The Radioactivity of Solids Determined by Alpha-Ray Counting

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The complete theory of alpha-ray counting for thin and thick radioactive sources is developed. Tables of the numerical constants required for the interpretation of practical measurements are given. Apparatus and experimental verification of the theory are described. Several new methods for determining the thorium content of geological samples are described. Analyses of a suite of igneous rocks show Th contents between 3.9 and 16.5×10^{-6}g Th per g, with an average Th/U ratio of 7. Therefore thorium is more important in age and geothermal problems than uranium.

I. INTRODUCTION

RECENT refinements in detection technique permit ionization measurements[1] on the alpha-rays emitted by all ordinary materials because of their natural contamination by traces of radioactive substances. Accurate alpha-ray measurements permit the determination of the thorium content of geological specimens, hence lead toward satisfactory solutions of such basic problems as the age of igneous rocks, the age of the stellar system as determined from meteorite analyses, and the excessive, radioactivity-derived internal heat of the earth.

Feebly radioactive sources such as igneous rocks emit the order of one alpha-ray per cm^2 per hour. Any measurements are therefore subject to statistical fluctuations due to the random time of emission of these alpha-rays. Ionization measurements are further impaired by ionization fluctuations due to the variation in residual track length of the emitted alpha-rays, which must be compounded with the effects of random rate of emission in accord with the statistical theory of independently random processes.[2] Ionization measurements must also deal with the unavoidable background due to cosmic-ray and local gamma-ray ionization in the detecting chamber.

These difficulties are circumvented by counting the individual alpha-rays with a detector which is unresponsive to cosmic or local gamma-radiation. Precision is then limited only by the statistical fluctuations in the rate of emission of alpha-rays, and by the accuracy of the computations dealing with the finite thickness of the source. The statistical problem concerning several alpha-emitters of widely different half-periods but in radioactive equilibrium has been treated by Adams.[3] The equations describing thickness corrections for sources containing any number of radioactive elements are here developed and confirmed by direct experiments.

II. THEORY OF PARTICLE COUNTING

Consider the section of a radioactive solid bounded by the planes AA' and BB' in Fig. 1, and surmounted by absorbers of thickness a_1 and a_2 cm. The source emits N alpha-rays per cm^3 per sec., of mean range R air-cm (i.e., measured in air at 0°C, 760 mm Hg). The full alpha-ray range in the source would be μR cm, and in the absorbers $\mu_1 R$, and $\mu_2 R$, respectively. The source is $\mu\tau$ cm (or τ air-cm) thick and some of the alpha-rays which it emits are observable in the detecting chamber located above the plane DD'.

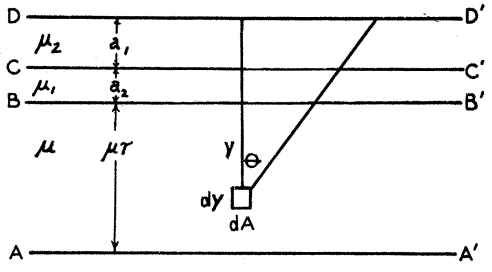

FIG. 1. Geometry for Eqs. (1) *et seq. Errata:* Subscripts for a_1 and a_2 should be interchanged.

[1] Evans, Phys. Rev. **45**, 29, 38 (1934).
[2] Evans and Neher, Phys. Rev. **45**, 144 (1934).
[3] N. I. Adams, Jr., Phys. Rev. **44**, 651 (1933).

A. Thin sources

There are $N\,dy\,dA$ alpha-rays per sec. emitted in all directions from the volume element $dydA$ located y cm below BB'. Alpha-rays which have equivalent air ranges of r or less below the interface DD', hence residual ranges equal to or greater than $(R-r)$ in the detecting chamber above DD', will be emitted within a cone of half-angle θ, or fractional solid angle $(1-\cos\theta)/2$, where $r\cos\theta=y/\mu+a_1/\mu_1+a_2/\mu_2$. Hence the number of such rays from the elementary volume is $N\,dy\,dA(1-\cos\theta)/2$, and n_a' the total number of such rays per cm² of surface of a source of large area (where edge effects are negligible) is obtained by integrating over y between the limits: $y=0$, and $y=\mu\tau$

$$n_a'=\frac{N}{2}\int_0^{\mu\tau}(1-\cos\theta)dy$$

$$=\frac{N}{2}\int_0^{\mu\tau}\left[1-\frac{y/\mu+a_1/\mu_1+a_2/\mu_2}{r}\right]dy \qquad (1)$$

$$=\frac{N\mu\tau}{4}\left[\frac{2(r-a)-\tau}{r}\right] \quad \text{where}$$

$$a\equiv a_1/\mu_1+a_2/\mu_2\cdots\text{air cm.} \qquad (2)$$

Now if the detecting instrument above DD' responds only to alpha-rays having a residual range equal to or greater than ρ air-cm, then the number of counted alpha-rays per cm² per sec. is given by Eq. (2) when $r=R-\rho$, that is:

$$n'_a=(N\mu\tau/4)\cdot[2(R-\rho-a)-\tau]/(R-\rho), \qquad (3)$$

which is the counting rate for thin sources surmounted by absorbers.[4]

If the thin source has an area A then the total alpha-ray counting rate is $n_a'A$. When d is the density of the source, its total mass $m=\mu\tau Ad$, and $n_a''\mu\tau Ad=n_a'A$, where n_a'' is the total alpha-ray counting rate per g of source. Therefore:

$$n_a''=n_a'/\mu\tau d$$
$$=(N''/4)[2(R-\rho-a)-(m/\mu Ad)]/(R-\rho), \qquad (4)$$

where $N/d\equiv N''$ alpha-rays emitted per sec. per g of source. When an infinitely thin source is used without an absorber $\tau=m=a=0$, and Eq. (4) reduces to $n''=N''/2$. Only in this limiting case is the counting rate independent of R. Under these conditions the alpha-ray counting rate is a direct measure of the rate of helium production, hence could be directly combined with a helium analysis on a bulk rock specimen to determine its age.

