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An Improved Geiger-Counter Arrangement for Determination of Radium Content

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Geiger-counter apparatus in which several counters may be placed in any position relative to each other is described. A circular arrangement of counters to give a counting rate approximately independent of the position of a source near the center of the circle is discussed. The relation of the position of the source to the counting rate of a single Geiger counter is discussed. A method of calculation of the self-absorption of a sample for the Geiger-counter arrangement used is given.

I. Introduction

The usual method of using a Geiger counter for the measurement of radium is to use a single counter and to place the sample at some distance from the counter and compare the counting rate with that of a standard in the same position.[1 2] In the measurement of radium samples by this method, the sample and standard must be accurately placed. If the source is at a distance which is great compared to the dimensions of the counter, the counting rate varies as the inverse square of the distance. At distances close to the counter, the counting rate varies approximately inversely as the distance. For samples of low content, which must be placed close to the counter, the shape and size of the sample (if different from the standard) become important. The different parts of the sample or standard will be at different distances from the counter, and it becomes difficult to determine the effective distance.

A Geiger-counter arrangement is described in which the counting rate is independent of the position of the sample; the only effect of the size and shape of the sample is the change in the percentage of absorption. A method will be shown whereby the absorption can be easily calculated for cylindrical samples.

[1] W. Vogt, Physik. Z. **34**, 79 (1933).

[2] Method of analysis, AOAC, 6th ed, 663 (1945).

II. Apparatus

The arrangement as originally planned was to use six Geiger counters in a ring. However, four in a square proved adequate. A somewhat similar apparatus was described by H. Neufeldt,[3] who used a counter consisting of two concentric cylinders for the cathodes, with four wires stretched between them as anodes. However, this provides a fixed geometry and sensitivity, limiting the use to samples small enough to be placed in the central cylinder, and also to samples of low radium content that can be counted at a reasonable rate.

The apparatus is shown in figure 1. The counters are in a vertical position and may be placed in any arrangement. They are constructed with outer electrodes of 1-in. copper tubing 10½-in. long and ⅓₂-in. wall thickness. The center electrode is 1-mil tungsten wire. The filling of the counters is 10 percent of *n*-heptane and 90 percent of argon at a total pressure of 10 cm Hg. A lead shield ½-in. thick surrounds each counter, and the first amplification stage (6C5 tube) is mounted on one end. The high-voltage lead and supply-voltage leads for each counter tube and first amplification stage are in separate shielded cables. The output of the high-voltage supply of approximately −1,000 v is

[3] Hans Neufeldt, Physik. Z. **30**, 494, (1929).

FIGURE 1.—*View of counters and scaler assembled for use.*

P, power supply and scaling circuit; C, Geiger counters encased in ½-in. lead shields; *S*, position of source in the center of the four counters; *T*, first amplification stage (type 6C5 vacuum tube).

regulated by ten $\frac{1}{25}$-watt neon lamps with a 1-megohm resistor in series. The voltage pulses from the 6C5 tubes are fed to a pulse shaper of a biased multivibrator type and thence to a scaler. The scaler is of the hard-vacuum twin-triode type with 6SL7 tubes, giving a scaling ratio of 128. The circuit for the scaler is shown in a previous paper.[4] The mechanical counter used is a telephone message register. The samples are supported either from above by thread or set on a small pedestal. The size of the square arrangement determines the sensitivity. The smallest square, with lead shields touching, is 6.2 cm on a side. This allows a cylindrical sample 2.5 cm in diameter to be placed in the center. For this arrangement the solid angle subtended by the counters from a point source in the center is approximately one-third of the total 4π solid angle. The counters should be tested individually and should have the same sensitivity. In case of slight differences in sensitivity, the two most sensitive should be placed diagonally opposite to give the least variation in counting rate with position of source

III. Effect of Position of Source

A Geiger counter, if uniformly sensitive over the sensitive area independent of the direction of the gamma rays, has a counting rate proportional to the solid angle at the point source subtended by the area. In figure 2 the points O and O' are the centers of the ends of the cylinder representing the counter cathode, and XX' is a line through the source S and parallel to OO'. Lines DOA and $CO'B$ are arcs of circles generated by rotating O and O' around the axis XX'. Points A, B, C, D are defined by the intersection of these arcs with the tangent planes ASB and CSD. The solid angle subtended by the cylinder is assumed to be bounded by surfaces ASB, BSC, CSD, and DSA. Conical surfaces BSC and DSA are generated by rotating lines OS and $O'S$ around axis XX'. Throughout the greater portion of the solid angle, gamma rays will pass through two surfaces of the cylinder. Gamma rays passing through end regions $HADH$ and $GBCG$ are not included in the solid angle, but these gamma rays pass through only one surface of the cathode and will be approximately compensated by the gamma rays passing through the end regions $EADE$ and $FBCF$, which are within the solid angle and pass through only one surface of the cathode. These end effects become more negligible the longer the counter tube compared to its diameter.

