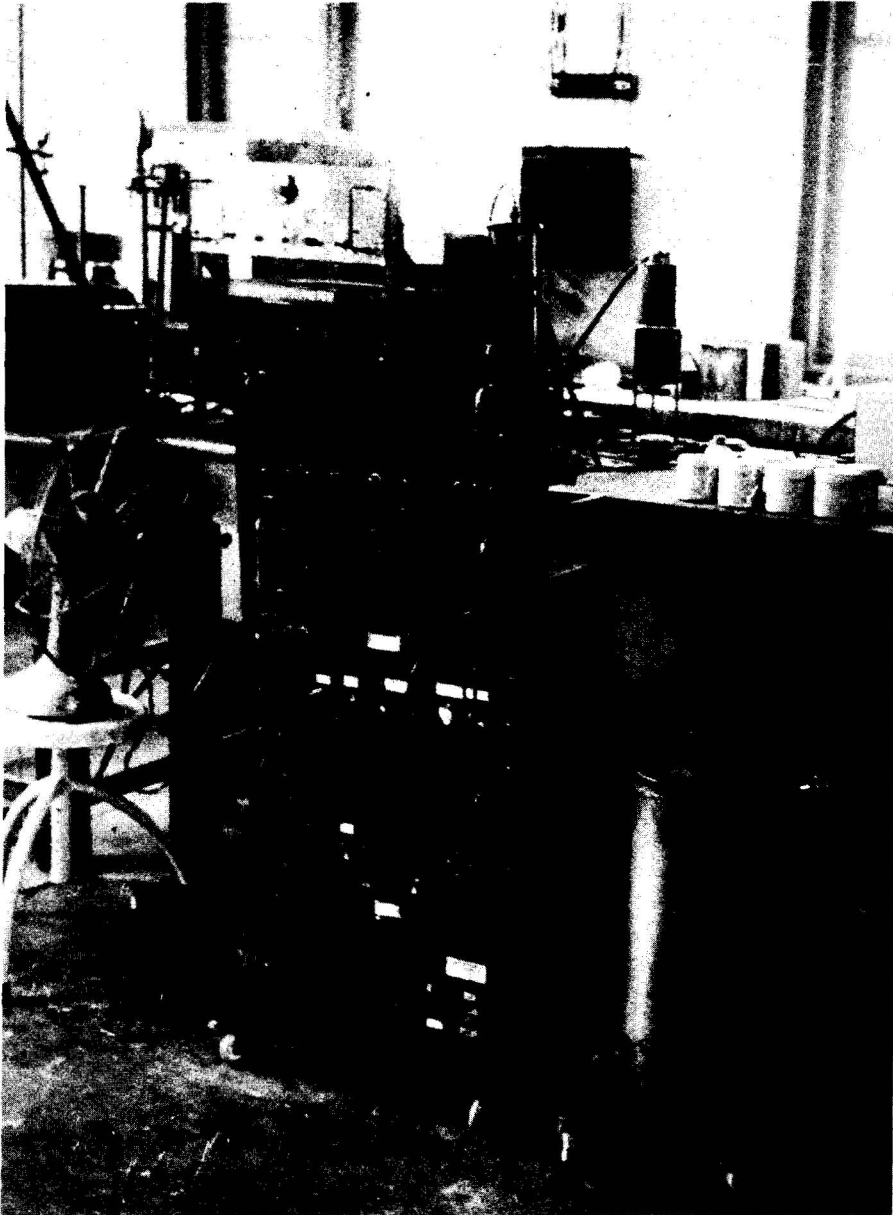Figure 1. Block diagram of the γ —Ray Spectrometer.

Figure 2. Photograph of the γ —Ray Spectrometer.

a transparent viscous medium such as Dow Corning 200 silicon fluid. Light leaving the crystal through the sides and rear face is reflected towards the photocathode by a reflecting material such as Alumina.

PREAMPLIFIER AND MAIN AMPLIFIER

The circuit of the base connections of the photomultiplier together with the preamplifier connected to it is given in Fig. 3. The voltage difference between the photocathode and the first dynode is kept about two times larger than that between the other dynodes to improve the resolution of the photomultiplier. The preamplifier is simply a cathode follower consisting of a 6AK5 tube, the plate voltage of which is supplied by a stabilized low voltage (300V) supply. The output of the photomultiplier is fed through a capacitor to the grid of this tube whose output is fed to the main amplifier with the help of a coaxial cable.

The main amplifier is essentially the A_1 amplifier designed originally by Jordan and Bell (1947) and later improved for its linearity and overload performance by Bell, Kelly and Goss (Price 1958). This refined version of the A_1 amplifier has been used in the present instrument. It consists of two degenerative feed-back sections and a cathode follower output stage. Each section has two gain stages and a cathode follower. While the original circuit uses three pairs of RC networks for controlling the rise and decay times, the present circuit employs only one such pair of RC networks resulting in a rise and decay times of 0.7 and $4\mu sec$ respectively. The response of the amplifier is found to be linear for output pulses upto 12OV. The overall gain of the amplifier (max. 10^4) can be varied by two controls — one stepwise by factors of two and the other continuous in between.

