DESIGN OF A SIMPLE URANIUM-THORIUM DISCRIMINATOR FOR WEAKLY ACTIVE ORES CONTAINING LESS THAN 1% U_3O_8 EQUIVALENT

Part I.—Study of the Self-Absorption of the β and γ Rays in Relation to Optimum Sample Thickness and Discrimination

KHURSHID HUSAIN, MOHAMMAD AZEEM,* NURUL AHMED AND MAZHAR MAHMOOD QURASHI

Central Laboratories, Pakistan Council of Scientific and Industrial Research, Karachi

1. Introduction

In the initial stages of the radioactive assay of mineral samples, the two most important questions to be answered are (a) what is the major radioactive constituent and (b) what is its percentage concentration in the sample. For relatively rich ores, a semi-quantitative chemical assay can be carried out, but these methods are always tedious and, for concentrations of less than 2 or 3%, precise results are often difficult to obtain due to interference from silica and iron.[1] Therefore, it is of great practical utility to develop physical methods of analysis. One of the most useful methods uses the characteristic X-rays excited in the elements composing the mineral. This method however suffers from two drawbacks, (a) the lower limit to the sensitivity is in the region of 0.05 to 0.1% uranium or thorium, and (b) interference from the K-lines of rubidium, yttrium, and zirconium (for uranium) and rubidium and bromine (for thorium) is serious if any of these elements is present in quantities greater than 10%. A method involving the estimation of radon,[1] and two methods using electronic discrimination[2] between (a) particle energy and (b) the half life of radium C′ and thorium C′, have been suggested but are rather complicated in application.

A powerful and accurate method is that of β-ray spectorgraphy, by which the energies of the γ-spectra of the active nucleii can be measured and thence the elements identified. The apparatus is elaborate, and quantitative data are obtainable after considerable time and trouble. The discrimination obtained is very high, because no two elements give the same γ-ray spectrum, but such high discrimination is not essential for the survey of minerals, because there one is looking primarily for uranium and thorium, while interference can be expected from the radioactive isotope of potassium.

2. Principle of the Simple Method

By sacrificing the excessive discrimination of the β-ray spectrograph, and reducing the examination to a simple analysis by absorption of the emitted β and γ-rays, we obtain a simple means of identifiying thorium and uranium, and of estimating their relative concentrations. Ideally, if potassium is assumed to be absent, two measurements of the total $(\beta + \gamma)$ counts through two suitable thicknesses of absorbing matter should provide the necessary discrimination between thorium and uranium. This technique is already employed in a type of discriminator and gives satisfactory results for rich ores. However, for the poor ores, the absorption in the samples itself becomes considerable, and must be allowed for.

The present development consists in making use of the mineral sample as its own absorber and in selecting the two thicknesses so that the smaller thickness, d_β, is of the order of the β range, and the larger thickness, d_γ is of the order of the γ-range in the powdered mineral. This has the advantage of making the ratio n_γ/n_β of the γ to the β counts more favourable for accurate measurement and discrimination than the ratio of γ counts through two different thicknesses suggested by Behounek.[1] Figure 1 shows two possible

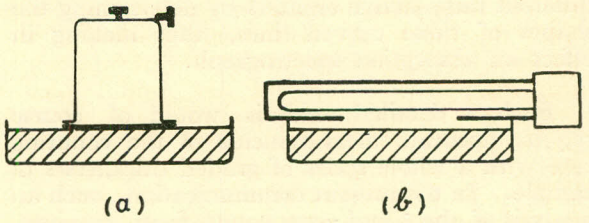


Fig. 1.—Two possible arrangements for investigating the variation of the counting rates with sample thickness using (a) an end window counter (left) and (b) a tube counter (right).

experimental arrangements, where either the end-window or the tube type of Geiger Muller counter measures the total counts due to the β and γ radiations from any one thickness of sample. If a substance gave off β-rays only, the plot of the counting rate n_β against thickness 'd' would be somewhat as in Fig. 2 (a), but when only γ-rays are given off, the curve is as shown in Fig. 2(b)

* Present address: Atomic Energy Commission, Karachi.