N and N'' may be expressed in terms of the fractional concentration of radioactive material Q g per g, its decay constant λ sec.$^{-1}$, its atomic weight W, and the density d of the source; thus:

$$N=6.06\times10^{23}Q\lambda d/W, \qquad (5)$$

$$N''=N/d=6.06\times10^{23}Q\lambda/W. \qquad (6)$$

When the source thickness increases to $\tau=R-\rho-a$, any further increase in thickness will not increase the counting rate because even the alpha-rays directed normal to the plane DD' will emerge with residual ranges less than ρ and hence cannot be counted. The thickness $\tau=R-\rho-a$ is the transition between thin sources, whose theory has been given above, and thick sources, which are treated later. The variation in counting rate with source thickness may be obtained by employing Eq. (3) to derive the ratio of n_a' the counting rate above a thin source to n_a the rate above a thick source, both having the same absorber a superposed.

The counting rate n_a for thick sources (i.e., $\tau\geqslant R-\rho-a$) follows from Eq. (3) for a source of critical thickness $\tau=R-\rho-a$. Substituting, we find:

$$n_a=(N\mu/4)(R-\rho-a)^2/(R-\rho). \qquad (7)$$

Dividing Eq. (3) by Eq. (7) we obtain the variation in counting rate with source thickness.

$$n_a'/n_a=\tau[2(R-\rho-a)-\tau]/(R-\rho-a)^2$$
$$=1-[1-\tau/(R-\rho-a)]^2 \qquad (8)$$
$$\equiv1-(1-F_\tau)^2, \qquad (9)$$

where F_τ is the fractional thickness $\tau/(R-\rho-a)$ of the source, and $0\leqslant F_\tau\leqslant1$, as $0\leqslant(n_a'/n_a)\leqslant1$. In Fig. 2, the ordinates at the right are n_a'/n_a, when the abscissae are F_τ.

B. Thick sources

Sources are called thick when $\tau\geqslant R-\rho-a$, in which case the counting rate is independent of source thickness and is given by Eq. (7).

[4] Evans, Phys. Rev. **45**, 29 (1934).

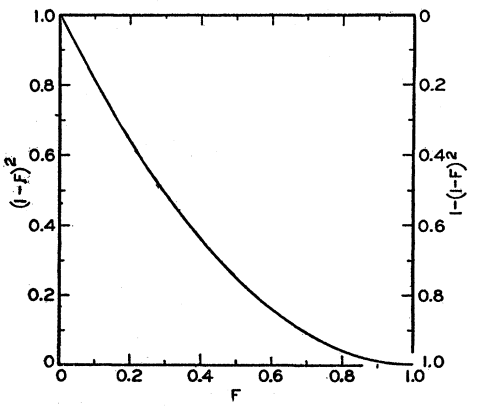

FIG. 2. Left-hand ordinates show fractional counting rate from thick sources surmounted by absorbers of fractional thickness $F_a=a/(R-\rho)$; see Eq. (11). Right-hand ordinates show fractional counting rate from thin sources having a fractional thickness $F_\tau=\tau/(R-\rho-a)$; see Eq. (9).

The variation with absorber thickness of the counting rate due to a thick source is best computed in terms of the counting rate n for zero absorption. Thus from Eq. (7):

$$n_a/n=(R-\rho-a)^2/(R-\rho)^2$$
$$=[1-a/(R-\rho)]^2 \quad (10)$$
$$\equiv(1-F_a)^2, \quad (11)$$

where F_a is the fractional thickness $a/(R-\rho)$ of the absorber, and $0\leqslant F_a\leqslant 1$, as $0\leqslant(n_a/n)\leqslant 1$. In Fig. 2, the ordinates at the left are n_a/n when the abscissae represent F_a.

Combining Eq. (8) for $a=0$, with Eq. (10), we find:

$$n'/n+n_a/n=1-[1-\tau/(R-\rho)]^2+[1-a/(R-\rho)]^2. \quad (12)$$

Hence if the thin source has a thickness τ, and no absorber, its counting rate n' may be immediately obtained from curves for the counting rate n_a due to a thick source with an absorber of thickness a equal to τ. Or, analytically, Eq. (12) reduces to:

$$n'/n+n_a/n=1 \quad \text{when} \quad \tau=a. \quad (13)$$

Eq. (13) holds for each particular alpha-ray range R, hence also for the integrated effects of several alpha-ray emitters in the same source. It is the basic correspondence theorem correlating the theories of thin and thick sources, and its use halves the numerical calculations required for applications to sources emitting alpha-rays from many or all members of the U, Th and Ac series. It can, of course, be verified by inspection of the geometry involved.