LOW VOLTAGE UNIT

This is a 300V stabilized power supply whose design is based on the model 50 power supply (Elmore and Sands 1949). The output voltage is stable to within $\pm$ 0.05% for a $\pm$ 10% change in the input at 200

mA output. The ripple content of the output is < 10 mV peak to peak. This unit supplies power to the preamplifier and the main amplifier.
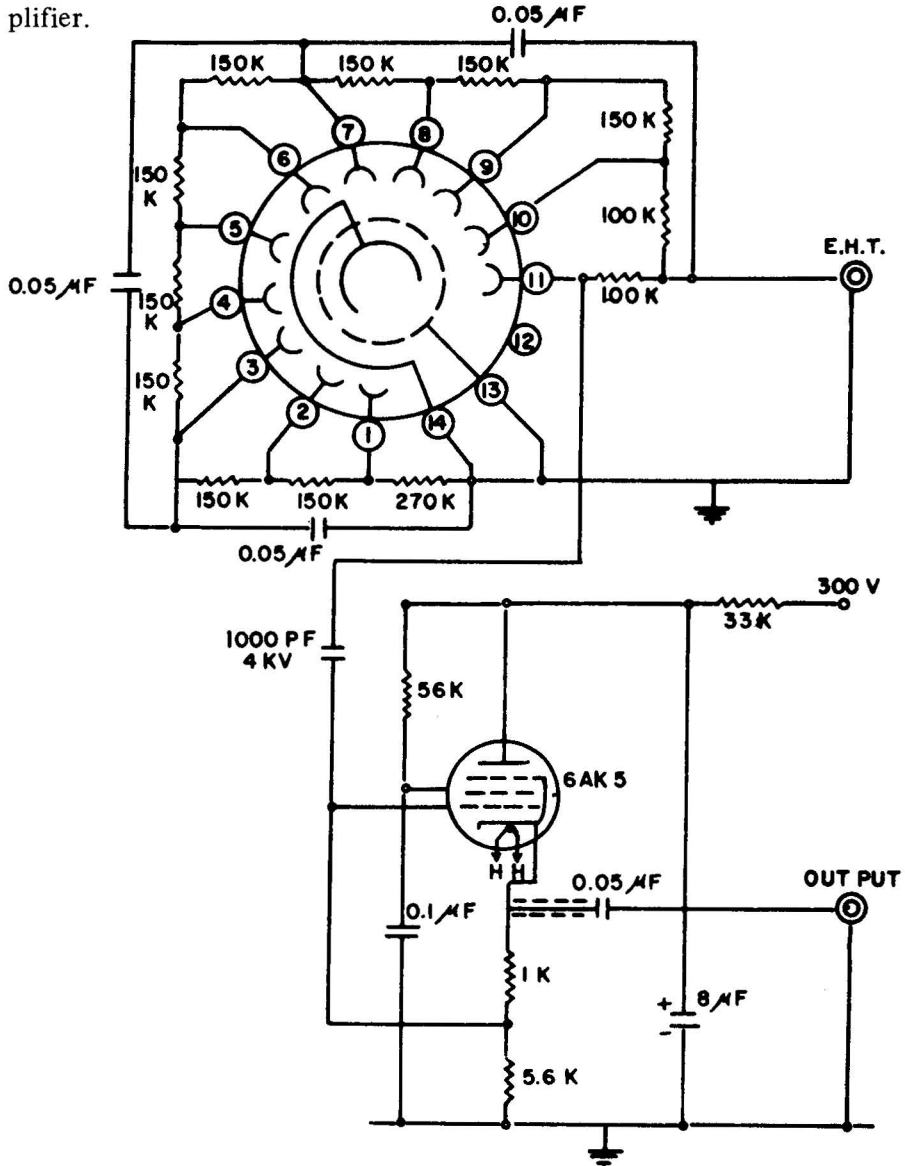

Figure 3. Photomultiplier and preamplifier circuits.

STABILIZED HIGH TENSION SUPPLY

Since the gain in a photomultiplier is very sensitive to the voltage applied to it, a stable voltage source is essential for spectroscopic applications. The circuit of the high voltage unit used is shown in Fig. 4. It is of the series stabiliser type and the design is based on that of O'Toole (1959). This has the following important advantages over conventional types using special valves with high anode voltage rating.

1. The variation of output voltage can be easily effected by varying the potentiometer R_2.

2. Since the voltage at the point A increases by the same amount as the output, the net drop across the amplifier in the feed back circuit is kept relatively small and roughly constant, so that receiver type valves such as 6SL7 can be used instead of the expensive valves with high anode voltage rating.

3. For a wide range of the output voltage, the triodes V_{2a} and V_{2b} operate under essentially the same conditions. The chief advantage is that one optimum condition of operation can be chosen and designed.

4. The balanced twin triode arrangement reduces the effects of valve aging and heater voltage variation.

5. The feedback amplifier is modified to a cascade circuit by using one half of the double triode, for regulating the series valve screen supply. Considerable reduction of the output impedance and increase of the stabilization ratio thereby result from the increased gain. The shunt regulation valve V_{5a} is stabilized by the voltage across V_3 and V_4.

