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## A COUNTING METHOD FOR THE DETERMINATION OF SMALL AMOUNTS OF RADIUM AND OF RADON

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### ABSTRACT

A method for determining small quantities of radon is described, in which the alpha particles from the radon and RaA and RaC are counted in an ion-counting chamber. Details of an arrangement for automatically making a printed record of the hourly totaled count are given. Advantages of this method over that using an ionization chamber with electrometer are discussed.

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### I. INTRODUCTION

The most common method<sup>1</sup> for determining radon quantitatively in amounts between  $10^{-9}$  and  $10^{-13}$  curie is to measure the ion current produced in a gas when the radon mixed with the gas is introduced into an ionization chamber. This method has the following disadvantages, particularly for the smaller amounts: (1) A sensitive electrometer with low electrostatic capacity must be used to measure the ion current. This entails the use of insulators of the highest quality, scrupulously protected from the effects of humidity, and requires that the electrometer be placed in an enclosure at constant temperature. (2) It requires carefully measured calibrating voltages as well as reproducible and constant voltages for knife-edges, in the case of a string electrometer, or for the needle of a quadrant electrometer. These difficulties are not greatly reduced if a low grid-current vacuum tube is substituted for the electrometer. (3) A double ionization chamber is required to eliminate effects of radiation arising outside the chambers. This increases the difficulty of insulation and also increases statistical errors arising from the background or contamination of the chamber. At the same time the double chamber increases

<sup>1</sup> R. D. Evans, *Rev. Sci. Instr.* **6**, 99 (1935). W. D. Urry, and C. S. Piggot. *Am. J. Sci.* **239**, 633 (1941).

the smallest amount of radon that can be measured, since the background of a single chamber enters into the computation of the probable error of the final result with a coefficient of 2. (4) For automatic recording, a practical necessity when dealing with small amounts of radon, a fairly elaborate photographic-recording system is required to give a record of the ion current and calibrating voltages at regular intervals over a period of 10 or more hours. (5) The computation of the results is complicated, involving the measurement of photographic records and corrections because of changes in the capacity of the electrometer at various sensitivities. (6) The calibrating voltages and the sensitivity of the electrometer must be adjusted to the size of each sample.

All these disadvantages can be eliminated by a method which directly counts the alpha particles produced in a single chamber by recording each individual current pulse resulting from the passage of an alpha particle through the gas. In this method no electrometer is required, and the quality of the insulation need not be stringently maintained at the best obtainable. A further pronounced improvement is that only one chamber is required. The probability is very small of any outside source of radiation producing a current pulse inside the chamber comparable to that of an alpha particle. This is limited to dense cosmic-ray showers, which are of very rare occurrence over an area of the magnitude involved here. Finally, the pulses may be enumerated and recorded on a printed record made automatically, so that the record is available immediately after the counting is completed and may be terminated at any time.

The principles involved in counting amplified individual current pulses produced in a gas are well known. For successful application to the measurement of small quantities of radon, it is necessary to adopt special precautions. These arise from the requirement that the current pulses produced by each alpha particle must be sharp so that they may be amplified sufficiently to actuate a recording mechanism. That is, the total current for each particle must be collected and amplified in a brief interval of the order of  $10^{-3}$  second or less. Furthermore, this must be accomplished in an ion chamber having a gas capacity of several liters, with the radon distributed through the gas. This can be done in an atmosphere of pure nitrogen, if the electric field of the chamber is arranged to collect negative ions, which will, in this case, be almost exclusively electrons, following a suggestion by Ortner and Stetter.<sup>2</sup> Pure nitrogen (containing less than 0.001 percent of oxygen) has a very low electron affinity, so that the high mobility of electrons can be utilized even when the ion paths may be as long as 15 centimeters. Under these conditions the current pulses are sufficiently sharp to obtain readily a resolving time of  $10^{-3}$  second.

## II. EXPERIMENTAL METHOD

The preparation of nitrogen of sufficient purity for this method is shown diagrammatically in figure 1. A combustion tube, *G*, filled with reduced copper and surrounded by a furnace, *F*, is connected through the drying bulbs, *B*<sub>1</sub> and *B*<sub>2</sub>, to the ion chamber, *I*, which

<sup>2</sup> G. Ortner and G. Stetter, *Weiz. Anz.* 70, 341, (1933); *Phys. Z.* 35 563 (1934).