It has been shown previously[4] that the decreased counting rate due to alpha-particle straggling in the solid is almost completely offset by the gain in counting rate occasioned by the symmetrical character of the error function describing straggling. The fractional loss is $\Delta n/n=\frac{1}{2}(\rho_3)^2$, where $\rho_3 r$ is Brigg's range straggling coefficient. The fractional loss is thus always less than 0.3 percent.

III. Total Counting Rate for Sources Containing Many Radioactive Elements

Measurements on chemically prepared materials containing only one alpha-ray emitting element are easily interpreted by Eqs. (3) and (7). A more complicated case is met in measurements of the total alpha-ray emission from materials containing all members of the U, Th and Ac radioactive series, such as rocks. We shall consider the case of thick sources; values for thin sources may be readily obtained by use of the correspondence theorem of Eq. (13).

We shall regard the specimen as sufficiently old to have reached radioactive equilibrium. Then the number of alpha-rays produced per cm^3 per sec. is essentially the same for each member of a radioactive decay series. Substituting[5] the decay constant and atomic weight of U and of Th in Eq. (5) we find:

$$N_U=\frac{6.06\times10^{23}\times4.77\times10^{-18}}{238.14}\cdot U\cdot d$$
$$=1.21\times10^4\,U\cdot d. \quad (14)$$

$$N_{Th}=\frac{6.06\times10^{23}\times1.7\times10^{-18}}{232.12}\cdot Th\cdot d$$
$$=0.44\times10^4\,Th\cdot d. \quad (15)$$

$$N_{Ac}=0.04N_U=0.05\times10^4\,U\cdot d. \quad (16)$$

[5] Kovarik and Adams, Phys. Rev. **40**, 718 (1932) give $\lambda=4.865\times10^{-18}$ sec.$^{-1}$ for UI. When corrected for AcU, this value becomes 4.77×10^{-18} sec.$^{-1}$. Geiger and Rutherford, Phil. Mag. **20**, 691 (1910), and H. Fesefeldt, Zeits. f. Physik **86**, 605 (1933), find $\lambda=1.7\times10^{-18}$ sec.$^{-1}$ for Th. A. v. Grosse, Phys. Rev. **42**, 565 (1932) reviews the work on Ac and recommends 0.04 for the Ac/U activity ratio.

Since each alpha-ray body in the U series emits 1.21×10^4 U$\cdot d$ alphas per sec., the total counting rate for a thick source containing only the U series would be:

$$(1.21\times10^4\ \mathrm{U}\cdot\mu\cdot d/4)\Sigma(R-\rho-a)^2/(R-\rho)$$

where the summation extends over all the range values R which are members of the uranium series.

In a natural source, such as a granitic rock, the Th and Ac series will be present also, hence similar expressions must be written for them. The total observed counting rate n_a will then be due to terms from the U plus Ac series and from the Th series, or

$$n_a=v_a\mathrm{U}+w_a\mathrm{Th} \tag{17}$$

where U and Th are the uranium and thorium concentrations in g per g of source. Writing

$$H\equiv\Sigma(R-\rho-a)^2/(R-\rho) \tag{18}$$

we have:

$$v_a=(1.21\times10^4\mu d/4)H_{\mathrm{U}}+(0.05\times10^4\mu d/4)H_{\mathrm{Ac}}$$

$$=[3.03H_{\mathrm{U}}+0.12H_{\mathrm{Ac}}]10^3\mu d \tag{19}$$

$$w_a=[1.11H_{\mathrm{Th}}]10^3\mu d \tag{20}$$

where the summations are made over the members of the U, Th and Ac series as indicated, and H for the CC′C″ bodies considers their respective branching ratios.

The computation of H_{U}, H_{Ac} and H_{Th} may be considerably simplified by the following devices. Let Δj be a numerical parameter defined as the ratio of the number of alpha-rays per second from any element, such as Ra, to the number from its parent, in this case U I. Then, without practical error, $\Delta j=1$ for all alpha-emitting elements, except those in the CC′C″ branches, when radioactive equilibrium is present. For Ra C, $\Delta j=0$ (accurately, 0.0004); for Ra C′, $\Delta j=1$ (0.9996); for Th C, $\Delta j=0.35$; for Th C′, $\Delta j=0.65$; for Ac C, $\Delta j=1$ (0.997); and for Ac C′, $\Delta j=0$ (0.003). To account analytically for branching we therefore introduce the parameter Δj into Eq. (18), obtaining:

$$H=\Sigma\Delta j(R-\rho-a)^2/(R-\rho) \tag{21}$$

$$=\Sigma\Delta j(R-\rho)[1-a/(R-\rho)]^2$$

$$=\Sigma R\cdot\Delta j+a^2\Sigma\Delta j/(R-\rho)-(\rho+2a)\Sigma\Delta j. \tag{22}$$

TABLE I. *Mean range R in air at 0°C and 760 mm Hg; and Δj, j, Pj, Qj (see Eq. (23)) for all alpha-ray emitters in the U, Ac and Th series.*