The solid angle W, which is equal to the area included on a unit sphere at S, can be shown to be

$$W=\frac{4b}{(a^2+b^2)^{1/2}}\sin^{-1}\frac{c}{a}, \tag{1}$$

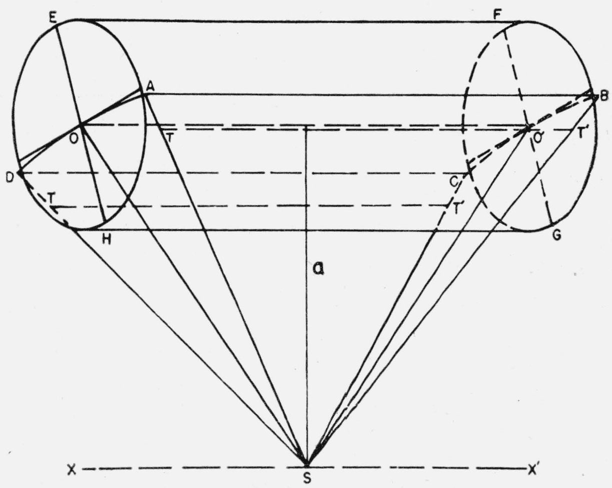

FIGURE 2.—*Diagram of the solid angle at the source, S, subtended by a counter.*

TT' are the tangent lines of the planes ASB and CSD; XX' is parallel to OO' through source S; $DOAS$ and $CO'BS$ are conical surfaces generated by rotating lines OS and $O'S$ around axis XX'.

[4] L. F. Curtiss and F. J. Davis, J. Research NBS **31** 181, (1943) RP1557.

where a is the distance from the source to the axis of counter, b is one-half the length of the counter, and c is the radius of the counter.

Curve B in figure 3 shows the variation of the theoretical relative counting rate with the distance of the source from the counter according to eq 1. This curve is for a counter used in the square arrangement, which has a ratio of half-length to radius (b/c) equal to 11.43. The coordinate scales used are logarithmic. The points shown as crosses are the observed counting rates for a source at the distances indicated and with the counter without its lead shield. The observed points, shown as circles, represent counting rates with the same counter in a ½-in. lead shield. Both sets of points are fitted to the curve at the point $c/b=5$. At close distances the observed points indicated by circles fall below the theoretical curve. This is probably due to the greater absorption by the lead at close distances, where the average path length of the gamma rays in the lead is longer.

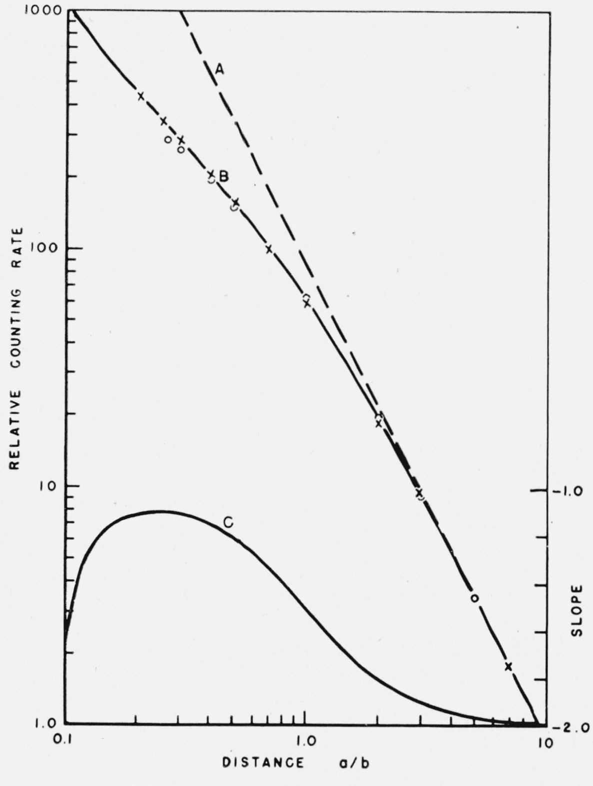

FIGURE 3.—*Counting rate versus distance of source from the counter for the counters used.*

Curve A represents an inverse square law variation. Curve B, theoretical curve for counter ratio of half length to radius (b/c)=11.43. The circles and crosses are observed points for a counter with and without a ½-in. lead shield, respectively. Curve C is the slope of curve B (ordinate on the right).