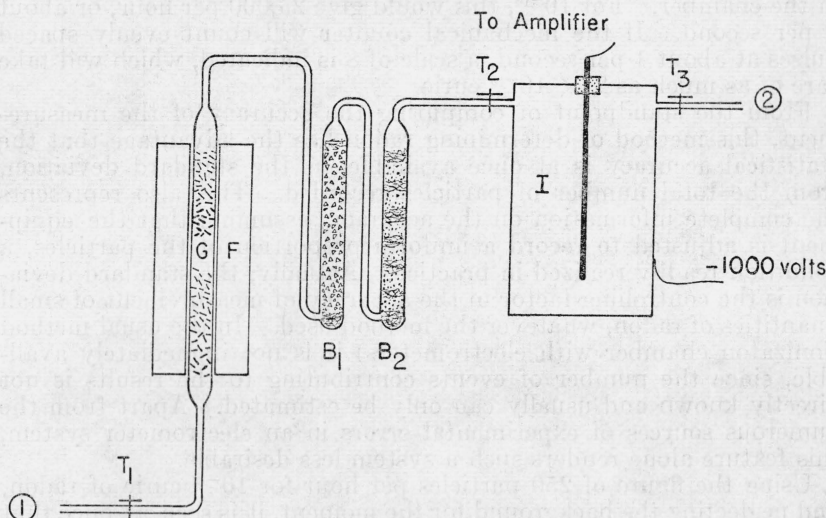


FIGURE 1.—Diagram of system for removing oxygen from radon samples.

*G*, Reduced copper; *F*, furnace; *B*<sub>1</sub>, Drierite; *B*<sub>2</sub>,  $P_2O_5$ ; *I*, ion-counting chamber.

is provided with a connection to a vacuum pump at (2). The drying bulb, *B*<sub>1</sub>, contains Drierite and *B*<sub>2</sub> contains  $P_2O_5$ . In operation the system is evacuated to approximately 0.1 mm Hg through the ion chamber up to stopcock *T*<sub>1</sub>. Then, with the furnace hot (about 500° C) the gas, usually air, containing the radon sample is introduced at (1) with stopcock *T*<sub>3</sub> closed. The rate is adjusted so that all oxygen is taken up by the reduced copper in *G*. If required, additional nitrogen is introduced through the sampling flask into the system to bring the pressure in the ion chamber to atmospheric pressure, as indicated by a manometer, not shown, connected to the chamber. Stopcock *T*<sub>2</sub> is closed and the chamber is ready to begin the counting.

To collect the electrons a negative potential, around 500 to 1,000 volts, is applied to the chamber. This drives the electrons to the collecting rod, *R*, which is connected to the input of a linear amplifier. This amplifier may operate a telephone message register through a scaling circuit. If automatic recording is desired, this can be accomplished most conveniently by using the mechanism of a traffic recorder. This device will print, on the hour or half-hour, the accumulated scale count, together with the time and day of the week, thus giving a complete printed record.

The choice of the scaling ratio is determined by the quantity of radon likely to be measured and by the resolving time of the mechanical recorder. For example,  $10^{-12}$  curie of radon in equilibrium with RaA and RaC will yield at most 266 particles per hour,<sup>3</sup> assuming that one-half of the RaA and RaC particles are counted, since they are solids that will be deposited on the inner surfaces of the chamber. In the work presented here, about 250 per hour actually are counted

<sup>3</sup> Using  $3.7 \times 10^{10}$  alpha particles per second per gram of radium, as given by Braddick and Cave, Proc. Roy. Soc. (London) 121, [A] 367 (1928). On the basis of  $3.5 \times 10^{10}$ , a value resulting from the work of Gladstish and Foyne (Am. J. Sci. 299, 233 (1935)), this number is 252 particles per hour.

in the chamber. For  $10^{-10}$ , this would give 25,000 per hour, or about 7 per second. If the mechanical counter will count evenly spaced pulses at about 1 per second, a scale of 8 is indicated, which will take care of as much as  $2 \times 10^{-10}$  curie.

From the standpoint of computing the accuracy of the measurement, this method of determining radon has the advantage that the statistical accuracy is at once available, as the standard deviation, from the total number of particles recorded. This also represents the complete information on the accuracy, assuming that the equipment is adjusted to record a uniform proportion of the particles, a condition readily realized in practice. Actually, the standard deviation is the controlling factor in the accuracy of measurement of small quantities of radon, whatever the method used. In the usual method (ionization chamber with electrometers) it is not immediately available, since the number of events contributing to the results is not directly known and usually can only be estimated. Apart from the numerous sources of experimental errors in an electrometer system, this feature alone renders such a system less desirable.