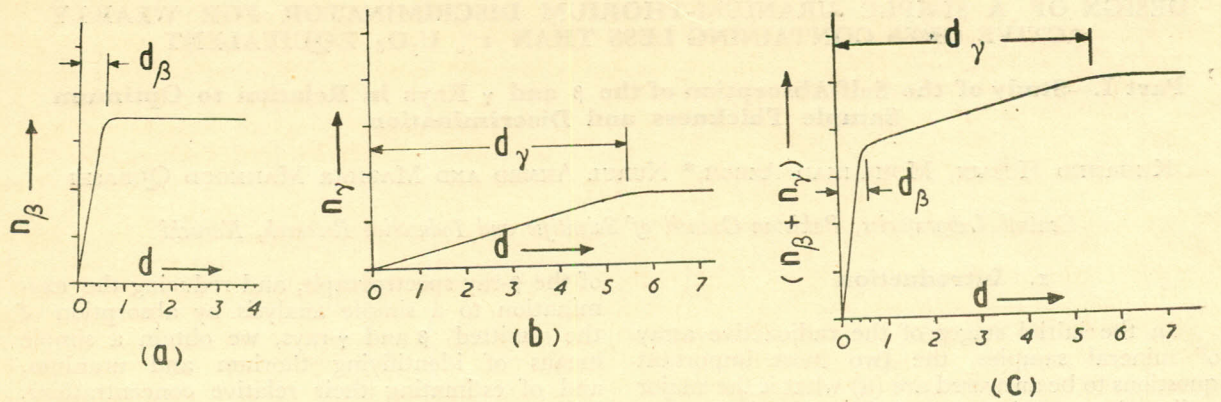


Fig. 2.—Idealized representation of variation of counting rate with sample thickness for (a) unique energy beta emitter, (b) unique energy gamma emitter, and (c) combination of both emissions.

where d_β and d_γ may be called the ranges of the β and γ-rays, respectively . When both these emissions are combined, we get Fig. 2 (c). For a unique energy of the β-rays and a simple γ-ray line, the plot should consist of two inclined straight lines, the first and steep one corresponding essentially to the β-rays and the second one to the γ-rays. The bend between the two occurs at a thickness corresponding to the range of the β-ray. The second part of this graph reaches saturation at a much larger value of thickness, corresponding to the large range of the γ rays. Since the actual β and γ-ray spectra of radioactive substances are complex, especialy when accompanied by decay products, each of the two straight lines of Fig. 2(c) will be slightly curved, being in fact made up of a series of lines, each terminating at the vlaue of d corresponding to the range of a particular group of β or γ-rays. This presents the possibility of a definite discrimination between different radio-active emitters by determining the shapes of these curved lines, thus making in effect an absorption spectrograph.

Such a detailed analysis would of course require accurate measurements of the counting rate with a whole series of graded thicknesses of sample. In the present communication , such an analysis of the β and γ-ray counts from uranium, thorium, and potassium is presented and the necessary conditions are determined for (a) efficient discrimination between them, and (b) accurate estimation of the quantities of the three metals in any given sample. On the basis of these results, the essential features of a combined discriminator-estimator are worked out, detailed tests on which form the subject matter of Part II of this paper.

3. Experimental Details

(a) *Preparation of Standards.*—The standard samples were prepared by successive dilution of the active material with dry calcium carbonate. The dilutions were carried out in stages, with thorough mixing at each stage, the concentration being reduced by a factor of 5 to 10 in each stage. The four standards used contained : (i) 0.060% of uranium in equilibrium (from analysed low-grade pitchblende), (ii) 0.50% thorium nitrate (prepared about five years ago), (iii) 30% potassium chloride (*i.e.*, 16% potassium) in calcium carbonate, and (iv) 0.25$_5$% of uranium in the form of chemically separated uranyl nitrate. The fourth standard was used to study the effect of lack of equilibrium in the uranium series.

(b) *Measurement of Counting Rate.*—Various quantities of the material under examination were taken in a 9 cm. diameter flat-bottomed petri dish, across the top of which a Geiger Muller tube counter* was held, the surface of the tube being about 4 mm. above the top of the dish. The counting rate was measured by noting the number of counts recorded by a Dynatron Scalar Unit in ten to twenty successive intervals of one or two minutes. Since a suitable lead castle was not available, the back-ground rate of the counter had to be determined when similarly placed, but without any sample in the petri dish. This back-ground rate was measured at the commencement and also at regular intervals all through the experiment. By subtraction, the total $(\beta + \gamma)$ counts, $n\beta + n\gamma$, recorded by the counter for any one quantity of material in the petri dish were obtained. The mean mass per unit area of the sample was obtained from its weight and the diameter of the dish, and this could be correlated with the thickness, which was measured correct

* The tube counter was a Frieseke and Hoepfner beta-gamma Geiger Muller tube (No. FHZ 69) with an operating plateau voltage of 700 volts.

to 0.2 mm. with a travelling microscope. This correlation is shown in Table 1, which also gives the γ and β+γ counting rate for various thicknesses of the uranium (pitchblende) standard. Since the petri dish was only 1.5 cm. high, some modification was necessary for working with larger thicknesses of material, going upto 8 cm. A dish of adjustable height was improvized by surrounding the petri dish with a stout well-fitting cardboard cylinder 10 cm. in height. By supporting the dish on a wooden block and sliding the cardboard cylinder up and down, various thicknesses of material upto 8 cm. could be used satisfactorily. During these experiments, the counter tube was placed so that its lower surface was about 4 mm. above the (upper) flat surface of the material being tested, in which position the β-shield just rested on top of the cardboard cylinder.