	R	Δj	j	P_j	$Q_j(\rho=0.5)$
URANIUM SERIES					
Ra C′	6.51₃[1]	0.9998[2]	1	6.51	0.1665
Ra A	4.38[1]	1	2	10.89	0.425
Ra C (av)	3.80[2]	0.0002[2]	2	10.89	0.425
Rn	3.80[1]	1	3	14.69	0.728
Ra F	3.61[1]	1	4	18.30	1.050
Ra	3.16[3]	1	5	21.46	1.426
UI I	3.05[3]	1	6	24.51	1.818
Io	2.97[3]	1	7	27.48	2.223
U I	2.51[3]	1	8	29.99	2.721
ACTINIUM SERIES					
Ac C′	6.17[1]	0.003[3]	0	0	0
Ac A	6.08[1]	1	1	6.08	0.179
An (av)	5.29[1]	1	2	11.37	0.388
Ac C (av)	5.04[1]	0.997[3]	3	16.41	0.608
Rd Ac	4.36[3]	1	4	20.77	0.867
Ac X	4.07[3]	1	5	24.84	1.147
Pa	3.42[3]	1	6	28.26	1.489
Ac U	3.0[4]	1	7	31.26	1.889
THORIUM SERIES					
Th C′	8.09[1]	0.65[3]	0.65	5.26	0.0857
Th A	5.31[1]	1	1.65	10.57	0.294
Tn	4.71[1]	1	2.65	15.28	0.532
Th C	4.45[1]	0.35[3]	3	16.84	0.620
Th X	4.06[3]	1	4	20.90	0.901
Rd Th	3.74[3]	1	5	24.64	1.210
Th	2.66[5]	1	6	27.30	1.673

[1] Lewis and Wynn-Williams, Proc. Roy. Soc. **A136**, 349 (1932).
[2] Rutherford, Wynn-Williams, Lewis and Bowden, Proc. Roy. Soc. **A139**, 617 (1933).
[3] Rutherford, Chadwick and Ellis, *Radiations from Radioactive Substances* (1930), p. 26. The extrapolated ionization ranges R_i there given are converted to mean ranges using the relation[5] $R_i-R=0.042(R_i-1.51)^{\frac{3}{2}}$ cm.
[4] Evans, Phys. Rev. **45**, 29 (1934), estimated from the half-period, A. v. Grosse, Phys. Rev. **42**, 565 (1932), and the Geiger-Nuttall relation using Geiger's, Zeits. f. Physik **8**, 45 (1921), data for the Ac series.
[5] Kurie and Knopf, Phys. Rev. **43**, 311 (1933), give the extrapolated ionization range R_i in air at 0°, 760 mm as 2.71±0.03 cm. The mean range is estimated by deducting $0.042\ (R_i-1.51)^{\frac{3}{2}}$ cm [Briggs, Proc. Roy. Soc. **A114**, 313 (1927); Kurie, Phys. Rev. **41**, 701 (1932); Evans, Phys. Rev. **45**, 29 (1934)]. Using Lewis and Wynn-Williams (reference 1, Table I) relationship, $R_2-R=0.8\rho-0.06$, where ρ is the straggling coefficient ($\rho\equiv\rho_{3X}$ used by Briggs) one obtains $R=2.67$ for Th. G. H. Henderson and Nickerson, Phys. Rev. **36**, 1344 (1930) obtained a lower value $R_i=2.59\pm0.05$ cm at 15°C, 760 mm, while Geiger, Zeits. f. Physik **8**, 45 (1922) found $R_i=2.90$ cm at 15°C, 760 mm.

Now we define $P_j\equiv\Sigma R\cdot\Delta j$, $Q_j\equiv\Sigma\Delta j/(R-\rho)$, and $j\equiv\Sigma\Delta j$; then Eq. (22) becomes:

$$H=P_j+a^2Q_j-j(\rho+2a). \tag{23}$$

In evaluating P_j, Q_j and j the summation extends only over those alpha-rays having $R\geqslant a+\rho$, since only these contribute countable rays. Table I gives the values of R, Δj, j, P_j, and Q_j for all alpha-rays in the U, Ac and Th series.

As an example, consider a thick radioactive source containing only the Th series. Let the minimum detectable track $\rho=0.5$ air-cm and the absorber $a=4$ air-cm. Then only alpha-rays having $R\geqslant4.5$ air-cm may be counted. From Table I this is seen to include Th C′, Th A and Tn, but neither Th C nor shorter members. Reading across the row corresponding to the shortest

TABLE II. *Dependence on absorber thickness, a air-cm, of the relative counting rate v_a/v for the U plus Ac series in equilibrium, w_a/w for the Th series, and v_a/w_a the ratio of U plus Ac series to Th series counts.* All values correspond to $\rho=0.5$ air-cm. Values of v_a and w_a (see Eqs. (19) and (20)) are also given.

a	$v_a/10^3\mu d$	$w_a/10^3\mu d$	v_a/w_a	v_a/v	w_a/w
0	82.1	27.0	3.04	1.000	1.000
1	40.4	15.52	2.60	0.493	0.575
2	15.62	7.75	2.02	0.1905	0.287
3	5.63	3.36	1.68	0.0687	0.124
4	2.12	1.40	1.51	0.0258	0.052
5	0.529	0.633	0.84	0.00645	0.0236
6	0	0.237	0	0	0.0089

countable emitter, Tn, we find $j=2.65$, $P_j=15.28$, $Q_j=0.532$. Hence by Eq. (23), $H_{\mathrm{Th}}=15.28+(4)^2\times0.532-2.65(0.5+2\times4)=1.26$. Finally, from Eqs. (17) and (20), the number of alpha-rays counted per sec. per cm² of source surface is $n_a=w_a\mathrm{Th}=1.11\times10^3H_{\mathrm{Th}}\mu d\cdot\mathrm{Th}=1.40\times10^3\mu d\cdot\mathrm{Th}$, when $a=4$, $\rho=0.5$ air-cm. This value will be found in Table II.

