

# A Trial Method of Determining Uranium and Thorium Content in Rocks by Means of an Alpha-ray Scintillation Counter and a Radioscope

By

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## **Abstract**

The determination of the content of uranium, thorium and potassium in rocks is a matter of great concern for the geologist. The usual method, using the gamma-ray spectrometer, requires not only relatively expensive apparatus, but also considerable time. This is especially true when the samples have a feeble radioactivity, such as in rocks. In this paper a new approach is described. In this approach the content of uranium and thorium can be obtained by means of the parallel measurements with an alpha-scintillation counter and a radioscope.

## **Introduction**

Up to the present, the radioactivity distribution in granitic bodies has been investigated by the authors by making use of the radioscope with a Lauritsen element, designed specially for the purpose of measuring feeble radioactivity in pulverized rock samples.<sup>1),2)</sup> As a result, interesting information on the granite problem has been obtained.<sup>1),2),3),4)</sup> It is desired, however, to make clear to which nuclides these radioactivity distributions due.

Recently, an alpha-scintillation counter, fit for the measurement of radioactivity of pulverized rocks, has been designed by the authors and manufactured by the Nihon Musen Co., (Aloka ZD-3B). The total alpha particles emitted from the unit surface area of the sample per unit time can be calculated from the observed counting rate in the same way as HATUDA and NAGAI have done in the geological age determinations by the lead/alpha-ray method.<sup>5)</sup> In this reduction, the absorption of alpha-rays in the body of

the sample itself, the air intervening between the scintillator and the sample, and also the thin aluminium foil which serves as a cover protecting the scintillator against contamination, are taken into consideration. It was found that by combining the results obtained by this scintillation counter and the radioscope, the uranium and thorium content of a rock sample can be obtained. It is a reasonable assumption that the contribution of potassium to the ionization within the radioscope is negligible as compared with the ionization due to thorium and uranium.

### Description of the alpha-scintillation counter

The alpha-scintillation counter mentioned above is shown in Fig.1. It has

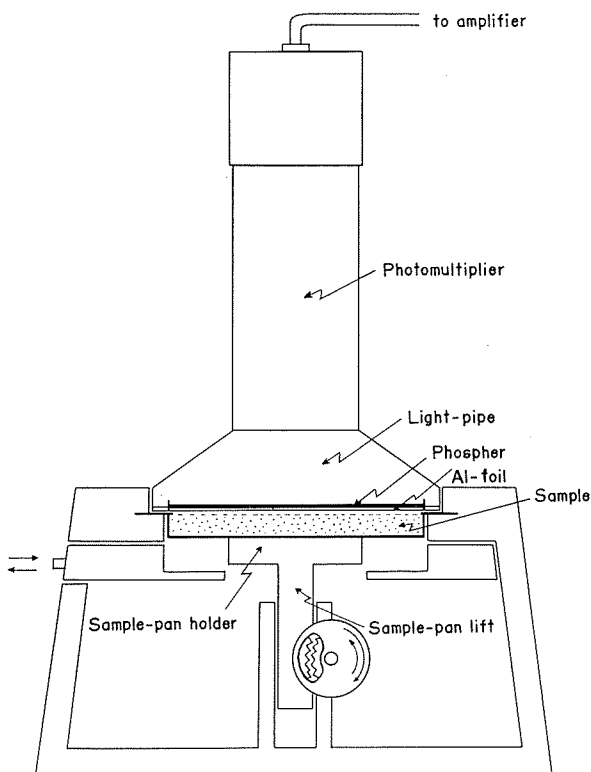


Fig. 1 Alpha-ray scintillation counter.

a phosphor of multi-crystalline ZnS(Ag) powder deposited on a transparent plastic circular disk of 11.8 cm in diameter, backed with a conical light-pipe of lucite, 2.5 cm in height, surrounded by light reflector as a light guide. The other end of the light-pipe is closely attached to the face of the  $1\frac{1}{2}$  " photo-multiplier (Du-Mont 6292), on the back side of which is deposited the photo-cathode.