Since the area of the petri dish and therefore of the cylinderical sample is fixed, the effect on the counter of the rays from a particular layer will depend on its distance, x, from the counter (Fig. 3). The variation with x is complex, and in order to minimize this variation, the slotted metal β-ray shield was retained on the counter tube, thus constituting an opening of fixed angular width for the β rays. For depths of several centimetres, this angular spread falls within the diameter of the petri dish, and therefore the correction for distance may be neglected.

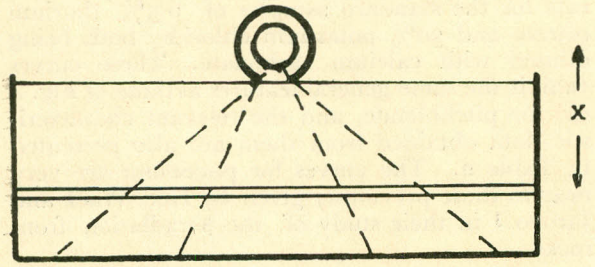


Fig. 3.—Schematic representation of the effect of counter shield with slotted opening.

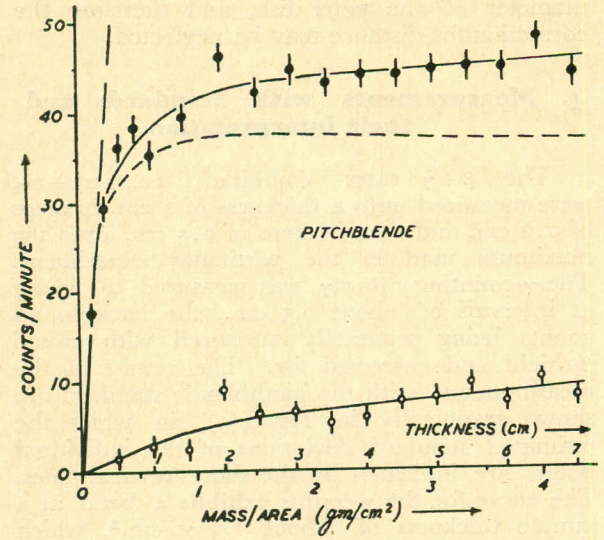
4. Measurements with Standards and their Interpretation

The β + γ rates combined, *i.e.*, $n\beta + n\gamma$, were measured upto a thickness of 1 cm. in steps of 0.2 cm. and then in steps of 0.5 cm. upto the maximum used in the particular experiment. The γ-counting rate $n\gamma$ was measured all along at intervals of about 0.5 cm., the background counts being separately measured with closed β-shield and corrected for. The results of the measurements with the picthblende standard are shown graphically in Fig. 4(a), in which the estimated standard deviations of the individual points are indicated by the short vertical lines. The curve for the γ-counts exhibits a bend at a sample thickness of about 1.5 g./cm.^{2}, which shows that the first part of this curve corresponds essentially to the relatively soft γ-rays (mostly internal-conversion K-series radiation), for which the mass absorption coefficient, μ/ρ, is known to be of the order of 0.7 cm.^{2}/g. in the diluting material used. The second, less steep, part of the curve naturally corresponds to the harder components of the nuclear γ-rays with $\mu/\rho \sim$ 0.1 cm.^{2}/g. in the diluting material, cf. Behounek.[1]

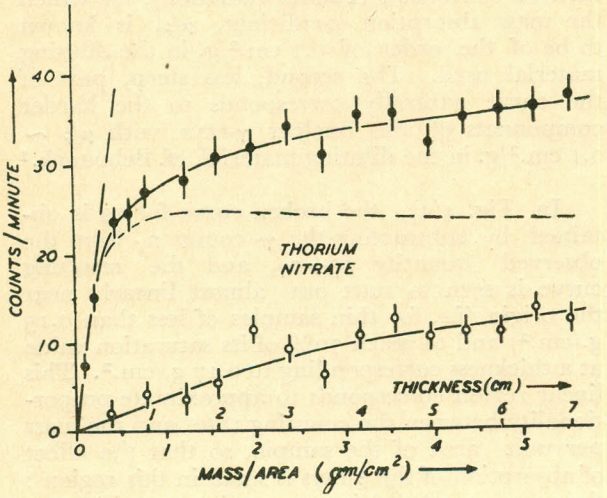
In Fig. 4(a), the broken curve for $n\beta$ is obtained by subtracting the γ- counts $n\gamma$ from the observed quantity $n\beta+n\gamma$, and the resulting curve is seen to start out almost linearly near the origin (*i.e.* for thin samples of less than 0.15 g./cm.^{2}) and to reach 70% of its saturation value at a thickness corresponding to 0.17 g./cm.^{2}. This linear region corresponds to approximate proportionality between the counting rate and the mass per unit area of the sample, so that the effect of absorption of the β rays is small in this region : thus at 0.17 g./cm.^{2} (corresponding to a thickness

TABLE 1.—β + γ AND γ COUNTING RATE FOR VARIOUS THICKNESSES OF 0.06% URANIUM (PITCHBLENDE) STANDARD.