Writing subscripts 1 and 2 to characterize two values of absorber thickness, a_1 and a_2, we may write the equations for absorption measurements which might permit separation of the U and Th effects, and hence provide a direct measure of either or both of these elements.

$$n_1=v_1\mathrm{U}+w_1\mathrm{Th},\quad n_2=v_2\mathrm{U}+w_2\mathrm{Th}. \tag{24}$$

Combining these equations we obtain:

$$\mathrm{Th}=(n_1/w_1)(n_2/n_1-v_2/v_1)/(w_2/w_1-v_2/v_1), \tag{25}$$

$$\mathrm{U}=(n_1/v_1)(w_2/w_1-n_2/n_1)/(w_2/w_1-v_2/v_1), \tag{26}$$

$$\mathrm{Th/U}=(v_1/w_1)(n_2/n_1-v_2/v_1)/(w_2/w_1-n_2/n_1), \tag{27}$$

$$n_1/n_2=(w_1/w_2)(v_1/w_1+\mathrm{Th/U})/(v_2/w_2+\mathrm{Th/U}). \tag{28}$$

Here n_1 and n_2 are values which can be observed experimentally, while v_1, v_2, w_1, w_2 are computed theoretically, or obtained experimentally by internal standardization, as described in Section IV.

Numerical examination of Eqs. (25), (26), (27), (28) quickly shows that an impossibly high experimental precision is required if U and Th concentrations are to be obtained from experiments in which the two absorbers differ in thickness by only about an air-cm. If, however, a_1 approaches zero, while a_2 is made the order of 5 or 6 air-cm, then separation of U and Th becomes possible even for quite feebly radioactive sources. Due to the low background counting rate attainable in counting experiments these measurements become possible, whereas they are forbidden by the relatively high gamma- and cosmic-ray background if ionization methods are employed.[4]

It will be noticed by comparison with Eqs. (19) and (20) that Eqs. (25) and (26) express the Th and U content in terms of $(1/\mu d)$ since the product μd occurs once in each term involving v or w. Experimentally, the effective stopping power $(1/\mu)$, is not readily determinable for rocks, particularly if they are coarse grained. The product μd can be approximated, however, from the Bragg-Kleeman rule which gives

$$\mu d_g=d_aW_g^{\frac{1}{2}}/W_a^{\frac{1}{2}}, \tag{29}$$

where $d_a=$ density of air at 0°C, 760 mm; $d_g=$density of the mineral grain; $W_g^{\frac{1}{2}}=$sum of the atomic percent times the square root of the atomic weight of each of the chemical constituents of the mineral grain; $W_a^{\frac{1}{2}}=$the similarly computed mean square root of the atomic weight for air. Direct measurements of μd for mica have been made by Briggs[6] and by G. H. Henderson[6] who obtained 1.547×10^{-3}g/cm² and 1.659×10^{-3}g/cm², respectively. Computations of μd for mica have been given by G. H. Henderson[6] and by Kerr-Lawson,[6] who found 1.629×10^{-3}g/cm² and 1.639×10^{-3}g/cm², respectively. Since $W_g^{\frac{1}{2}}$ is about 4.79 for mica (Kerr-Lawson) we may say that within an uncertainty of about ±3 percent

$$\mu d_g=3.4\times10^{-4}W_g^{\frac{1}{2}}. \tag{30}$$

This value is also obtained when the known values of d_a and $W_a^{\frac{1}{2}}$ are substituted in Eq. (29).

For practical applications of Eqs. (24) to (28) we need tables of v_2/v_1, w_2/w_1, and the values of v and w when $a=0$. These are obtained from the data of Table I combined with Eqs. (19) and (20), and are given in Table II for the case in which $\rho=0.5$ air-cm, which is an easily

[6] G. H. Briggs, Proc. Roy. Soc. **A114**, 341 (1927). G. H. Henderson, Proc. Roy. Soc. **A145**, 563 (1934). D. E. Kerr-Lawson, Univ. of Toronto Studies, Geological Series, No. 24, p. 54 (1927). Each of the values of μd quoted above has been corrected to 0°C, 760 mm Hg pressure.