The alpha-particle to be detected produces a flash of light on the phosphor screen. This faint light is transmitted efficiently to the photo-cathode of the multiplier tube, and the photo-electrons produced at the photo-cathode, in turn, are multiplied about  $10^6$

times as a current by means of 10 dynodes in the tube. The current pulse is then supplied to the input of the scaler endowed with the discriminator

(VS-105 III, mfd. by Tokyo Atomic Industrial Co., Ltd.) through the pre-amplifier (Aloka, PAM-7). By making use of the dual timer (TM 1A), the counting rate is obtained by measurement of at least 1000 counts per sample.

### Principle of alpha-particle counting with a scintillation counter

An alpha-particle is emitted from the nucleus of the alpha-rayer without fail as it disintegrates into the next daughter element, so that alpha-particle emission is equal to the disintegration of alpha-rayers in number. Of the emitted alpha-particles only a portion can attack the surface of the scintillator with a sufficient energy to give the count. The number of alpha-particles countable depends upon some factors, such as (i) geometrical relation between alpha-ray source and receiver, including space-angle relation and absorptions by layer of air and also by metal foil absorber, if they exist, (ii) absorption in the body of the sample itself, i.e., so-called "self-absorption", and (iii) sensitivity of the detector. The effect due to back-scattering is negligible for alpha-particles and counting loss due to recovery time of the scintillation counter is also negligible for a weak radioactive source such as rocks.

The number of alpha-particles actually to be counted can be expressed in the similar way obtained by one of the authors (H) and NAGAI<sup>5)</sup> as

$$n = \frac{K\mu}{4} \left\{ N_{UI} \sum_{UI} \frac{(R_{UI} - \rho - a)^2}{R_{UI} - \rho} + N_{AcU} \sum_{AcU} \frac{(R_{AcU} - \rho - a)^2}{R_{AcU} - \rho} + N_{Th} \sum_{Th} \frac{(R_{Th} - \rho - a)^2}{R_{Th} - \rho} \right\} \quad (1)$$

where,  $n$  is expressed as counts per sec. per cm<sup>2</sup>;  $\mu$  is the ratio of range of alpha-particle in sample to that in air;  $R_{UI}$  etc. are ranges of alpha-particles from each members of uranium series, etc.;  $\rho$  is the minimum energy expressed in terms of the range (air-cm), which can excite the scintillator to cause a count;  $a$  is the thickness (cm in air-equivalent) of the absorber foil plus the layer of air intervening between the sample and the scintillator; and  $N_{UI}$  etc. are numbers of atoms of the each parent element  $UI$  etc. decaying per cm<sup>3</sup> per second. The summation is made for each alpha-rays from each series, respectively.

Now that,

$$N_{UI} = 8 \cdot 6.06 \cdot 10^{23} \cdot \lambda_{UI} \cdot UI \cdot d / W_{UI}$$

$$N_{AcU} = 7 \cdot 6.06 \cdot 10^{23} \cdot \lambda_{AcU} \cdot AcU \cdot d / W_{AcU}$$

$$N_{Th} = 6 \cdot 6.06 \cdot 10^{23} \cdot \lambda_{Th} \cdot Th \cdot d / W_{Th}$$

where  $W$ 's are atomic weights;  $UI$ ,  $AcU$  and  $Th$  are content in grams of the respective elements;  $d$  is density of the sample and  $\lambda$ 's are decay constants which are

$$\lambda_{UI} = 4.86 \cdot 10^{-18} \text{ sec.}^{-1}$$

$$\lambda_{AcU} = 3.11 \cdot 10^{-17} \text{ sec.}^{-1}$$

$$\lambda_{Th} = 1.58 \cdot 10^{-18} \text{ sec.}^{-1}$$

Using these relations, the equation (1) becomes:

$$n = K \mu d \cdot 10^3 \left[ UI \left\{ 3.11 \sum_{UI} \frac{(R_{UI} - \rho - a)^2}{R_{UI} - \rho} + 0.14 \sum_{AcU} \frac{(R_{AcU} - \rho - a)^2}{R_{AcU} - \rho} \right\} + 1.03 Th \cdot \sum_{Th} \frac{(R_{Th} - \rho - a)^2}{R_{Th} - \rho} \right] \dots\dots\dots (2)$$

(i) Calculation of  $\mu d$  (absorption factor):