The output voltage is positive and can be continuouly varied from 800 to 1100V. Maximum available current is about 5 mA, and the ripple at 1000V is $< 0.5V$ peak to peak. A 10% change in the line voltage gives a variation in the high voltage which is less than 0.1% at 1000V.
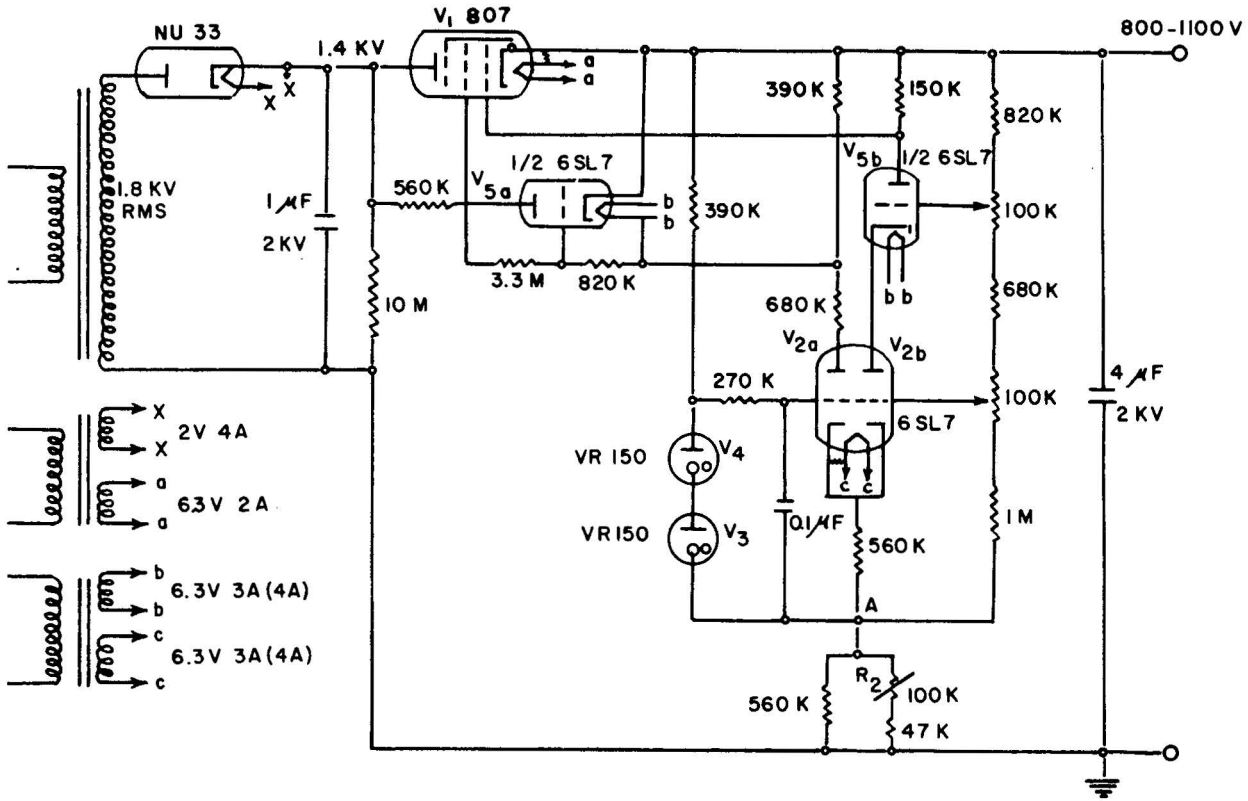
Figure 4. Stabilized high tensions supply.

SINGLE CHANNEL ANALYZER

This unit (type AN 800) is supplied by the Bhabha Atomic Research Centre, Bombay, India. As is well known, the purpose of a single channel analyzer is to provide an output pulse only if the input pulse lies between two preset voltages, and to reject all pulses above the upper voltage and below the lower voltage. It consists of two discriminators conventionally called lower and upper, connected in anticoincidence. The discriminator level of the lower discriminator is controlled by a ten turn helical potentiometer, while that of the upper discriminator lies above that of the lower by an amount which is controllable by an ordinary potentiometer —'window width'. The setting of the discriminator bias and window-width control provides a channel or window adjustable from 0 to 6 volts in width over the range 3 to 103 volts.

An expander amplifier to amplify the window 10 times ensures very accurate discrimination by the upper discriminator. If the input voltage is large enough to trigger both the discriminators, the anticoincidence circuit is actuated to stop the pulse output from the lower discriminator. The output of the single channel analyzer can be directly fed to a scaler for counting.

SCALER

It is a Radart Decatron scaler, type 902A, supplied by the Eastern Electronics, Faridabad, India. It is designed to count and register the number of electrical pulses arriving in a random manner over a period of time. The counter-circuit of the instrument is formed by three decade tubes connected in cascade to form a scale of 1000. It has its own discriminator unit with a pre-amplifier stage incorporated before the discriminator for better sensitivity. The events are recorded on a relay-operated, impulse recorder. All the stages are operated from an electrically regulated power supply (also incorporated in the unit) for better stability.

SOURCE HOLDER ASSEMBLY

In order to lower the background (when radioactively weak samples are to be measured), a cylindrical lead shield of 1" wall thickness is used. This can be graded with Cd or Cu for better definition of the spectrum in low energy region.

Aluminium canisters used as sample holders were cylindrical in shape with an annular recess to cover the crystal. The Rock samples crushed to ~ 50 mesh were packed inside the canister so that the sample weights for nearly the same geometry were ~ 200 gms.

Fig. 5 shows schematically the scintillation detector along with the source unit, the preamplifier and the lead shield.

TESTING OF THE INSTRUMENT

A few drops of the radioisotope ^{65}Zn (equivalent to a fraction of a mcurie; ^{65}Zn-Eγ 1.11 Mev and T$_{1/2}$ 250 days) were deposited on an aluminium plate and the source was placed above the scintillator crystal. The photomultiplier voltage and the amplifier gain can be adjusted so as to bring the ^{65}Zn γ-ray peak well inside the discriminator range (i.e., below 100V setting) at any desired value Fig. 6 gives a typical γ-ray spectrum of ^{65}Zn for a particular setting of the instrument gain. It is clearly seen from this spectrum that the γ-ray spectrometer has a resolution of ~ 8% at this γ-ray energy of 1.11 Mev.

Next, the gain parameters were adjusted so as to record the γ-ray spectra of the uranium and thorium series in secular equilibrium. Samarskite and uraninite were used to study the γ-ray spectrum of the uranium series, and monazite and thoria for the thorium series. These spectra are shown in Figs. 7 and 8. respectively. They have been recorded with different gain-settings. The γ-ray spectrum of potassium was also recorded using large quantity of natural K_2CO_3 (^{40}K ~ 0.0119%) and is shown in Fig. 9.