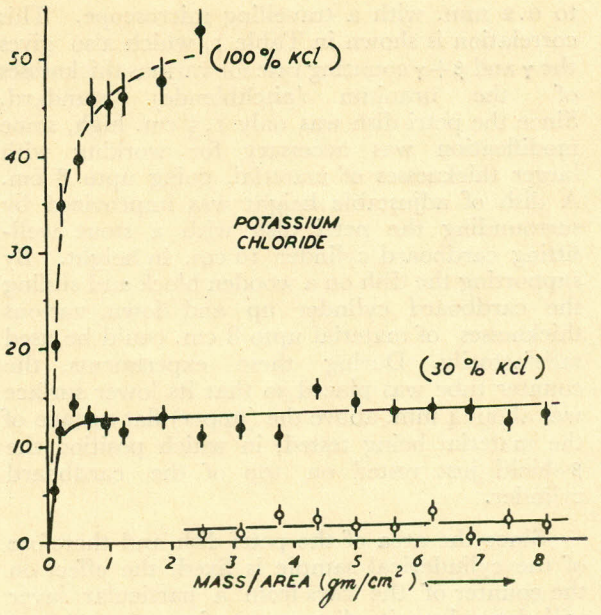
Approx. thickness (cm.) from microscope ..	0.20	0.40	0.60	0.80	1.00	1.50
Mass per unit area (g. sq. cm.)	0.095	0.216	0.347	0.476	0.627	0.891
Corrected thickness of sample (cm.)	0.16	0.36	0.57	0.79	1.03	1.47
$n\beta + n\gamma$ (counts/minute)	17.6	29.4	36.2	38.5	35.3	39.7
$n\gamma$ (counts/minute)	..	..	1.6	..	2.8	2.4



(a)



(b)



(c)

Fig. 4.—Experimental graphs showing total counting rate (solid circles), gamma counting rate (hollow circles) and beta counting rate (broken line) for the three standard samples (a) 0.06% uranium as pitchblende, (b) 0.5% thorium nitrate,(c) 30% potassium chloride, the broken line curve being for 100% KC1 in this case.

of nearly 0.3 cm.) the curve falls only about 15% below the tangent at the origin. For larger thicknesses, the absorption is so great that the number of β counts becomes constant and no longer increases with sample thickness. All the important characteristics of the β and γ counts from pitchblende as deduced from the curves of Fig 4(a) are collected in the first row of Table 2, in which all the figures have been reduced to 0.1% uranium content, *i.e.*, 1 mg. uranium in 1 g. of sample. The last column gives the estimated γ counting rate for a thickness of 8 g./cm.,² which should yield about 80% of the saturation γ counts.

Figures 4(b) and 4(c) show the counting rate for the standard samples of 0.5% thorium nitrate and 30% potassium chloride, both being diluted with calcium carbonate. These curves exhibit the same general features as those of Fig. 4 (a) for pitchblende, and the relevant characteristic data obtained from them are also presented in Table 2. The curves for potassium are very near to those previously given by Hee, Wack and Jarovo [3] in their study of the β–radiation from rocks.

It is to be noted that if an exponential law is assumed for the absorption of the complex β-ray spectrum, then the figures in columns 4 and 5 of Table 2 should be equal. In actual fact, the two values are seen to be in the ratio of 1.30 ± 0.03, thus showing that such a law is not a good approximation. The lack of decay equilibrium of the thorium nitrate standard requires comment here. One of the decay products of thorium, *viz.*, radio-thorium (half life=1.9 years) is isotopic with thorium and is therefore to be found in the equilibrium proportion in chemically separated thorium compounds. All the decay products of radiothorium have short half-lives, the longest being 3.6 days for thorium X, so that these products will all be present in equilibrium with the radiothorium after a lapse of 10 days or so. The total activity of the thorium compound at any time can thus be estimated from the decay of

radiothorium and the formation of mesothorium 1 (half-life=0.7 years) and mesothoruim 2 (half-life=6 hours), which are products intermediate between radiothorium and thorium (half-life $=10^{10}$ years). The detailed estimates are given in Appendix I, and the conclusion is that the ratio, R, of the activity of a certain quantity of thorium as the chemical compound to that of the same quantity of thorium together with all its products in equilibrium is

$$R_\beta = 0.56 \pm 0.08 \quad \text{for the } \beta\text{-activity}$$

and $$R_\gamma = 0.65 \pm 0.06 \quad \text{for the } \gamma\text{-activity}$$

over the first 10 years. Since the age of the thorium nitrate used was known to be of the order of 3 to 7 years, the β and γ measurements made with it can be approximately corrected for lack of equilibrium by dividing by these factors, R_β and R_γ, respectively. The essential adequacy of this correction is confirmed by the fact that the corrected β-counts for thorium in equilibrium (columns 2 and 3 of Table 2) are found to be nearly two sevenths of the corresponding counts for pitchblende, *i.e.*, in the inverse ratio of the half-lives of thorium and uranium.