If the permeability of the medium to alpha-particles defined in terms of its atomic composition is denoted by  $\psi$ , we have by the Bragg-Kleeman rule,

$$\psi = a \sqrt{W_A} + b \sqrt{W_B} + c \sqrt{W_C} + \dots\dots\dots (3)$$

where the medium is assumed to be composed of atoms,  $A, B, C, \dots\dots\dots$ , in the relative proportions by weight  $a, b, c, \dots\dots\dots$ . Thus the permeability of standard dry air is as tabulated:

| Atom     | Composition | $\sqrt{W}$ | Permeability         |
|----------|-------------|------------|----------------------|
| Oxygen   | 0.23024     | 4.000      | 0.921                |
| Nitrogen | 0.75539     | 3.742      | 2.827                |
| Argon    | 0.01437     | 6.324      | 0.091                |
|          | 1.00000     |            | $\psi_{air} = 3.839$ |

The density of dry air at NTP is 0.001226 g per ml, therefore

$$\mu = \frac{R_s}{R_a} = \frac{d_a \psi_s}{d_s \psi_a} = \frac{0.001226}{3.839} \cdot \frac{\psi_s}{d_s} \dots\dots\dots (4)$$

the suffixes  $a, s$  showing air and sample respectively, or

$$\mu d = 3.194 \cdot 10^{-4} \cdot \psi_s \dots\dots\dots (4')$$

where  $d_s$  is simply denoted by  $d$ .

For the available rocks of the known chemical composition shown in Table 1, similar calculations were made to find the permeability  $\psi$ , and the results as well as  $\mu d$  are shown in Table 2.

Table 1. Chemical compositions of the rocks in the calculation

|                                | NBS Milford<br>granite | Rokkô<br>granite | Tanakami<br>granite | Hira<br>granite | Hiei<br>granite | Ryôke<br>granite | NBS Deccan<br>Trap |
|--------------------------------|------------------------|------------------|---------------------|-----------------|-----------------|------------------|--------------------|
| SiO <sub>2</sub>               | 80.57%                 | 74.07%           | 75.28%              | 74.17%          | 69.14%          | 65.42%           | 49.72%             |
| Al <sub>2</sub> O <sub>3</sub> | 10.02                  | 12.56            | 13.06               | 13.14           | 15.65           | 18.57            | 13.41              |
| Fe <sub>2</sub> O <sub>3</sub> | 0.22                   | 2.98             | 0.38                | 0.62            | 1.04            | 1.32             | 5.53               |
| FeO                            | 1.32                   | .....            | 0.50                | 1.42            | 1.87            | 1.54             | 9.49               |
| MgO                            | 0.12                   | 0.45             | 0.48                | 0.25            | 0.34            | 0.60             | 5.64               |

|                               |       |       |        |        |        |       |       |
|-------------------------------|-------|-------|--------|--------|--------|-------|-------|
| CaO                           | 0.56  | 1.09  | 0.97   | 1.66   | 3.08   | 2.48  | 9.11  |
| Na <sub>2</sub> O             | 2.62  | 3.98  | 4.35   | 3.84   | 4.71   | 6.34  | 2.29  |
| K <sub>2</sub> O              | 4.00  | 4.32  | 4.52   | 4.34   | 3.38   | 2.74  | 0.78  |
| H <sub>2</sub> O              | 0.13  | ..... | 0.54   | 0.06   | 0.60   | 0.53  | 1.20  |
| CO <sub>2</sub>               | 0.03  | ..... | .....  | .....  | .....  | ..... | 0.27  |
| TiO <sub>2</sub>              | 0.11  | ..... | 0.07   | 0.31   | 0.35   | 0.15  | 2.63  |
| P <sub>2</sub> O <sub>5</sub> | 0.01  | ..... | 0.19   | 0.02   | 0.06   | ..... | 0.27  |
| MnO                           | 0.06  | 0.09  | .....  | 0.01   | 0.03   | 0.07  | 0.22  |
| Total                         | 99.81 | 99.54 | 100.34 | 100.65 | 100.25 | 99.76 | 99.91 |