Figure 5. Scintillation detector.
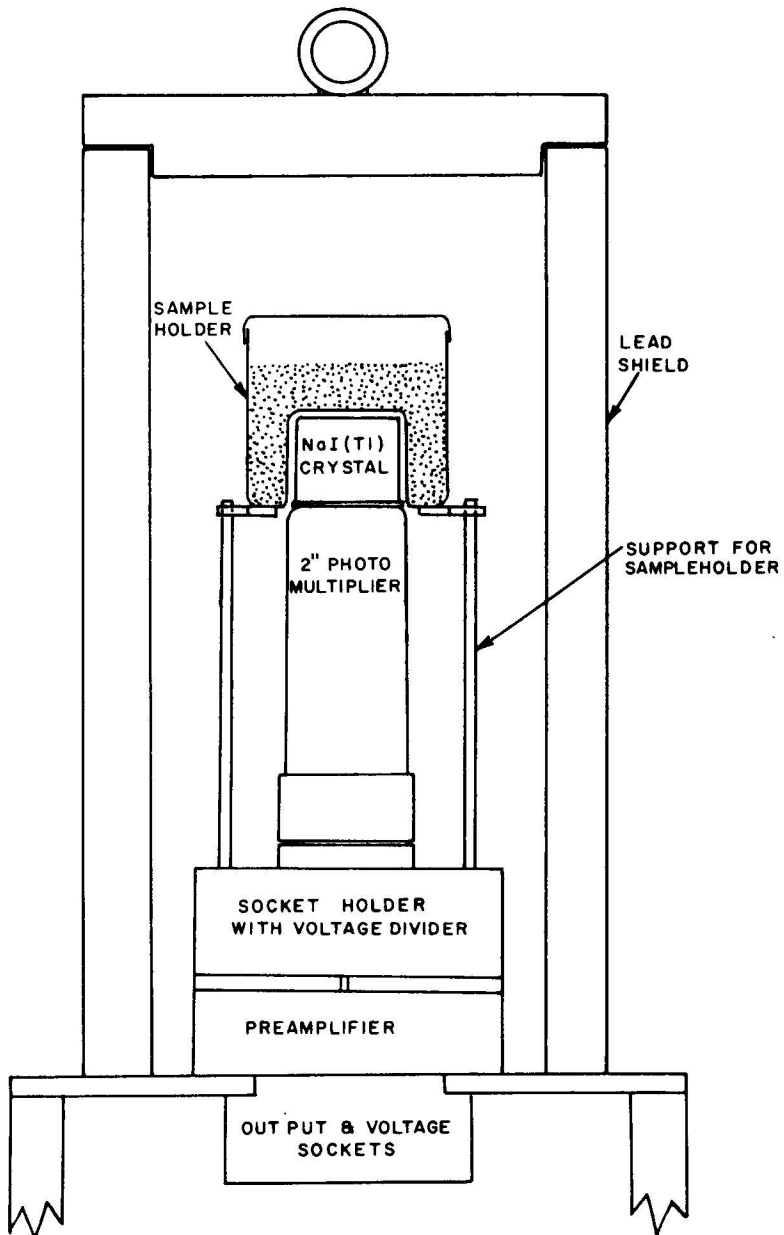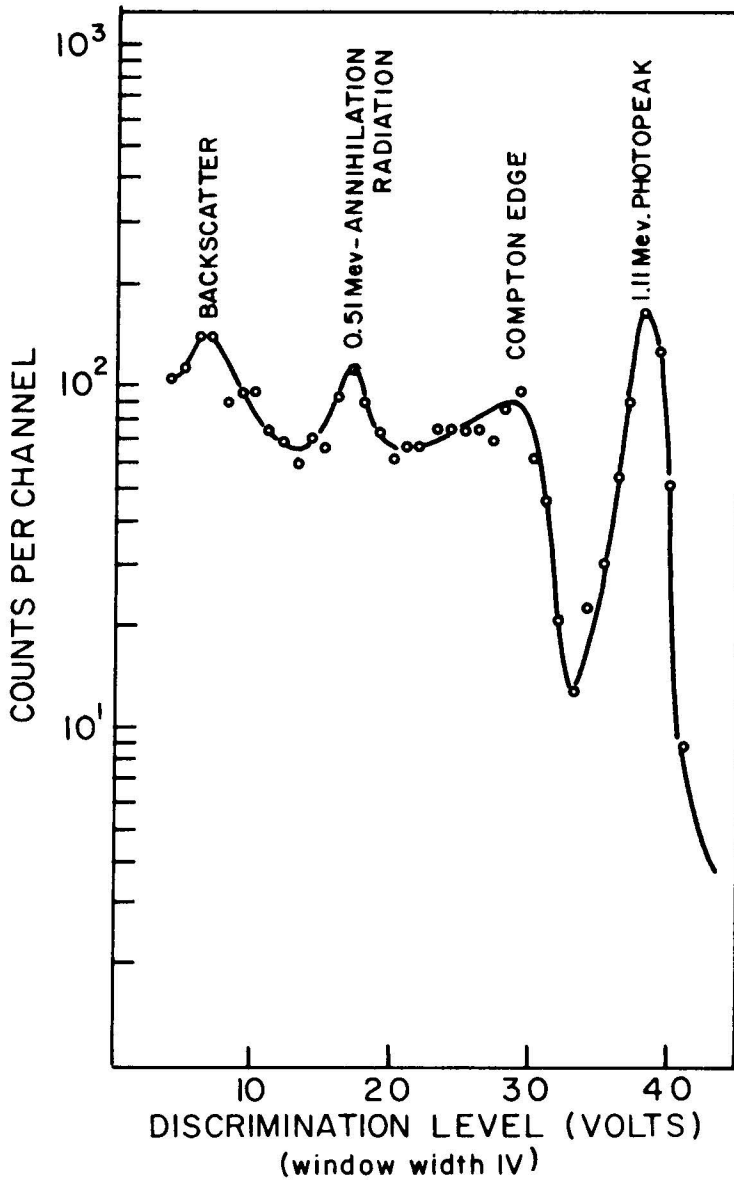
Figure 6. γ Ray spectrum of 65Zn.
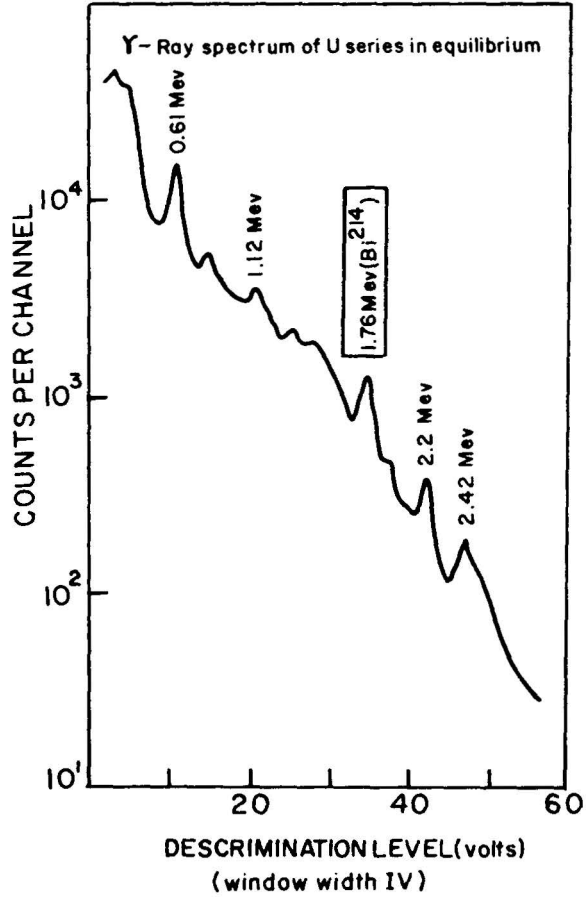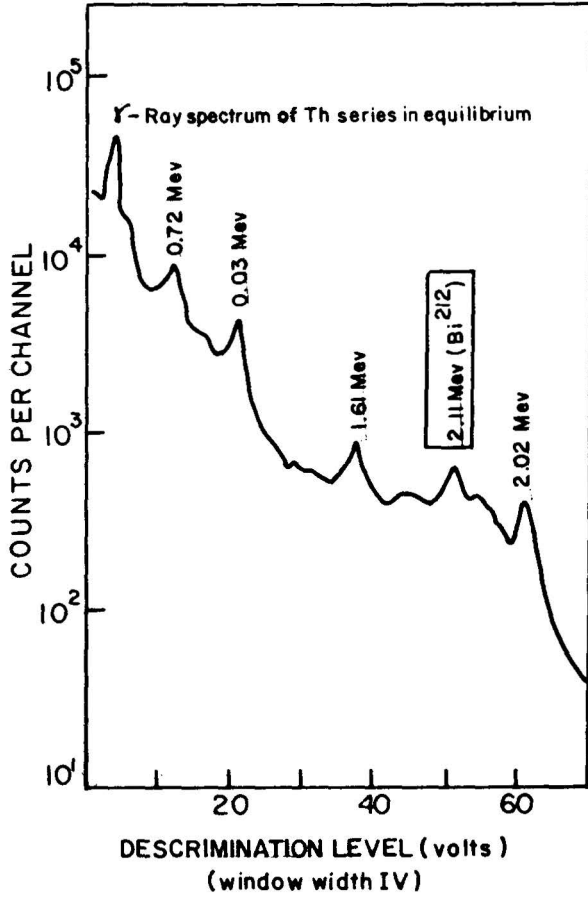
Figure 8. γ Spectrum of Th-series in equilibrium