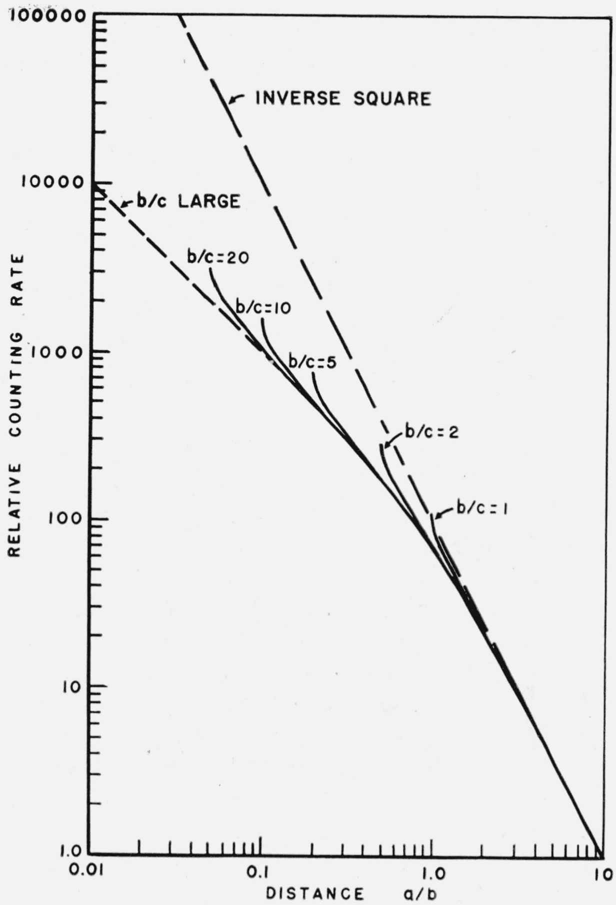

FIGURE 4.—*Counting rate versus distance of source from counter in units of counter half-length.*

a, distance of source from counter; b, half-length of counter; c, radius of counter.

As on log-log coordinates a power function $F=px^q$ has its slope equal to the power q, the slope of the curve B in figure 3 should approach -2 for large values of a/b. A straight line of slope -2 (curve A) is shown for comparison. The calculated slope of the curve according to the function

$$\frac{d \log W}{d \log (a/b)} = \frac{-c}{(a^2-c^2)^{1/2} \sin^{-1} c/a} - \frac{a^2}{a^2+b^2} \qquad (2)$$

is shown by curve C of figure 3. The ordinate of this curve is linear, as shown on the right; the abscissa is the same for the three curves. Any point on the curve then represents the power variation of the counting rate with distance of source. The maximum of the slope curve falls at $a/b=0.24$, with the value -1.10.

In figure 4 are shown several curves for different values of b/c ratio of half-length to radius of

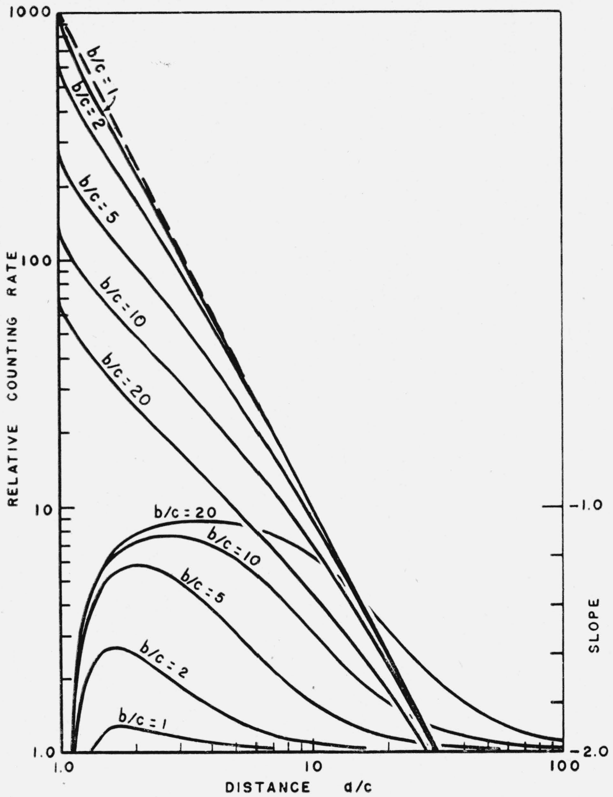

FIGURE 5.—*Counting rate versus distance of source from counter in units of counter radius.*