Table 2. Permeability  $\psi$  and the values of  $\mu d$  of several rocks

|                     | $\psi$ | $\mu d$               |                                        |
|---------------------|--------|-----------------------|----------------------------------------|
| NBS Milford granite | 4.69   | $1.50 \times 10^{-3}$ | mean: $(1.50 \pm 0.01) \times 10^{-3}$ |
| Rokkô granite       | 4.69   | 1.59                  |                                        |
| Tanakami granite    | 4.55   | 1.45                  |                                        |
| Hira granite        | 4.75   | 1.52                  |                                        |
| Hiei granite        | 4.75   | 1.52                  |                                        |
| Ryôke granite       | 4.65   | 1.49                  |                                        |
| NBS Deccan trap     | 5.02   | 1.60                  |                                        |

The values of  $\mu d$  for the granites are very close, so that  $1.50 \times 10^{-3}$  is adopted tentatively as value of  $\mu d$  for granite samples.

(ii) Determination of  $\rho$ ,  $\alpha$  and  $K$  :

For the determination of  $\rho$  and  $\alpha$ , alpha-ray countings were carried out with the pulverized borax glass containing chemically pure  $U_3O_8$   $7.53 \times 10^{-3}$  g/g used as a "thick source" just as the same manner described by HATUDA and NAGAI (*loc. cit.*).

Fig. 2 shows the results of the pulse-height analyses with the uranium standard mentioned above and thorium standard, the pulverized borax glass fused with allanite from Kitashirakawa, Kyoto, containing  $6.05 \times 10^{-4}$  gTh/g. The former was proved to be suitable for the purpose of the present experiment.

Alpha-ray countings have been made with different number of sheets of the absorber Al-foil of thickness  $3.4 \mu$  and the ratios of counts per unit time with the combination of different number ( $m, n$ ) of sheets of the absorber were obtained by the following equation.

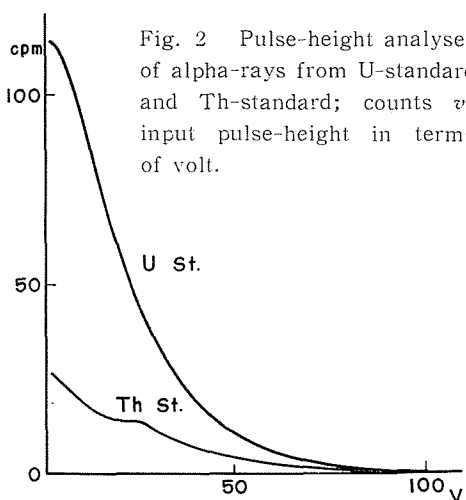


Fig. 2 Pulse-height analyses of alpha-rays from U-standard and Th-standard; counts vs input pulse-height in terms of volt.

$$\begin{aligned}\frac{n_m}{n_n} &= \frac{\frac{(R_{UI}-\rho-ma)^2}{R_{UI}-\rho}}{\frac{(R_{UI}-\rho-na)^2}{R_{UI}-\rho}} + \frac{\frac{(R_{UII}-\rho-ma)^2}{R_{UII}-\rho}}{\frac{(R_{UII}-\rho-na)^2}{R_{UII}-\rho}} \\ &= \frac{(R_{UI}-\rho-ma)^2}{(R_{UI}-\rho-na)^2} + \frac{(R_{UII}-\rho-ma)^2}{(R_{UII}-\rho-na)^2}\end{aligned}$$

With the six observation equations for different values of  $\frac{n_m}{n_n}$  the most probable values of  $\rho$  and  $a$  were determined by means of the method of least squares, giving the following values:

$$\begin{aligned}a &= 0.78 \pm 0.02 \\ \rho &= 1.20 \pm 0.05 \quad (\text{in cm}).\end{aligned}$$

In the case of rock samples,  $a$  is reduced to 0.60, owing to the decrease in the thickness of air layer caused by the difference in bulk of the uranium standard and rock samples.