Figure 7. γ Spectrum of U-series in equilibrium

Figure 9. γ ray spectrum of ^{40}K in natural patassium
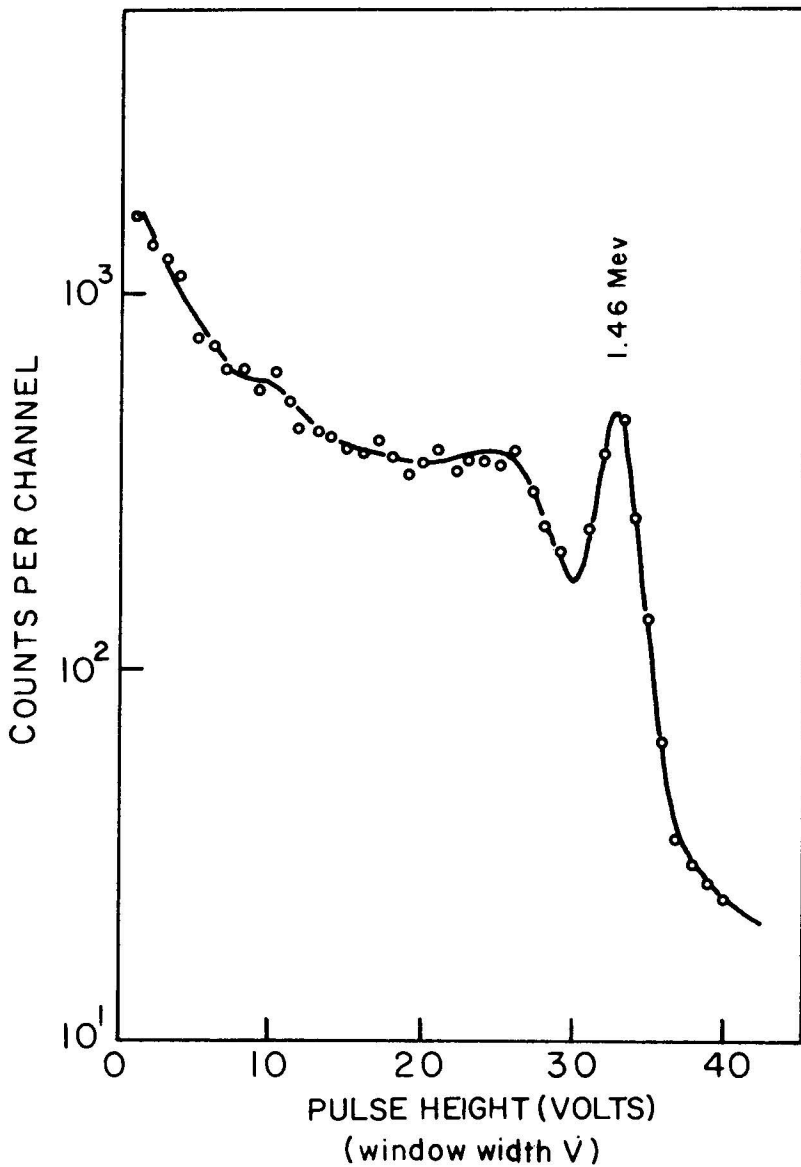
CALIBRATION AND THE METHOD OF MEASUREMENT

For uranium and thorium measurements, the counting rates were taken in two channels each centered around a γ-ray peak – one from the uranium and the other from the thorium series. Higher energy peaks were chosen so as to avoid any interference from potassium. Lower energy peaks as used by Hurley (1956), could be chosen, but the counts should be corrected for the potassium contribution.