Upper curves for counters of various ratios of half-length to radius (b/c) Lower curves are the slopes of the upper curves.

the counter. These curves show the theoretical relative counting rate with distance of source in units of the half-length of the counter. It is seen that the smaller the ratio b/c, the more truly the inverse-square law holds. The upper ends of the curves correspond to the position of the source at the electrode. The dotted curved line is the asymptotic line for a counter whose length is very long compared to its diameter. This curve approaches a slope of -1 for small values of a/b.

The curves shown in the upper portion of figure 5 give the theoretical relative counting rate for the source at different distances from the counter in units of the radius (c) of the counter. The curves in the lower portion give the slope of the upper curves, indicating the power relation of the counting rate with distance of source. It is seen that as the source approaches the cathode the slope approaches negative infinity. It would be difficult at very close distances to have an arrangement that would have a counting rate independent of position of the source. For the square arrangement, this region is not accessible because of the ½-in. lead shield, making the closest distance of the source $a/b=0.265$. This is seen to be near the maximum of the slope curve shown in figure 3.

The effect of a vertical displacement of a source within the square arrangement would be the same as for a single counter. Assuming a counting rate N_0 before displacement and a counting rate N_h after a vertical displacement h, the ratio N_h/N_0 derived from eq 1 is

$$\frac{N_h}{N_0}=\frac{(a^2+b^2)^{1/2}}{2b}\left[\frac{b-h}{[a^2+(b-h)^2]^{1/2}}+\frac{b+h}{[a^2+(b+h)^2]^{1/2}}\right]. \tag{3}$$

A horizontal displacement, k, in the direction of a counter of the square arrangement using eq 1 will give a ratio of counting rates

$$\frac{N_k}{N_0}=\frac{(a^2+b^2)^{1/2}}{4\sin^{-1}\frac{c}{a}}\left[\frac{\sin^{-1}\frac{c}{a-k}}{[(a-k)^2+b^2]^{1/2}}+\frac{\sin^{-1}\frac{c}{a+k}}{[(a+k)^2+b^2]^{1/2}}+\frac{2\sin^{-1}\frac{c}{(a^2+k^2)^{1/2}}}{[a^2+k^2+b^2]^{1/2}}\right] \tag{4}$$

where N_0 and N_k are the counting rates before and after displacement k.

The ratio of the counting rates N_m/N_0 where N_0 and N_m are the counting rates before and after a displacement, m from the center and in the direction of the center of one side of the square counter arrangement is given by

$$\frac{N_m}{N_0}=\frac{(a^2+b^2)^{1/2}}{2\sin^{-1}\frac{c}{a}}\left[\frac{\sin^{-1}\frac{c}{(a^2+m^2+\sqrt{2}\,am)^{1/2}}}{(a^2+b^2+m^2+\sqrt{2}\,am)^{1/2}}+\frac{\sin^{-1}\frac{c}{(a^2+b^2-\sqrt{2}\,am)^{1/2}}}{(a^2+b^2+m^2-\sqrt{2}\,am)^{1/2}}\right] \tag{5}$$

If we assume that the inverse-square law for distance holds (a/b large), the first three terms of series representing horizontal displacements give the following approximate formulas valid for small displacements.

$$\frac{N_k}{N_0}=1+\left(\frac{k}{a}\right)^2+3\left(\frac{k}{a}\right)^4 \tag{6}$$

$$\frac{N_m}{N_0}=1+\left(\frac{m}{a}\right)^2-3\left(\frac{m}{a}\right)^4 \tag{7}$$

If we assume that the inverse first power law holds, the first three series terms are

$$\frac{N_k}{N_0}=1+\frac{1}{4}\left(\frac{k}{a}\right)^2+\frac{11}{16}\left(\frac{k}{a}\right)^4 \qquad (8)$$

$$\frac{N_m}{N_0}=1+\frac{1}{4}\left(\frac{m}{a}\right)^2-\frac{13}{32}\left(\frac{m}{a}\right)^4 \qquad (9)$$

A comparison of the results of displacing the source is shown in table 1. The percentage displacement to cause a change of counting rate of 0.1 percent and 1.0 percent is shown in table 2.