Assuming all the member elements of each series are in radioactive equilibrium, the summations in the equation (2) were calculated as shown in the following by using the values of  $\rho$  and  $a$  thus obtained, in which as the values of ranges of alpha-rays from each alpha-emitter, those summarized by H. YAGODA (1949)<sup>6)</sup> were adopted and also members of very small branching ratio were neglected as usual. The results are

$$\begin{aligned}\sum_{UI} \frac{(R_{UI}-\rho-a)^2}{R_{UI}-\rho} &= 13.66 \\ \sum_{AcU} \frac{(R_{AcU}-\rho-a)^2}{R_{AcU}-\rho} &= 16.97 \\ \sum_{Th} \frac{(R_{Th}-\rho-a)^2}{R_{Th}-\rho} &= 15.18\end{aligned}$$

Hence, the equation (2) becomes:

$$\begin{aligned}n &= K \mu d \cdot 10^3 (44.86UI + 15.64Th) \\ &= 1.50 \cdot K (44.86UI + 15.64Th) \dots\dots\dots(7)\end{aligned}$$

On the other hand, uranium and thorium contents of Milford granite (NBS) were determined by P.M. HURLEY (1959)<sup>7)</sup> as follows:

Table 3. U and Th content of Milford granite and Deccan Trap by means of gamma-scintillation counting.

| Rocks                 | U (ppm) | Th (ppm) |
|-----------------------|---------|----------|
| Milford granite (NBS) | 0.8     | 9.0      |
| Deccan Trap (NBS)     | 0.7     | 2.8      |

Using these values for Milford granite in the table and the average value of  $n$  obtained from five runs of measurement, the factor  $K$  was found from

the equation (7) to be

$$K = 2.5 \pm 0.2$$

Then the equation (7) is reduced to

$$n = 3.7(44.86 UI + 15.64 Th) \dots\dots\dots(8)$$

where  $n$  is expressed as counts per sec. per  $\text{cm}^2$  of the surface area of the sample as a thick source, and  $UI$  and  $Th$  are counts in gram per gram.

In the future, the numerical coefficients in the above equation (8) may be slightly modified to better values by the accumulation of the reliable data on uranium and thorium content of rocks whose chemical analyses are available.

### Measurement with radioscope

The details of the radioscope for use in the measurement of radioactivity of rocks were previously reported by the authors.<sup>1)</sup>

The shifting rate of the indicator of the radioscope is a measure of radioactivity of the samples. In the measurement with rocks which are usually of weak radioactivity, the aluminium screen at the bottom of the ionization chamber was removed so as to utilize efficiently the ionization by alpha-rays from the surface of the specimen placed beneath it.<sup>8)</sup>

In the case of ionization within the limited space of an ionization chamber, the rôles of beta- and gamma-rays are highly reduced owing to their far longer ranges and smaller specific ionizations along their paths as compared with those of alpha-rays. Practically, as reported before,<sup>2)</sup> the contribution of beta- plus gamma-rays in the ionization measurement with a pulverized sample of granite by the electroscope showed nearly 1/6 to 1/7 of the total activity.

Besides this small share in ionization of beta- and gamma-rays within the ionization chamber, the effects of the aluminium foil as absorber are very small for beta-ray, not to mention gamma-ray. For example, that for beta-rays is proved to be ca. 10% with Al-foil of  $9.6 \text{ mg/cm}^2$  in thickness (see Appendix I), so that the following assumption may be reasonably allowed.

Denoting the shifting rate of the indicator of the radioscope by alpha-, beta- and gamma-rays without the Al-foil absorber as  $S_{\alpha+\beta+\gamma}$ , and  $\bar{S}_{\beta+\gamma}$  that by beta- and gamma-rays with absorber, we have

$$S_{\alpha} = S_{\alpha+\beta+\gamma} - S_{\beta+\gamma} \doteq S_{\alpha+\beta+\gamma} - \bar{S}_{\beta+\gamma}$$

Consequently, taking into consideration the absorption of alpha-rays in the sample and in the air before entering the effective space within the ionization chamber,  $S_{\alpha}$  can be approximately expressed as

$$S_{\alpha} = K' \left\{ UI \left( 0.96 \sum_{UI} I_{UI} G_{UI} \frac{(R_{UI} - h)^2}{R_{UI}} + 0.04 \sum_{AcU} I_{AcU} G_{AcU} \frac{(R_{AcU} - h)^2}{R_{AcU}} \right) + Th \sum_{Th} I_{Th} G_{Th} \frac{(R_{Th} - h)^2}{R_{Th}} \right\} \dots\dots\dots (9)$$