In the present work, the 1.76 Mev (^{214}Bi) peak of the U-series as used by Bloxam (1962), was chosen for the uranium channel and the 2.11 Mev (^{212}Bi) peak of the Th-series for the thorium channel. The 2.11 Mev peak was found to yield a better sample to background ratio than the 2.62 Mev (^{208}Tl) peak used by Bloxam (1962). At 1.76 Mev, a channel width of 4V ($\sim$ 0.14 Mev) was used but was increased to 6V ($\sim$ 0.21 Mev) at 2.11 Mev. It is apparent from the energy distribution of U and Th γ - rays that in a sample containing uranium and thorium, the thorium will contribute counts to the uranium channel and the uranium to the thorium channel. These contributions should therefore be taken into account in the calculation of the uranium and thorium contents of a sample. Potassium could also be estimated by taking counts in a third channel containing the 1.46 Mev γ -ray peak of ^{40}K and deducting the contribution of uranium and thorium. The contribution of ^{40}K γ -rays in uranium and thorium channels, however, was found to be negligible.

One fundamental assumption that we make in this method of uranium and thorium determinations, is that both uranium and thorium series are in secular equilibrium, which is fortunately the case with most of the rock and mineral occurrences. In the uranium series, 95% of equilibrium is established in 3.5×10^5 yrs. In the thorium series only 30 years are required.

Calibrations were made using synthetic standards prepared from analyzed specimens obtained from the Atomic Minerals Division, Department of Atomic Energy, India. Two primary standards (500 ppm U and 1000 ppm Th) were prepared in large quantities by dilution, with very low active trap rock (total activity $<$ 1 ppm equivalent U)

of samarskite (8.614% U) and pure ThO_2. From these standards substandards containing known amounts of U (25-300 ppm) and Th (50-800 ppm) were prepared. Weights of 200 gms were filled in sample-holders and the measurements of U and Th on these substandards agreed to within 5%. The calibration curves drawn from these measurements are given in Fig. 10.

Every time while analyzing unknown samples, counting rates with the standards (both U and Th primary standards) were taken in order to check the calibration. The spectrometer was allowed to warm up and stabilize for a few hours before taking the counting rates. Samples and background counts were alternately taken and repeat-counts were made to ensure that there had been no drift during the period of measurements. Total counting times (for sample as well as background) of 24 hours and more were used. The statistical errors in these measurements are calculated by using the method given by Hurley (1956). The errors vary from 3% at 20 ppm Th to 8% at 5 ppm Th and 12% at 1 ppm U to 4% at 5 ppm U. Repeat measurements at intervals of 1 month agree to within 5%.

TYPICAL CALCULATION

The suffixes 1 and 2 refer to the observations in uranium channel (1.76 Mev peak) and thorium channel (2.11 Mev peak) respectively. (U_1, U_2), (T_1, T_2) and (R_1, R_2) are the counting rates in the two channels for the uranium standard, thorium standard and the sample respectively (subtracting the background counts from the net counting rates). U and Th are the amounts of uranium and thorium in ppm in the uranium and thorium standards respectively.

$$U_2/U_1 = Q_U, \; T_2/T_1 = Q_T, \; R_2/R_1 = Q_R,$$

$U/U_1 = C_U$ and $T/T_1 = C_T$ (both say, in ppm/count/hr). The sample counting rate arises from contributions A from the uranium and B from the thorium present in it. Thus
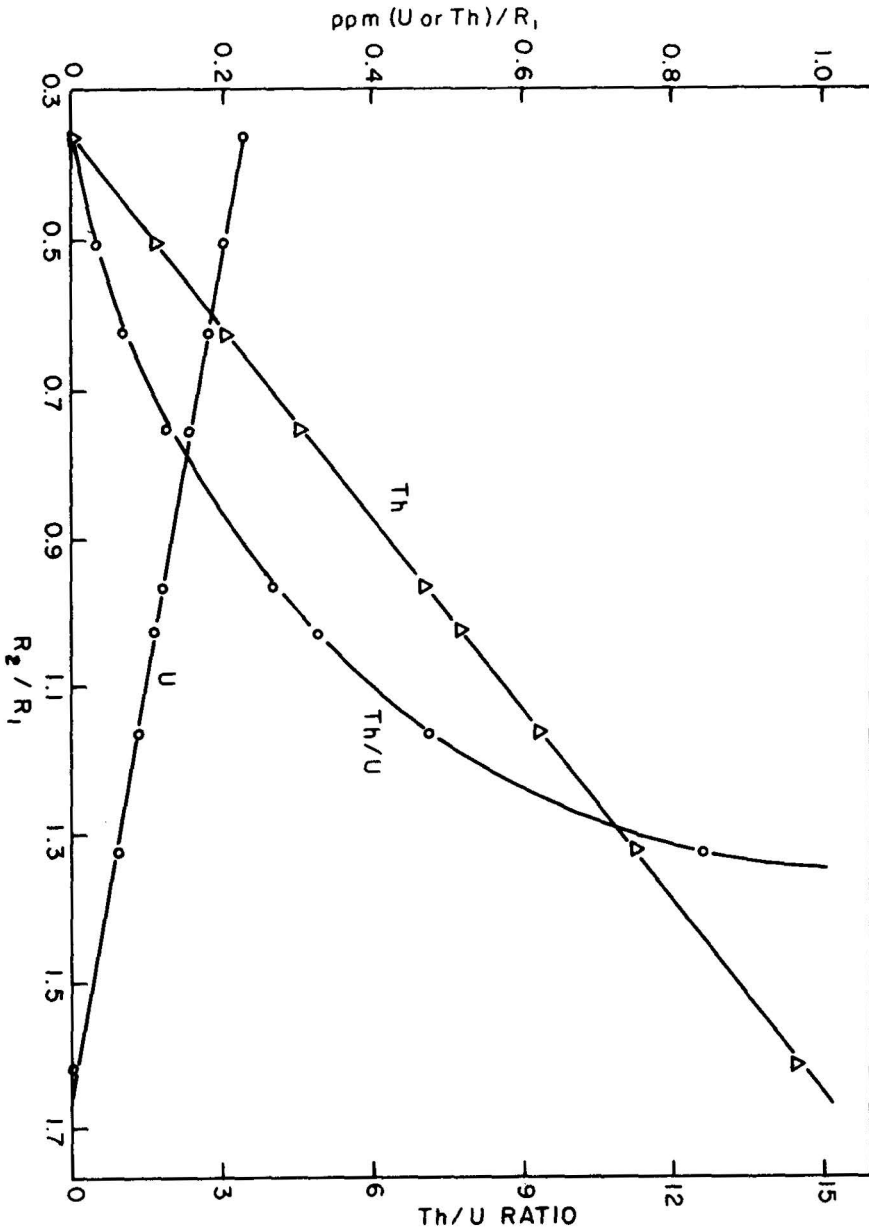
Figure 10. Calibration curves for Uranium, Thorium and Thorium/Uranium ratio