TABLE 1.—*Effect of displacement of the source on the counting rate*

Displacement ratio	Ratio of the counting rate after displacement of source over the counting rate before displacement		
	Experimental	Approximate formula	Theoretical
$h/b=0.5$	0.930	-------	0.936
$h/b=1.0$	.763	-------	.770
$k/a=0.2$	1.038	1.045	1.034
$k/a=\ .5$	1.340	1.438	1.333
$m/a=\ .2$	1.028	1.035	1.029
$m/a=\ .5$	1.132	1.062	1.126

TABLE 2.—*Displacement changing the counting rate by 0.1 and 1.0 percent*

Percentage displacement	Percentage displacement required to cause a change of counting rate of 0.1 percent	Percentage displacement required to cause a change of counting rate of 1.0 percent
	Percent	*Percent*
100 k/a, assuming inverse square	3.0	10.0
100 m/a, assuming inverse square	3.0	10.0
100 k/a, assuming inverse first power	6.3	20.0
100 m/a, assuming inverse first power	6.3	20.0
100 h/b at $a/b=0.5$	6.5	20.0
100 h/b at $a/b=1$	5.5	16.5
100 h/b at $a/b=2$	6.0	20.6
100 h/b at $a/b=5$	13.5	42.6

IV. Self-absorption of Sample

To calculate the self-absorption of a cylindrical sample,[5] it is assumed that the gamma rays counted from the element of volume $dxdy$ times the length of sample, shown in figure 6, are parallel to the x-axis, and that the counting rate is independent

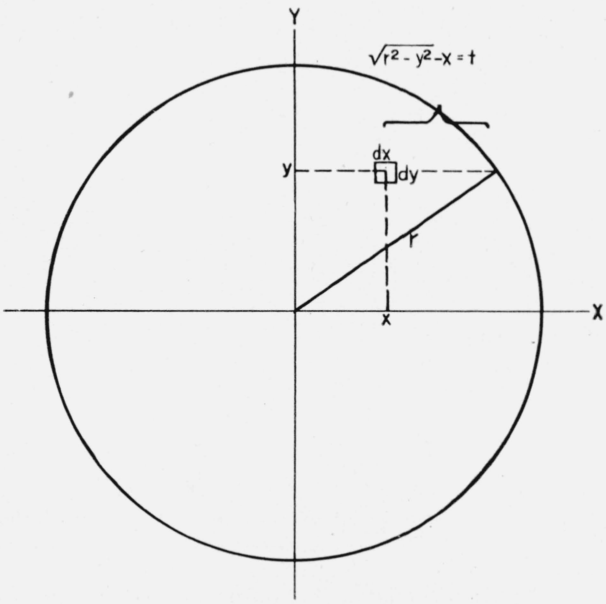

FIGURE 6.—*Diagram for the calculation of self-absorption.*

of the position of the volume element and dependent only on the absorption through the cylinder according to the exponential law $e^{-\mu t}$, where t is the thickness of material through which the gamma ray travels, and μ is the absorption coefficient. If N_0 is the counting rate, assuming no absorption and N_a the counting rate with absorption, then

$$\frac{N_a}{N_0}=\frac{2}{\pi r^2}\int_0^r\int_{-\sqrt{r^2-y^2}}^{\sqrt{r^2-y^2}} e^{-\mu\{[r^2-y^2]^{1/2}-x\}}dxdy$$

$$=\frac{2}{\mu\pi r^2}\int_0^r[1-e^{-2\mu(r^2-y^2)^{1/2}}]dy \qquad (10)$$

Expanding the exponential into a series, we have

$$\frac{N_a}{N_0}=\frac{2}{\mu\pi r^2}\left[r-\int_0^r[1-2\mu(r^2-y^2)^{1/2}+\frac{4\mu(r^2-y^2)}{2!}-\frac{8\mu^3(r^2-y^2)^{3/2}}{3!}+\frac{16\mu^4(r^2-y^2)^2}{4!}-\cdots]dy\right]$$

which reduces to

$$\frac{N_a}{N_0}=1-\frac{8}{3}\frac{\mu r}{\pi}+\frac{1}{2}\mu^2r^2-\frac{32}{45}\frac{\mu^3r^3}{\pi}+\cdots$$

$$=1-0.848\mu r+0.5(\mu r)^2-0.2264(\mu r)^3+\cdots.$$

In figure 7 is plotted the theoretical curve N_a/N_0 for various values of μr. Assuming a relation between density and absorption coefficient μ, one can estimate the absorption of an unknown sample by measuring the density to compute μ and obtain the absorption from figure 7.