where  $h$  is the thickness of air layer between the sample and the effective space of the ionization chamber;  $K'$  is a proportional constant and  $G_{UI}$  etc. are the geometrical reduction factors for the rays from each member of the three radioactive series.  $G_{UI}$  is given as (see Appendix II)

$$G_{UI} = \tau d^2 \left( \frac{l}{d} \right) \left\{ \frac{1}{2} \left( \frac{d}{l} - \frac{d}{R} \right) + \frac{3}{16} \left( \frac{d}{R} \right)^3 \left\{ \left( \frac{l}{R} \right)^2 - 1 \right\} - \frac{5}{128} \left( \frac{d}{R} \right)^5 \left\{ 7 \left( \frac{l}{R} \right)^4 - 10 \left( \frac{l}{R} \right)^2 + 3 \right\} + \text{etc.} \right\} \dots\dots\dots (10)$$

where  $d$ ,  $l$  and  $R$  are shown in Fig. 3.

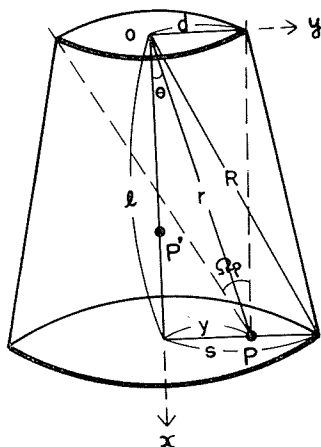


Fig. 3 Illustration figure showing solid angle relation of the sample and the window (the bottom circular area of the effective space) of the ionization chamber.

For the calculation of

$$\sum_{UI} I_{UI} G_{UI} \frac{(R_{UI} - h)^2}{R_{UI}}, \quad \sum_{AcU} I_{AcU} G_{AcU} \frac{(R_{AcU} - h)^2}{R_{AcU}} \quad \text{and} \quad \sum_{Th} I_{Th} G_{Th} \frac{(R_{Th} - h)^2}{R_{Th}},$$

the standard sample of NBS Milford granite and Deccan Trap were used, and from the equation (9), we get

$$S' = K' (0.50 UI' + 0.54 Th') \dots\dots\dots (11).$$

Finding the values of  $S'$  by the actual measurement with NBS Milford granite of which uranium and thorium contents were known as shown in Table 3, the value of  $K'$  was obtained, and finally the equation (11) was reduced to

$$S' = 0.067 (0.50 UI' + 0.54 Th') \dots\dots\dots (12)$$

in which  $S'$  is the shifting rate of the indicator of the radioscope expressed as div/min, and  $UI'$ ,  $Th'$  are respectively the uranium and thorium content in ppm.

Measurements were made to test the validity of the constants used in Eqs. (8) and (12). Assuming the radioactive equilibrium, uranium content was calculated from the known content of radium. Two of the samples used are Ra-standard rocks furnished from the National Bureau of Standard of U. S. A., and the rest one is biotite granite whose radium content has been determined by ASAYAMA, T. The result and the sample were graciously offered to the present authors by him.

Thorium content determined by the alpha-scintillation method using Eq. (8) was compared with that obtained by the radioscope using Eq. (12). As seen in Table 4, the thorium content obtained by the two different methods shows a fair agreement. This assures the safety of the simultaneous use of the two equations, Eq. (8) and Eq. (12).

Table 4. Th-content of Ra-standard rocks determined by two different methods assuming the radioactive equilibrium.

| Rock sample                | <i>U</i> (ppm)             | <i>Th</i> (ppm)                           |                          |
|----------------------------|----------------------------|-------------------------------------------|--------------------------|
|                            | calculated from Ra-content | determined by alpha-scintillation counter | determined by radioscope |
| Graniteville granite (NBS) | 10.0                       | 3.4                                       | 3.4                      |
| Deccan trap basalt (NBS)   | 0.6<br>(0.7) *             | 2.0<br>(2.8) *                            | 2.0                      |
| Biotite granite (ASAYAMA)  | 4.7                        | 18.2                                      | 16.8                     |

\*The parenthesized values are those obtained by Hurley (loc. cit.)