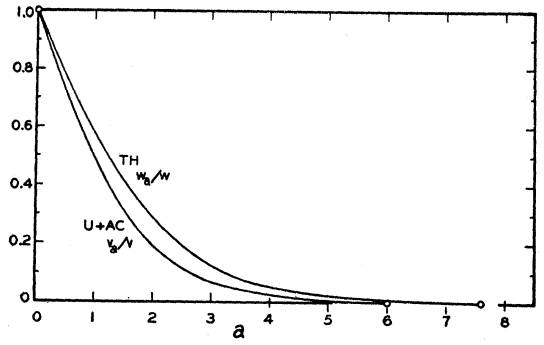

FIG. 3. Dependence of the relative counting rate v_a/v for the U plus Ac series in equilibrium, and w_a/w for the Th series, on absorber thickness a air-cm, when $\rho=0.5$ air-cm.

obtained experimental condition. Table II corresponds to Eqs. (24) to (28) when $a_1=0$, $a_2=a$; $v_1=v_0\equiv v$, $w_1=w_0\equiv w$; and $v_2\equiv v_a$, $w_2\equiv w_a$. Fig. 3 shows graphically the dependence of v_a/v and w_a/w on a.

IV. Determination of the Thorium Content of Geological Specimens

Completely satisfactory experimental methods are available for determining the radium (hence uranium) content of rocks.[7] Geological age and thermal problems also require accurate knowledge of the thorium content of rocks. The results of Section III lead to several new methods for determining the thorium content of rocks[8] by counting the alpha-rays emitted from the rock surface.

Method A rests on absorption measurements and the use of Eq. (25). The value of μd for the source may be estimated from Eq. (30), or obtained experimentally by internal standardization, that is, by adding a known amount of a finely powdered Th mineral such as monazite sand to a powdered rock specimen and determining the change in the observed counting rate.

Method B also involves absorption measurements, but employs Eq. (27) to evaluate the Th/U ratio. This method is independent of the value of μd for the source, but must be combined with a separate determination of U by emanation measurements. These can be made with high precision. The method B is really equivalent to a

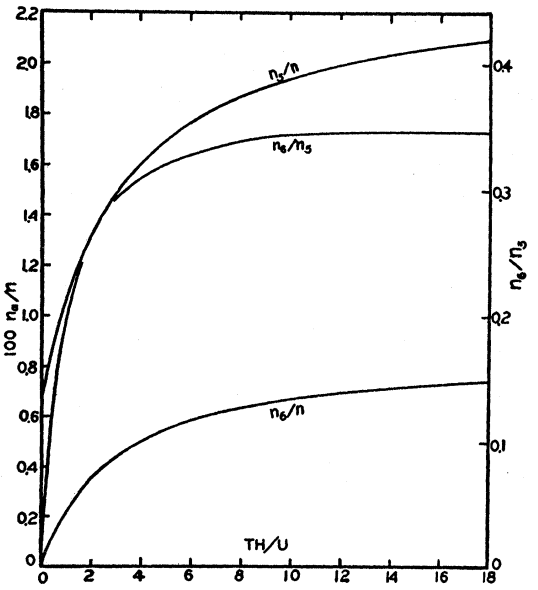

FIG. 4. Left-hand ordinates: ratio n_a/n of counting rates with and without absorbers; curve n_5/n for $a=5$ air-cm, curve n_6/n for $a=6$ air-cm. Right-hand ordinates: ratio n_6/n_5 of counting rates with $a=6$ to $a=5$ air-cm absorption. Abscissae: Th/U ratio in g per g.

combination of method A with a determination of μd by means of Eq. (26) and the independently measured uranium content. Fig. 4 shows the dependence of the relative counting rate n_a/n with 5 and 6 air-cm absorbers to the counting rate with no absorber, and also n_6/n_5 the relative counting rate with 6 air-cm and with 5 air-cm absorbers, each as a function of the Th/U ratio. It will be noted that the method is best suited to Th/U ratio less than about 6; for higher Th/U ratios, method C is preferable.

Method C is especially applicable to very weak sources in which absorption measurements would reduce the counting rate to an impractically low value. It is, therefore, generally applicable even when methods A and B become objectionally inaccurate due to statistical fluctuations in the counting rates. It consists of simply measuring the total counting rate from a source with no absorber, hence by Eq. (24) and Table II:

$$n=(82.1\mathrm{U}+27.0\mathrm{Th})10^3\mu d. \qquad (31)$$

As in method A, μd is evaluated by internal standardization using either a thorium or a uranium mineral, or μd is estimated from Eq. (30). In the latter case, it must be remembered

[7] Evans, Rev. Sci. Inst. **6**, 99 (1935).
[8] Evans, Finney, Kip and Mugele, Phys. Rev. **47**, 791 (1935).

that if a coarsely powdered rock is used as a source, the density of the source d in Eq. (31) is less than the density d_g of the individual grains treated in Eq. (30). Since the ratio d/d_g is not easily measured, the method of internal standardization gives a more reliable value of μd. A separate measurement of the uranium content is then used with Eq. (31) for the determination of the thorium content of the specimen. Since knowledge of the U content is always required in age or thermal problems, this involves no additional work and is therefore not objectionable.