5. Analysis of Results

We can now analyze Table 2 to determine what quantitites to use for the most satisfactory discrimination between thorium and uranium. The four possible quantities are the following ratios : R_1 = (saturation β-counts /origin β + γ slope), R_2 = (origin γ slope/origin β + γ slope), R_3 = (γ slope at 4 g./cm.²/origin β + γ slope) and R_4=(γ counts at 8 g. per cm.²/saturation β counts). Values of these ratios for the measured standards are collected in Table 3 below, which shows that all of them do give a certain measure of discrimination. The third row of figures in Table 3 are the differences, $(R_{Th}-R_U)$, between corresponding ratios for thorium and uranium, together with the standard error of measurement of this difference with the apparatus previously described. Examination of these figures brings out the fact that the first ratio (R_1) will give very poor discrimination, and can be used at best as a confirmatory measurement. The other three ratios, all of which make a comparison between the γ and β counts, will give good discrimination, because the standard error of the measurement is one fifth to one tenth of the corresponding value of $(R_{Th}-R_U)$.

The best discrimination is given by R_3 but this requires accurate measurements of γ-counts for several sample thicknesses in the neighbourhood of 4 g./cm.². R_4 is second best, but has the great advantage that only two observations are required, one of the γ counts for a thickness of 8 g./cm.², and the other of the saturation β counts. R_2 ranks with R_4 in discriminating efficiency, but sufferes from the same drawback as R_3. It appears therefore that R_4 is the best discriminating ratio for the present purpose.

6. Estimation of the Uranium to Thorium Ratio

Since the measured value of any one of the above discriminating ratios, R_2, R_3, R_4, for a sample containing both uranium and thorium (in equilibrium with their decay products) will be intermediate between the values for uranium and thorium given in Table 3, it follows that either of these three ratios can be used to determine the relative proportions of uranium and thorium in the sample. The expected values of R_2, R_3 and R_4 for various ratios of uranium to (thorium + uranium) can be easily calculated by simple proportion using the data of Tables 2 and 3, and the results are shown in Table 4 and graphically in Fig. 5(a). All the graphs of this figure are

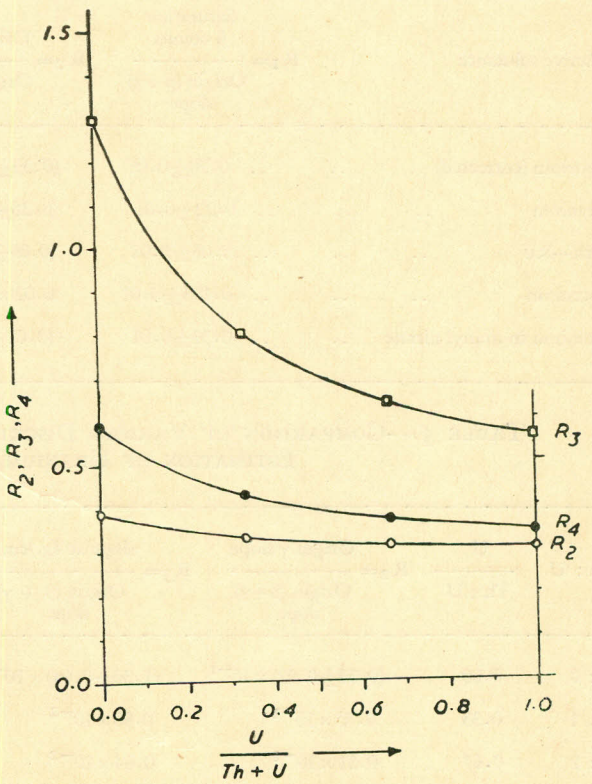


Fig. 5(a).—Graphs showing the values of the three best discriminating ratios, 10 R_2, 100 R_3, R_4, for different ratios of uranium/uranium + thorium).

TABLE 2.—SYNOPSIS OF MEASUREMENTS ON STANDARDS (REDUCED TO 1% OF ACTIVE SUBSTANCE).