$$R_1 = A_1 + B_1$$

$$\text{and } R_2 = A_2 + B_2 = \frac{U_2}{U_1} A_1 + \frac{T_2}{T_1} B_1 \quad \text{(assuming secular equilibrium)}$$

From these

$$A_1 = \frac{Q_T - Q_R}{Q_T - Q_U} R_1 = F.R_1,$$

$$\text{if } \quad F = \frac{Q_T - Q_R}{Q_T - Q_U}$$

Similarly $B_1 = (1 - F) R_1$.

Therefore   ppm Uranium in the sample = A_1/U_1 x ppm U in std.

$$= F R_1 . U/U_1$$
$$= C_U.F.R_1$$

Similarly   ppm Thorium in the sample = $C_T (1 - F) R_1$

In the present set up, we get for the 500 ppm U standard and 1000 ppm Th standard:

$$U_1 = 2150/hr. \qquad\qquad U_2 = 780/hr.$$
$$T_1 = 1030/hr. \qquad\qquad T_2 = 1670/hr.$$

The average background counting rates in either channel $\sim 180/hr$.

Therefore, $Q_U = 0.363$, $Q_T = 1.62$, $C_U = 0.2325$ and $C_T = 0.971$ (both C_U and C_T in ppm/count/hr).

For the specimen — Auriferous quartz vein 380L, Western reef, Kolar

Counting rate in uranium channel = 17.5/hr.

Counting rate in thorium channel = 16.7/hr.

Therefore $Q_R = 0.95$.

Uranium content of the sample = $C_U F.R_1 = 2.4 \pm 0.2$ ppm.

Thorium content of the sample = $C_T (1-F)R_1 = 7.9 \pm 0.5$ ppm.

The errors of the measurements are calculated by the method of Hurley (1956).

RESULTS AND DISCUSSION

The results of the measurements on rocks of the Nandidrug mines, Kolar Gold Fields are given in Table 1 and those on Charnockites from different localities in Table 2.

The geochemistry of uranium and thorium and their distribution among various rock-types have been discussed by e.g., Larsen et al (1954) and Adams et al (1959).

The measurements on hornblende schists clearly show that their uranium and thorium contents are remarkably uniform, while the Th/U ratios also show only a very small spread about the mean value of 3.5. The results for the schists altered by contact with the reef show a definitely large spread, especially in their Th/U ratios. It has been observed by several workers, e.g., Larsen and Gottfried (1960), Heier and Rogers (1963), that the scatter of the Th/U ratios from the average is greater, the more complex the history of the differentiation of the source material producing the rocks— this being undoubtedly due to the tendency of the uranium to be leached during the deuteric stages that characterise the later history of the igneous and metamorphic rock-bodies. Such secondary processes are unimportant for the hornblende schists — this being truer for basic rocks in general than their acidic intrusives.

Heat flow measurements in this area have been given by Verma and Rao (1965). They give for the hornblende schists, an average value of terrestrial heat-flow of 0.66×10^{-6} cal/cm² sec ± 14% based upon a mean value of thermal conductivity of $(6.3 \pm 0.7) \times 10^{-3}$ cal/cm sec °C, taking the temperature gradient to be 10.4 °C/Km (density of the rocks $\sim$ 3 gms/cm³). This value of average heat flow is well below the continental average of 1.4 μcal/cm² sec, but only a little lower than the average values given for other shield areas of the world (e.g., African 0.95, Canadian 0.87 and Western Australia 0.9 to 1.01 etc.).

The results for the auriferous quartz veins show a definite increase in the U and Th contents, which is even more pronounced for pegmatites. This is in accordance with the general trend for the concentra-

tions of these elements to increase during differentiation; the effect of the ionic radii of U^{4+} and Th^{4+} (0.97 and 1.02A° respectively) and their high ionic potentials resulting in a strong partitioning of these elements in the liquid phase during fractionation. The increase in U and Th concentrations from the mafic to felsic types is well known.