[5] C. C. Patterson, J. W. T. Walsh, and W. F. Higgins, Proc. Phys. Soc. **29**, 215 (1917).

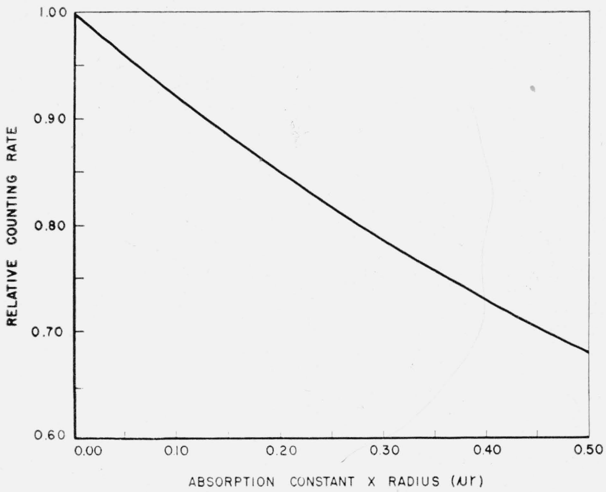

FIGURE 7.—*Relation between self-absorption and radius of ore containers.*

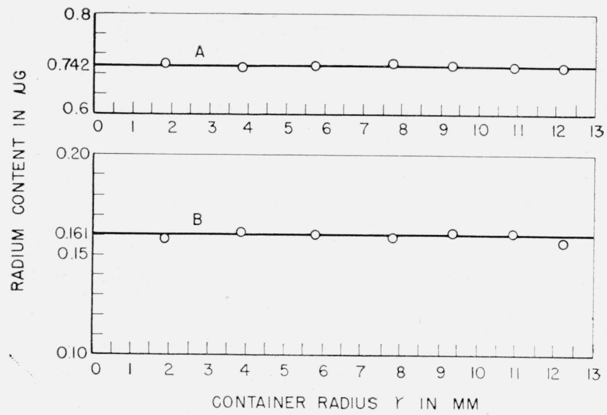

FIGURE 8.—*Observed radium content in containers of various radii.*

A, High-grade ore concentrate; *B*, low-grade ore concentrate.

In figure 8 is shown the observed radium content of a sample of high-grade ore concentrate, using different diameters of cylindrical containers and using the method of correction for absorption just given. The points show good agreement with the horizontal line, giving a value of 7.42×10^{-7} gRa/g. Figure 8 also shows a similar curve for a sample of low-grade ore concentrate giving a value of 1.61×10^{-7} g *Ra*/g. This sample was also determined by the radium emanation method giving 1.62×10^{-7} g Ra/g. This method consists of placing the ore in solution and collecting the radium emanation. The amount of radium emanation is then determined by counting the alpha particles emitted by the radium emanation and its short-life decay products and comparing this count with the emanation from a radium standard solution. The apparatus for this method has been described in a previous paper (see footnote 4). The containers used are glass cylinders, and the absorption of the glass is determined by comparing measurements of a source of radium in the glass and without the glass. Estimates of absorption of the standard are made in the same manner as for unknowns.

V. Performance of Apparatus

In the past year the instrument has proved to be useful for rapid analysis. Very few adjustments have been made since its initial construction. The sensitivity of the apparatus when the counters are as close together as possible is such that 0.1 μg of radium will double the background counting rate. With this value as a minimum and assuming a 100-g sample, the radium content would be 10^{-9} g Ra/g. Estimates can be made of the radium content that gives an increase of the counting rate as low as one-tenth of the background, but no great accuracy of determination can be claimed.

In addition to well-known requirements for statistical accuracy, the following factors limit the accuracy. The ratio of absorption coefficient and density is not a strict constant but may vary as much as 20 percent among light materials. If the absorption of the substance is of the order of 10 percent, the error in the measurement of radium content due to this cause would be of the order of 2 percent. Errors due to assumptions not strictly true in the derivation of the correction for self-absorption would show as a slope or curvature of the horizontal lines in figures 8 and 9. A comparison of determinations made by the method here described with determinations by the radon method showed agreement within 2 percent. Better agreement may be expected if all the materials are of the same nature.

All samples should be sealed for at least 30 days before measuring them unless standard samples of rock or other material are available in a similar condition so that the percentage of radon lost is the same.

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