Active substance	(β + γ) Counts				γ- Counts		
	Origin slope, counts per 1 g./cm.^{2}	Saturation β counts	Mass/area for 70% of saturation	Saturation β counts / Origin β slope	Origin slope, counts per 1 g./cm.^{2}	Slope at 4 g./cm.^{2}	Estimated** γ counts for a thickness of 8 g./cm.^{2}
Uranium (in pitchblende*) ..	280±10	62.4±1.0	0.17	0.23	8.3_3±0.2	1.58±0.15	21±1
Thorium (in thorium nitrate). ..	38±0.5	11.5±0.2	0.25	0.32	1.7_1±0.1	0.57±0.3	..
Thorium corrected to equilibrium ..	67.8	20.5	0.25	0.32	2.63	0.88	12±1
Potassium	(410±20)10^{-3}	(80±4) $\times 10^{-3}$	0.16	0.19_5	..	1×10^{-3}	$\sim 10 \times 10^{-3}$
Uranium (in uranyl nitrate) ..	184±5	64.3±0.9	0.26	0.35	0.4±0.2	0.4±0.2	..

* Some slight lack of equilibrium was suspected in the pitchblende sample, and it is possible that a correction of about 10% may be required for the experimental values.

** The estimate is a valid extrapolation because the value of μ/ρ is less than 0.1 for the harder group.

TABLE 3.—COMPARATIVE STUDY OF THE POSSIBLE DISCRIMINATING RATIOS.

Active substance	$R_1 = \frac{\text{Saturation } \beta \text{ counts}}{\text{Origin } (\beta+\gamma) \text{ slope}}$	$R_2 = \frac{\text{Origin } \gamma \text{ slope}}{\text{Origin } (\beta+\gamma) \text{ slope}}$	$R_3 = \frac{\gamma \text{ slope at 4g./cm}^2}{3 \text{ Origin } (\beta+\gamma) \text{ slope}}$	$R_4 = \frac{\gamma \text{counts at 8 g./cm}^2}{\text{Saturation } \beta \text{ counts}}$
Thorium (corrected)	0.30±0.01	(0.39±0.02) $\times 10^{-1}$	(1.30±0.04) $\times 10^{-2}$	0.59±0.05
Uranium	0.22±0.01	(0.30±0.01) $\times 10^{-1}$	(0.56±0.02) $\times 10^{-2}$	0.34±0.02
R_{Th}—R_U	0.08±0.02	(0.09±0.02) $\times 10^{-1}$	(0.74±0.04) $\times 10^{-2}$	0.25±0.05
Potassium	0.195±0.01	0.02×10^{-1}	(0.24±0.05) $\times 10^{-2}$	0.1
Uranium in uranyl nitrate	0.35±0.01	(0.02±0.01) $\times 10^{-1}$	(0.22±0.1) $\times 10^{-2}$	..

TABLE 4.—COMPARISON OF VARIOUS DISCRIMINATING RATIOS FOR THE QUANTITATIVE ESTIMATION OF URANIUM/(THORIUM + URANIUM).

Th : U	$\frac{U}{Th+U}$	$R_2 = \frac{\text{Origin } \gamma \text{ slope}}{\text{Origin } (\beta+\gamma) \text{ slope}}$	$R_3 = \frac{\text{slope at 4g./cm.}}{\text{Origin } (\beta+\gamma) \text{ slope}}$	$R_4 = \frac{\text{8g./cm. } \gamma \text{ counts}}{\text{Saturation } \beta \text{ counts}}$	$\frac{0.01}{R_2}$	$\frac{0.01}{R_3}$	$\frac{1}{R_4}$
3: 0	0.00	(0.39±0.02) $\times 10^{-1}$	(1.30±0.04) $\times 10^{-2}$	0.59±0.05	2.56±0.1	0.77±0.02	1.71±0.1
2: 1	0.33	0.33×10^{-1}	0.80×10^{-2}	0.43	3.06	1.25	2.30
1: 2	0.67	0.31×10^{-1}	0.64×10^{-2}	0.37	3.25	1.55	2.69
0: 3	1.00	(0.30±0.01) $\times 10^{-1}$	(0.56±0.02) $\times 10^{-2}$	0.34±0.02	3.36±0.1	1.77±0.05	2.97±0.2