Using the figure of 250 particles per hour for  $10^{-12}$  curie of radon, and neglecting the background for the moment, it is seen at once that about 40 hours of observation are required to give a standard deviation of 100 for 10,000 particles, or a statistical accuracy of 1 percent. This points clearly to a frequent source of error in measurements of the intensity of alpha particles, namely, insufficient data. When only 100 particles have contributed to the measurement, whether the quantity measured is an ionization current or a direct enumeration of the particles, the statistical accuracy is  $\pm 10$  percent. When the number is not known directly this easily can be overlooked.

The minimum quantity of radon that can be measured or estimated by the alpha-particle counting method is set by the so-called "natural" rate of emission of alpha particles from the inner metal surfaces of the ion chamber. It is difficult to reduce this rate of emission of particles below 10 per hour per 100 square centimeters of metal surface in a system that counts about 94 percent of all the particles released in the chamber. Bearden<sup>4</sup> reports natural counts as low as 5 and 3 per 100 square centimeters per hour for freshly machined brass, but his figures cannot be compared directly with ours, since efficiency of detection was not determined for his recording system. Metal surfaces that have been stored without particular care have rates of emission that are usually much higher. Various methods have been suggested from time to time to reduce this rate. The most effective is careful machining or sandpapering of the metal to expose a fresh surface just before assembling the chamber. It is evident that in making a determination of radon two measurements are therefore involved. One is in the increased counting rate resulting from the introduction of radon into the chamber, and the other is the natural rate of emission, or background count, of the alpha particles from the walls of the chamber. In the present chamber the area of the interior is approximately 1,400 square centimeters, and the background is about 180 particles per hour, or a little over 11 per 100 square centimeters. As has been pointed out by Urry and Piggot,<sup>5</sup> the observa-

<sup>4</sup> J. A. Bearden, *Rev. Sci. Instr.* 4, 271 (1932).

<sup>5</sup> W. D. Urry, and C. S. Piggot, *Am. J. Sci.* 239, 653 (1941).



tional limit is given by  $0.0036 \times 10^{-12} (B/n)^{1/2}$  curie of radon, where  $B$  is the background count per hour, and  $n$  the number of hours. Thus the observational limit is  $10^{-14}$  curie of radon for a run of 24 hours. This can only be reduced by either decreasing the inner area (that is, the volume) of the ion chamber or by attempting to reduce the rate of emission per unit area. For some purposes a smaller chamber can be used. The chief use of the chambers at present is to determine the radon concentration in the air of workrooms for radium-dial painting and in the expired breath of the workers.<sup>6</sup> A sample of about 2 liters is desirable, therefore a 4-liter ion chamber is used, so that the sample can be flushed by a stream of nitrogen to insure that all the radon passes into the ion chamber. Tests have shown this is accomplished with this chamber, but it would not be true for one of much smaller volume.

### III. APPARATUS

The details of the apparatus found satisfactory are given to illustrate the method as applied to the measurement of samples of air of about 2 liters and containing from  $10^{-13}$  to  $10^{-10}$  curie radon. This covers the usual range of measurements of this type, but as much as  $10^{-9}$  curie can be measured conveniently if the scalers have a ratio of 64:1 or 128:1.

The ionization chamber in which the alpha particles generate the pulses to be counted is shown in section to scale in figure 2. It consists of an inner cylinder,  $C_1$ , and the chamber proper, 9 inches high and 6 inches in diameter. This is surrounded by another metal cylinder,  $C_2$ , 9¼ inches high and 7 inches in diameter, which serves as an electrostatic shield and to protect the operator from the high voltage applied to the inner chamber. The inner chamber is closed by soldering disks in the ends. All joints must be vacuum tight. In soldering, care must be exercised to leave no exposed surfaces of solder in the interior. Lead solders have a high natural rate of emission of alpha particles. An insulator, surrounded by a grounded guard ring, supports the collecting rod,  $R$ , which extends along the axis of the chamber. The plate,  $P$ , fitted to the end of the outer cylinder, supports the shield,  $S$ , for the first amplifier tube and serves as a mounting plate to attach the assembly to a support. The support should be cushioned to reduce vibration of the chamber. Although the chamber is not extremely sensitive to vibrations, some protection is required. Glass stopcocks  $T_1$  and  $T_2$  are waxed into the inner chamber, as shown, for evacuation and for admitting the sample. Since the lower limit of the quantity of radon that can be measured is chiefly determined by the natural background emission from the walls of the chamber, great care is taken in cleaning the inner surfaces of the ionization chamber immediately before the parts were soldered together. The cleaning was accomplished by machining some inner surfaces and polishing others with fine sandpaper known to be free from radioactive contamination. Some abrasives are known to be unsuitable, probably because they are made from materials containing small quantities of radium. Some improvement of the background probably can be achieved by a selection of the metal from which the cham-