### Formulae giving U- and Th-content by combination of the two kinds of the measurement

As mentioned above, the number of counts by the alpha-scintillation counter per cm<sup>2</sup> of the surface of the thick source of a sample per second is given in the equation (8):

$$n = 3.7(44.86 U + 15.64 Th)$$

where *U* and *Th* are content of uranium and thorium in gram per gram, *UI* being simply denoted by *U*.

The surface area of the sample with our equipment is 113 cm<sup>2</sup>. Therefore, the above equation becomes;

$$n' = 1.125 U' + 0.392Th' \dots\dots\dots(13)$$

where *n'* denotes the observed counts per minute, *U'* and *Th'* content of uranium and thorium respectively in ppm.

From Eqs. (13) and (12), in which *UI'* is again expressed as *U'*, we get

$$\begin{aligned} Th' &= 40.7 S' - 1.21 n' \\ U' &= 29.9 S' - 1.08 Th' \dots\dots\dots(14) \end{aligned}$$

Thus we can find out the uranium and thorium content of the sample by the

combined measurements by an alpha-scintillation counter and a radioscope.

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### Appendix I

In order to see the effect of aluminium foil for beta-ray, the following ratio was calculated:

$$\frac{I_{9.6}}{I_0} = \frac{\sum_{UI} N \cdot F \cdot f_s \cdot f_A \cdot i + \sum_{AcU} N \cdot F \cdot f_s \cdot f_A \cdot i + \sum_{Th} N \cdot F \cdot f_s \cdot f_A \cdot i + N \cdot F \cdot f_s \cdot f_A \cdot i \text{ (for K-40)}}{\sum_{UI} N \cdot F \cdot f_s \cdot i + \sum_{AcU} N \cdot F \cdot f_s \cdot i + \sum_{Th} N \cdot F \cdot f_s \cdot i + N \cdot F \cdot f_s \cdot i \text{ (for K-40)}}$$

where  $I_0$  and  $I_{9.6}$  are intensities of ionization within the electroscope without and with aluminium foil of the thickness 9.6 mg/cm<sup>2</sup> respectively;  $N$  the beta-emissions/g/sec.;  $F$  branching ratios;  $f_A$  and  $f_s$  the correction coefficients for absorption by aluminium foil and the source respectively, and  $i$  ion density produced by beta-ray from each element.

Using the values of these shown in the following table, we have  $I_{9.6}/I_0 \doteq 0.90$  as the effect of the absorber used.

Table Correction coefficients for absorption of beta-rays by the aluminium foil and the source.

| Nuclides                 | $E$<br>(MeV) | $i$ | $F$<br>(%) | $f_s$ | $f_A$ |
|--------------------------|--------------|-----|------------|-------|-------|
| <i>U series</i>          |              |     |            |       |       |
| <sup>234</sup> <i>Th</i> | 0.193        | 96  | 100        | 0.014 | 0.232 |
| <sup>234</sup> <i>Pa</i> | 2.31         | 43  | 100        | 0.479 | 0.960 |
| <sup>214</sup> <i>Pb</i> | 0.65         | 49  | 99.96      | 0.085 | 0.759 |
| <sup>214</sup> <i>Bi</i> | 3.17         | 43  | 99.96      | 0.608 | 0.975 |
| <sup>210</sup> <i>Pb</i> | 0.023        | 440 | 100        | 0.000 | 0.000 |
| <sup>210</sup> <i>Bi</i> | 1.17         | 42  | 100        | 0.204 | 0.321 |

|                               |        |     |       |       |       |
|-------------------------------|--------|-----|-------|-------|-------|
| <i>AcU</i> series             |        |     |       |       |       |
| <sup>231</sup> Th             | 0.302  | 75  | 100   | 0.027 | 0.417 |
| <sup>227</sup> Ac             | 0.0455 | 116 | 98.8  | 0.000 | 0.002 |
| <sup>223</sup> F <sub>r</sub> | 1.15   | 43  | 1.2   | 0.199 | 0.889 |
| <sup>211</sup> Pb             | 1.39   | 42  | 100   | 0.265 | 0.916 |
| <sup>207</sup> Tl             | 1.44   | 42  | 99.68 | 0.280 | 0.920 |
| <i>Th</i> series              |        |     |       |       |       |
| <sup>228</sup> Ra             | 0.012  | 870 | 100   | 0.000 | 0.000 |
| <sup>228</sup> Ac             | 0.218  | 88  | 100   | 0.017 | 0.242 |
| <sup>212</sup> Pb             | 2.25   | 42  | 66.3  | 0.458 | 0.958 |
| <sup>208</sup> Tl             | 1.79   | 42  | 33.7  | 0.358 | 0.915 |
| <sup>40</sup> K               | 1.325  | 42  | 89    | 0.248 | 0.910 |