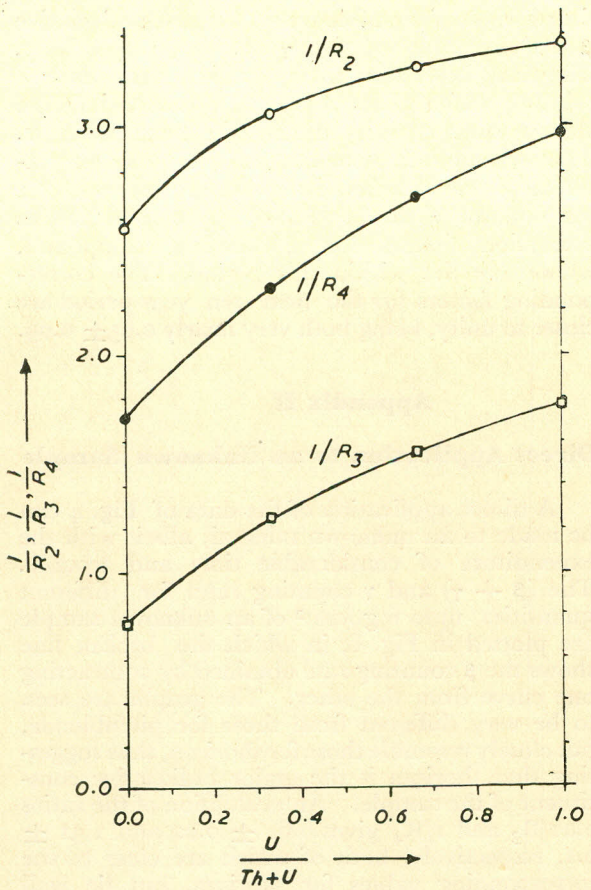


Fig. 5(b).—The corresponding graphs for the reciprocals of these ratios, viz. $0.1/R_2$, $0.01/R_3$ and $1/R_4$, showing the improved character of the curves.

seen to be strongly curved, especially that for R_3, and are therefore rather unsuitable for quantitative work. This defect can be easily remedied in the case of R_3 and R_4 by plotting $0.01/R_3$ and $1/R_4$ against relative percentage of uranium to (thorium+uranium), which gives the much improved graphs shown in Fig. 5(b). Combining this with the considerations of the previous section, we find that $1/R_4$ is the best discriminating ratio. If we assume an accuracy of 5% in the γ-counting rate, which is readily attained, we get a margin of about 0.1 in the measured value of $1/R_4$. It follows from Fig. 5(b) that we can determine the relative proportion of uranium and thorium to within 5% with this type of measurement. The design and testing of a discriminator based on the foregoing analysis will be described in a later communication, while Appendix II gives an example of the application of the detailed analysis along the patterns of Fig. 4.

As regards the influence of potassium on the counting rates, it is enough to note that both the β and γ counts for potassium are about 1/1000 of those obtained with corresponding quantities of uranium. Thus 10% of potassium in the sample will be equivalent to about 0.01% of uranium, and its presence will not therefore produce significant error unless the (uranium + thorium) content of the sample is as low as 0.1%. For such extremely poor ores, a correction for the activity of potassium can be readily applied using the data of Table 2, provided the potassium content is known approximately from a chemical determination.

Acknowledgements

The authors are grateful to Dr. Nazir Ahmad, Chairman, Atomic Energy Commission, for his interest in the work, and to Mr. Abdul Hai for carrying out some of the chemical analyses.

References

1. F. Behounek, Collection Czech Chem. Communs., **15**, 699 (1950).
2. D. H. Peirson, Proc. Phys. Soc. (London), **64B**, 876 (1951).
3. A. Hee, M. Wack and M. Jarovo, Ann. geophys. **8**, 323 (1952).

Appendix I

Estimate of the Activity of Thorium Nitrate Preparation

(a) Members of the thorium series isotopic with thorium and therefore initially present are : (1) thorium (half-life $= 10^{10}$ years) and (2) radiothorium (half-life = 1.9 years).

(b) The intermediate products are mesothorium 1 (half-life = 6.7 years) followed by mesothorium 2 (half-life = 6.1 hours).

(c) All the other products, *i.e.*, those after radiothorium have short half-lives with a maximum of 3.6 days for thorium X.

Thus the activity A (t) at any time t ($\gg$ 3.6 days) can be represented by

$A(t) = A_{Th} + (A_{Mes_1} + A_{Mes_2}) + (A_{Radth} + A(\text{of decay products}))$

$= A_{Th} + \text{Constant} \times A_{Mes_1} + \text{Constant} \times A_{Radth}$. Since thorium itself is only α-active, the problem reduces to estimating the relative activites of radiothorium and mesothorium 1, with which mesothorium 2 will be in equilibrium after a period exceeding a day. Let the number

of atoms of thorium disintegrating per second be N_0. Then

$A_{Mes_I} = N_I$ = number of atoms of mesothorium disintegrating per second.

$$= N_0 (1\text{-}\exp [-\lambda_I t])$$

where $\lambda_I = 0.693/6{,}7$ year^{-1},

and $A_{Radth} = N_2$ = number of atoms of radiothorium disintegrating per second.

$$= N_0 \exp [-\lambda_2 t] + N_0 (1 - \exp [-\lambda_1 t])$$

$$= N_0 (1 - \exp [-\lambda_I t] (1 - \exp [(-\lambda_2 - \lambda_I)t])).$$

Table 5 gives the values of N_I/N_0 and N_2/N_0 for various values of time elapsed since the separation of the thorium. Now there are two β-emitters among radiothorium and its equilibrium products, and two among mesothorium and its products. On the assumption of similar spectra for these four emitters, the β-activity of the thorium should be proportional to

$$\frac{1}{4}\left(2\frac{N_I}{N_0} + 2\frac{N_2}{N_0}\right) = \frac{1}{2}\left(\frac{N_I}{N_0} + \frac{N_2}{N_0}\right).$$