Where ΔTh is the known change in Th content, introduced as an internal standard, and Δn is the corresponding change in the counting rate n, we have

$$\text{Th} = n(\Delta\text{Th}/\Delta n) - (v_a/w_a)\text{U} \quad (32)$$

$$= n(\Delta\text{Th}/\Delta n) - 3.04\text{U}(\text{when } a=0). \quad (33)$$

Method D. For sufficiently strong sources a fourth method is available. This consists of a single measurement of the counting rate with an absorber sufficiently thick ($a+\rho=6.5$ air-cm at 0°C 760 mm) to stop all but the 8.08 air-cm alpha-ray from Th C′. This method is really a special limiting case of method A, for when $v_2=0$, i.e., complete U series absorption, Eq. (25) leads to:

$$\text{Th} = n_2/w_2 = 4.22\times10^{-3} n_a/\mu d \quad (\text{for } a=6,\ \rho=0.5), \quad (34)$$

where n_a is the counting rate per cm² per sec., and μd is obtained either by internal standardization or from Eq. (30). Taking $\mu d = 1.63\times10^{-3}$, as for mica, we then have $\text{Th} = 2.6 n_a$. If the source is a granitic rock having 10^{-5}g Th per g the total counting rate through the 6 air-cm absorber will then be $10^{-5}\times150\times3600/2.6 = 2.8$ alphas per hr. per 150 cm², which is detectable on the apparatus described in Section V. For sources containing as much as 10^{-4} g Th per g the method becomes quite accurate, as statistical fluctuations are then much less pronounced.

V. Apparatus for Counting Alpha-Rays from Large Surface Areas

The counting chamber (Fig. 5) is a parallel plate condenser having a lower collecting electrode 10 cm in diameter. This electrode is surrounded at a distance of 1 mm by a circular guard ring 2.5 cm wide. The edges of both are beveled to reduce the electrostatic capacity of the collector. The upper electrode is 2 cm above the lower collecting plate, and is mounted on a brass ring supported by Bakelite blocks. In the early experiments a grid of parallel, gauge 40, German silver wires spaced 5 mm apart was used. The alpha-ray source was mounted immediately over it with the active face downward and the active area defined by a circular opening in a brass mask which rested on the upper electrode ring and supported the source.

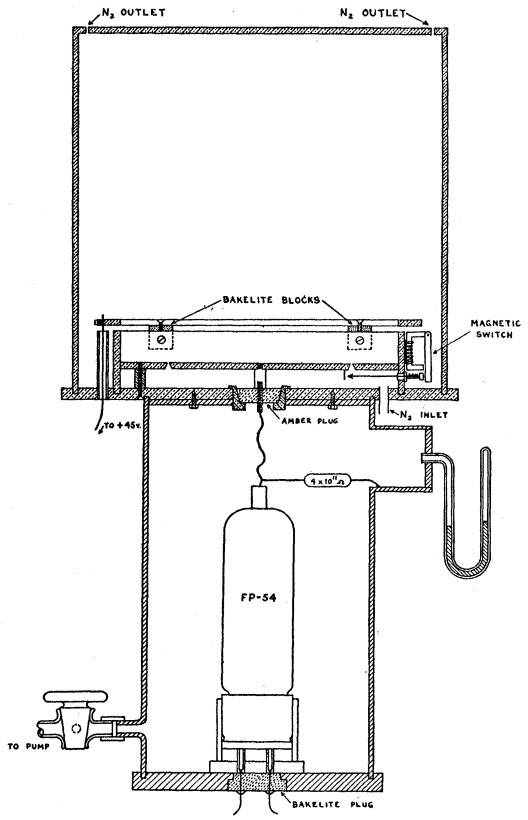

FIG. 5. Construction of the counting chamber, source holder, and electrometer-tube housing. Accurately to scale.

The source is either a flat surface polished onto a hand specimen of a rock; a chemically prepared precipitate painted onto an aluminum disk by using ethyl alcohol as a diluent; or a finely ground rock powder painted on in the same way. The source is covered with a sheet of thin aluminum or gold leaf (stopping power 0.03 air-cm) fastened to the periphery of the source disk

with vaseline. This covering provides a uniform electrostatic field normal to the surface of the source, and prevents minute free grains of the source from dropping into the counting chamber. Gold leaf will not prevent the escape of Rn and Tn from polished rock surfaces, but direct tests by several methods show that the powdered rock sources do not permit the escape of a significant amount of emanation.

Satisfactory electrical operation can be realized without the use of this covering foil, even when the wire grid is removed from below it, but we have always used the foil in order to provide exactly reproducible collecting fields for all specimens. In this case it is best to remove the wire grid, making the foil which covers the source act as the upper electrode of the collecting chamber. This procedure also eliminates the absorption of alpha-rays by the wires of the grid and by the gas between the source and the grid, and avoids the uncertainties of penetration of the collecting field beyond the wires of the grid. The equations of Section III permit accurate correction for the small absorption in the foil. For absorption measurements, aluminum foils are placed directly between the source and the chamber, and nearly in contact with the source.

In addition, powdered sources similarly prepared on Al foils may be placed directly on the lower collecting electrode with their active surfaces facing upward into the counting chamber. It is thus possible to utilize over 150 cm^2 of active source material.

The lead from the lower collecting electrode passes through an amber insulator into an evacuated iron tube-housing patterned after Bearden's[9] apparatus. The amplifying circuit follows the methods of DuBridge and Brown,[10] and the output galvanometer deflection is continuously photographed with a telechron driven drum camera nearly identical with that developed for string electrometer measurements by Evans.[7] Eastman P.M.C. No. 2 contrast bromide emulsion on smooth single-weight paper, 5 inches wide, is used for recording. The drum is 12 inches in diameter and rotates once an hour, hence the travel is 1.6 cm per min.