## Appendix II

The geometrical reduction factors  $G_{UI}$  etc. for rays from each member of the three radioactive series were calculated as follows: In order to calculate the proportion of the emitted rays which go beyond the circular top of the truncated cone (Fig. 3), the solid angle  $\Omega$  subtended by the top opening at any point  $P$  on the base must be known.

For a point  $P'$  on the axis of the truncated cone at a distance  $x$  from the top,

$$\Omega_{P'} = 2\pi \left[ 1 - \left\{ 1 + \left( \frac{d}{2} \right)^2 \right\}^{-\frac{1}{2}} \right]$$

For a point  $P$  at a distance  $r$  from  $O$ , where  $x=l$  and  $\cos\theta=l/r$ ,

$$\begin{aligned} \Omega_P = 2\pi \left[ \frac{1}{2} \cdot \frac{d^2}{r^2} P_1(\cos \theta) - \frac{1}{8} \cdot \frac{d^4}{r^4} P_3(\cos \theta) \right. \\ + \frac{5}{16} \cdot \frac{d^6}{r^6} P_5(\cos \theta) - \frac{35}{128} \cdot \frac{d^8}{r^8} P_7(\cos \theta) \\ \left. + \frac{63}{256} \cdot \frac{d^{10}}{r^{10}} P_9(\cos \theta) - etc. \right], \end{aligned}$$

where  $P_n(\cos \theta)$  are the zonal harmonics.

Now, if  $N_o$  is the number of alpha-particles of  $UI$  emitted from a unit area of the surface of the source;  $N$ , the number of alpha-particles of  $UI$  emerging through the top surface of the truncated cone, which corresponds to the bottom circular area of the effective space within the ionization chamber; and  $R = (S^2 + l^2)^{\frac{1}{2}} = \left[ d^2 + 2d \left\{ (R_{UI} - h)^2 - l^2 \right\}^{\frac{1}{2}} + (R_{UI} - h)^2 + 2l^2 \right]^{\frac{1}{2}}$ , where  $R_{UI}$  is range of alpha-particles from  $UI$ , then,

$$\begin{aligned}
N &= G_{UI} N_o \\
&= N_o \int_0^s \frac{\Omega}{4\pi} \cdot 2\pi y \cdot dy \\
&= \frac{N_o}{2} \int_l^R \Omega \cdot r dr \\
&= \pi d^2 N_o \left( \frac{l}{d} \right) \left\{ \frac{1}{2} \left( \frac{d}{l} - \frac{d}{R} \right) + \frac{3}{16} \left( \frac{d}{R} \right)^3 \left\{ \left( \frac{l}{R} \right)^2 - 1 \right\} \right. \\
&\quad - \frac{5}{128} \left( \frac{d}{R} \right)^5 \left\{ 7 \left( \frac{l}{R} \right)^4 - 10 \left( \frac{l}{R} \right)^2 + 3 \right\} \\
&\quad + \frac{35}{2048} \left( \frac{d}{R} \right)^7 \left\{ 33 \left( \frac{l}{R} \right)^6 - 63 \left( \frac{l}{R} \right)^4 + 35 \left( \frac{l}{R} \right)^2 - 5 \right\} \\
&\quad - \frac{63}{32768} \left( \frac{d}{R} \right)^9 \left\{ 715 \left( \frac{l}{R} \right)^8 - 1716 \left( \frac{l}{R} \right)^6 + 1386 \left( \frac{l}{R} \right)^4 - 420 \left( \frac{l}{R} \right)^2 + 35 \right\} \\
&\quad \left. + \text{etc.} \right\}.
\end{aligned}$$