By a similar reasoning, the γ-activity should be proportional to

$$\left(\frac{3}{4}\frac{N_I}{N_0} + \frac{1}{4}\frac{N_2}{N_0}\right).$$

These two ratios are tabulated in the third and fourth rows of the table under the labels of relative β activity and relative γ activity, respectively. They are both seen to fall rapidly and reach a minimum after about 2 years, and then to increase slowly towards unity during the next 20 years. The average values of these ratios obtained graphically over the first ten years are given in the last column of the table and they can be used as correction factors for a thorium compound, whose age lies within this period. The corresponding factors for the next ten year period are closer to unity, being both very nearly 0.85 ± 0.09.

Appendix II

Direct Application to an Unknown Sample

A direct application of the data of Fig. 4 can be made to an unknown mineral, albeit with the expenditure of considerable time and trouble. The $(\beta + \gamma)$ and γ counting rates for different quantities upto 6 g./cm.^{2} of an unknown sample are plotted in Fig. 6, in which the broken line shows the β counting rate obtained by subtracting one curve from the other. The graphs are seen to be very different from those for pitchblende, but closely resemble those for thorium, thus suggesting that thorium is the major radioactive constituent of the sample. An evaluation of the ratios $0.01/R_3$ and $1/R_4$ gives 0.61 ± 0.02 and 1.31 ± 0.1, respectively, both of which are close to the corresponding values for thorium, but lie well outside the range for thorium-uranium mixtures (Table 4 and Fig. 5). Thus there would appear to be no possibility of the presence of any uranium in the sample.

TABLE 5

Time (years)	0	1	2	4	6	8	10	15	20	Mean for first 10 years
N_I/N_0	1.000	0.834	0.714	0.643	0.659	0.706	0.759	0.866	0.930	
N_2/N_0	0.000	0.120	0.232	0.410	0.547	0.652	0.733	0.862	0.929	
Estimated relative β—activity	0.500	0.477	0.473	0.526	0.603	0.679	0.746	0.864	0.930	$0.56_5 \pm 0.08$
Estimated relative γ-activity	0.750	0.655	0.594	0.585	0.631	0.692	0.752	0.865	0.930	$0.65_2 \pm 0.06$
Relative (γ/β) .. .. ratio	1.50	1.37	1.26	1.11	1.05	1.02	1.01	1.00	1.00	1.15 ± 0.2

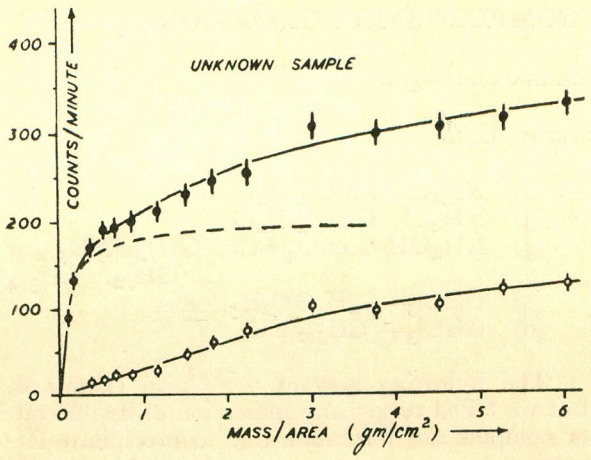


Fig. 6.—The gamma, beta, and total counting rates for various thicknesses of an unknown sample. Total rate, solid circles; gamms rate, hollow circles; beta rate, broken line.

Assuming first that all the activity is due to thorium, we get from the measured β and γ counting rates the following estimates of thorium content :

β Counting gives	..	1.10±0.1 %
γ Counting gives	..	1.40±0.1 %

The discrepancy between the two estimates points to the presence of some other constituent, which has a higher γ/β emission ratio than thorium. This idea was confirmed by the chemical analysis of the sample, which gave a thorium content of only 0.6±0.05%. This would account for a trifle more than half the β-counts and less than half of the γ-counts. The counts remaining unaccounted for are tabulated below :

Origin β+γ slope	Saturation β counts	Origin γ slope	γ slope at 4 g./cm.²	γ counts at 8 g./cm.²
400	70	16	9	70

Such a distribution of β and γ counts could be due to the decay products of uranium that have been separated from the parent material by weathering action. These decay products are known to be strongly γ-active, cf. for instance the various counting rates for uranium in pitchblende and in uranyl acetate given in Table 2. On this basis, the above residual activity would correspond to an original uranium content of about 0.4%.