Table 1

U and Th contents of rocks of Kolar Gold Fields

Specimen	U ppm	Th ppm	Th/U
Auriferous quartz veins, Western Reef, Nandidrug mines, Kolar			
380 L	2.4	7.9	3.3
670 L	1.8	8.3	4.6
1070 L	1.7	7.6	4.5
1450 L	2.0	8.0	4.0
2000 L	2.8	7.5	2.7
3000 L	3.7	6.3	1.7
4300 L	3.7	7.3	2.0
4800 L	4.0	6.3	1.6
5500 L	3.1	6.9	2.2
6200 L	3.9	7.4	1.9
Altered hornblende schists, Footwall Contact, Western Reef, Kolar			
380 L	2.1	6.1	2.9
670 L	1.7	7.2	4.2
1070 L	1.5	6.9	4.6
1850 L	1.9	7.1	3.7
2000 L	2.2	6.8	3.1
3000 L	2.3	5.8	2.5
4400 L	1.9	7.0	3.7
4800 L	2.4	5.8	2.4

(contd.)

(continuation)

Specimen	U ppm	Th ppm	Th/U
550 L	2.1	6.3	3.0
6000 L	2.0	5.6	2.8
6200 L	2.5	6.2	2.5
Hornblende schists, Western Reef, Kolar			
1450 L	1.3	4.4	3.4
1850 L	1.5	4.9	3.3
4300 L	1.4	4.7	3.3
4800 L	1.2	4.4	3.8
6000 L	1.4	5.0	3.6
Pegmatites, Western Reef, Kolar			
3300 L N22	7.2	10.0	1.4
3650 L N20	6.7	7.2	1.1
3650 L N34	14.1	19.8	1.4
3800 L N30	10.2	15.6	1.5

Table 2

U and Th contents of charnockite of S. India

Locality	Specimen	U ppm	Th ppm	Th/U
Holenarasippur	Magnetite bearing charnockite schist	4.1	2.9	0.7
Kanjamalai Hills, Salem	Acid charnockite	2.3	5.1	2.2
Yercaud, Salem	Acid charnockite	2.1	6.3	3.0
Doddayampatti, Salem	Feldspathised-charnockite	1.3	6.2	4.8

(contd.)

(continuation)

Locality	Specimen	U ppm	Th ppm	Th/U
Salem	Charnockite	1.2	4.7	3.9
	Charnockite	1.7	5.5	3.2
Ongole Hills	Acid charnockite	8.8	107	12.1
	Acid charnockite	7.2	112	15.6

The Th/U ratios of the vein rocks show an interesting trend in that they decrease with the depth of the zones of occurrence of the sam ples, this being due to the decrease in the uranium contents nearer the surface, the thorium contents remaining more or less constant. This can be ascribed to secondary processes as a variation of hydration with depth, leading to oxidation of the uranium to the hexavalent state and the removal of the soluble uranly ion UO_2^{2+}. The problem of such an oxidation of uranium during metamorphic differentiation is controversial; thus Heier (1962) found no evidence of oxidation and selective leaching of uranium from the rocks of the amphibolite facies from Lanjoy, Norway. On the other hand, Adams et al (1959) adduced some evidence that metamorphic processes tend to introduce more U than Th, so that this would be a factor in produccing lower values of Th/U ratios. Narayanaswamy and Venkatasubramanian (1969) report a decrease in Th/U ratios in the sequence gneiss-granite-pegmatite of the amphibolite facies rocks of this area.

Lambert and Heier (1967) found that acid and intermediate pyroxene granulite subfacies rocks are depleted in U and Th by factors of about 5 and 9 respectively when compared with common surface rocks of similar bulk composition. This depletion could result from a crust made up of a series of different layers with different chemistries as commonly considered in geophysics. The low Th and U granulites would come from a layer within the crust that has always been low in these elements. But the authors do not favour such static models because normal geological processes operating during crustal evolution are capable of causing large scale movement of elements. They also state

that the variations of chemical composition with depth in the crust result from different mobilities of the elements during these processes.

Heier (1965) and Heier and Adams (1965) discussed the movement of elements within the crust during regional metamorphism and observed that the trace elements are not quantitatively transferred from one mineral assemblage to the other by the suggested mineral transitions. The processes of regional metamorphism and magmatism merge at high temperatures. Partial melting begins at upper amphibolite facies conditions At granulite facies conditions, more melting may take place and the liquid may migrate upward to lower pressure environments. Different degrees of partial melting in the crust would add to the general effect of metamorphism and cause the continental crust to be progressively depleted in the granitophile elements with depth.

Our present measurements on a few charnockites show a wide spread in Th/U ratios – the U and Th contents being normal. Some of them have normal Th/U ratios characteristic of the earth's crust ($\sim$ 3.5), while others show low ratios (even less than 1) similar to those found in pegmatites. Two samples from Ongole Hills have much larger uranium and thorium contents with much higher Th/U ratios. Unpublished work by Narayanaswamy in this laboratory shows high Th/U ratios and high Th contents (though lower U contents) for charnockites of Pallavaram, Madras, India. These results thus do not show the expected depletion of U and Th in such rocks of granulite facies. The higher radioactivity observed is probably due to the presence of accessory minerals, as allanite, that have been observed in some cases.

CONCLUSIONS

The main results are the following:

i) The pure hornblende schists from Kolar gold fields have uniform uranium and thorium contents in the samples studied. They have normal Th/U ratios – mean value of 3.5.

ii) The altered schists in contact with the reef show a large spread specially in their Th/U ratios.

iii) The gold bearing auriferous quartz veins show some evidence of secondary processes leading to oxidation of uranium to the hexavalent state and the removal of the soluble uranyl ion UO_2^{2+}.

iv) The results on vein rocks and pegmatites confirm the general fractionation trend for U and Th to increase with differentiation.

v) The results on some charnockites from different localities do not generally show the expected depletion of these elements in such granulite facies rocks, probably due to the presence of accessory minerals, as allanite that have been observed in some cases.

AKNOWLEDGMENTS

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