This high film speed permits recording the full form of the alpha-ray deflection and gives a distinctive hump with a steep initial slope, corresponding to the ballistic period of the galvanometer; a maximum height corresponding to the range of the alpha-ray in the chamber; and an exponential return to the undeflected position, corresponding to the electrostatic time constant of the collecting system. From measurements on the shape of the return curve, and knowledge of the value of the grid resistance (4×10^{11} ohms), the electrostatic capacity of the collecting system and its leads is found to be 36 cm.

The sensitivity[11] is such that a 1 cm alpha-ray produces a 1 cm deflection of the galvanometer. The maximum noise level fluctuations are about 2 to 3 mm hence, while shorter alpha-rays can be clearly identified, we have taken ρ as 0.5 air-cm (0°C 760 mm of air) for then there can be no ambiguity.

Radioactively dead nitrogen, from tanks stored longer than one month, is flowed continuously through the collecting chamber and its cylindrical brass housing at a rate of about 20 cm^3 per minute, which causes no appreciable pressure rise in the chamber. This excludes room air from the apparatus and eliminates resultant contamination. Great care must be taken in preparing sources, and in all operations with the apparatus, to avoid traces of contamination. Numerous control and background runs accompany every measurement of any source, in order to avoid contamination effects. The parts of the counting chamber are readily removable for cleaning, and average background counting rates of 5 alphas per hour are realizable, while 10 per hour is an easily reproduced routine standard. The maximum resolvable counting rate is the order of 350 alphas per hour, and various classes of rock specimens normally run between 20 and 200 alphas per hour per 50 cm^2 of surface area.

The entire apparatus except the camera is housed in a double-walled masonite case for protection from temperature variations, and all electrical parts are electrostatically shielded.

[9] Bearden, Rev. Sci. Inst. **4**, 271 (1933).
[10] DuBridge and Brown, Rev. Sci. Inst. **4**, 532 (1933).

[11] The apparatus has been moved from the University of California to the Massachusetts Institute of Technology, where, with the assistance of Mr. Howard Tatel, a Western Electric 7567-A electrometer tube has been substituted for the FP-54 formerly employed.

The galvanometer spot is automatically moved across the camera slit so that successive hours' records lie side by side on a single photograph. Two methods, each telechron controlled, have been successfully used. By means of a 22 contact, rotary, magnetic telephone relay, which is fed hourly impulses from contacts on the minute and second hands of a telechron clock, the plate resistance and hence the galvanometer zero may be shifted over suddenly once each hour. Alternatively, the galvanometer lamp, mounted on a traveling carriage, may be moved uniformly by means of a screw-thread driven at any speed by a gear on the minute hand shaft of a telechron motor. The former method was used for over 2200 hours of photographic records without any failures, but the latter method has proved preferable at very high sensitivities, because of the low and critical value of the plate circuit resistance.

VI. Experimental Results

Analyses of suites of rock specimens, protactinium sources, kolm, etc., will be separately reported in other journals. In addition we have measured the alpha-ray emission from artificial rock sources in order to verify the theoretical equations of Section III. The specimens were prepared by mixing small amounts of a weak uranium ore (carnotite) and a weak thorium ore (monazite) with plaster of Paris, and have been previously described in connection with ionization measurements on alpha-rays emitted by solids.[12] The uranium series sources contain 2.53×10^{-5} g U per g and 2.53×10^{-6} g U per g; the thorium series sources 4.25×10^{-5} g Th per g and 4.25×10^{-6} g Th per g.

Taking the value of μd_0 from Eq. (30), and measuring d for the plaster of Paris ($=1.32$ g cm^{-3}), it is found that the observed counting rate, within the statistical fluctuation of ±3 to 5 percent occasioned by the random emission of alpha-rays, is equal to that predicted by the equations of Section III.

Further calibration tests were made by adding known small amounts of the uranium and thorium series to powders of igneous rocks, in the course of repeated internal standardization tests. Here again the experimental observations are in close agreement with Eqs. (30), (32), and the analog of Eq. (32) for the uranium series. The agreement in the case of the thorium standards tends to support the Geiger-Rutherford value of the Th decay constant which was chosen[5] in Section III as more probable than Kirsch's lower value.[13]

In general, the thorium analyses of igneous rocks have shown values between 3.9 and 16.5×10^{-6} g Th per g, and Th/U ratios ranging between 3.2 and 10.6, and averaging about 7. These results have been confirmed by independent measurements of the U and Th content of these specimens by the emanation method[14] and the γ-ray method.[15] They focus attention on the great importance of a reliable routine method for the determination of the Th content of geological materials, because for Th/U ratios greater than 4 the Th series produces more helium and more thermal energy than the U series, and hence is of greater importance geophysically.

The experimental work and a preliminary theory for thin absorbers which is equivalent to the present general theory was completed at the University of California during the National Research Council Fellowship tenure (1932–1934) of one (E) of the authors. The generous support and encouragement given this program by Professor Leonard B. Loeb is particularly appreciated.

[12] Evans, Phys. Rev. **45**, 38 (1934).

[13] G. Kirsch, Physik. Zeits. **31**, 1017 (1930).

[14] Evans, Finney, Kip and Mugele, Phys. Rev. **47**, 791 (1935); Physics (1935), in preparation.

[15] Evans and Mugele, Phys. Rev. **47**, 427 (1935).