<sup>6</sup> See National Bureau of Standards Handbook H27, Safe Handling of Radioactive Luminous Compound (1941).

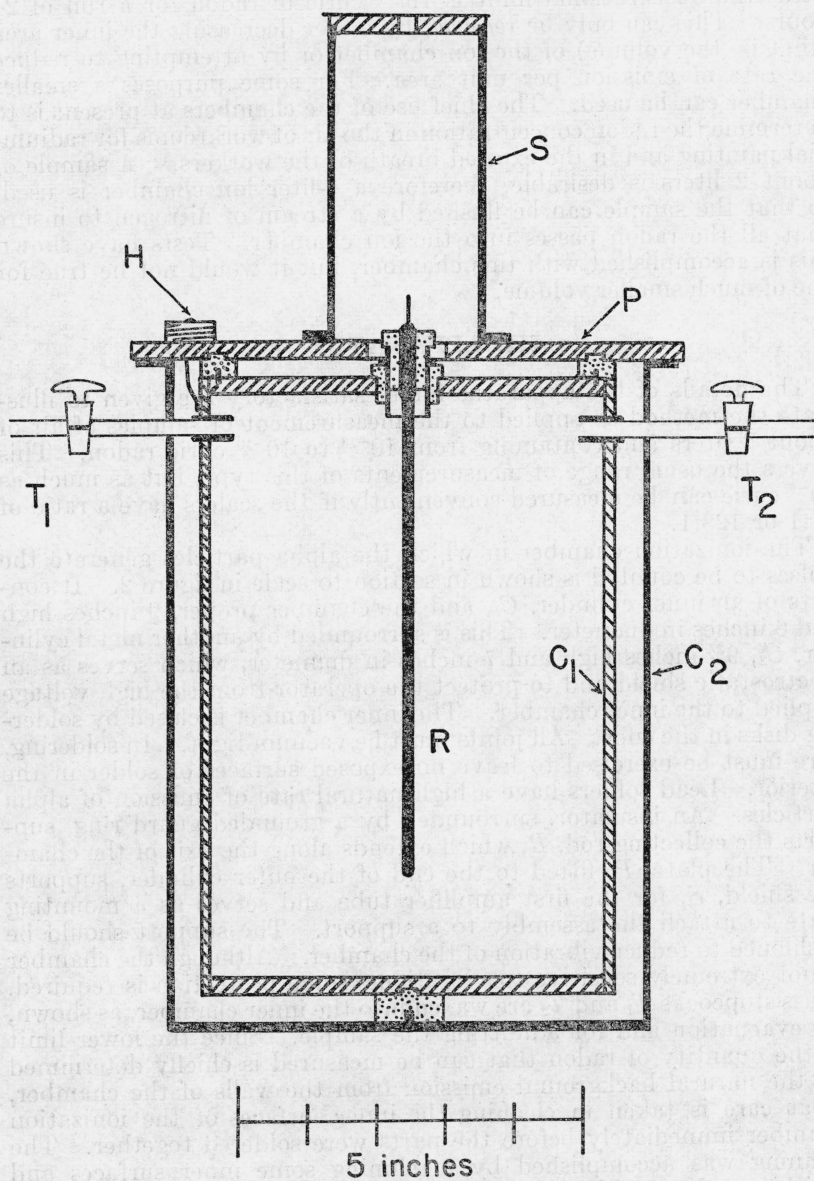


FIGURE 2.—Cross section of ion-counting chamber.

*S*, Shield can for first amplifier tube; *P*, mounting plate; *H*, high-voltage connection; *R*, collecting electrode; *C*<sub>1</sub>, ionchamber; *C*<sub>2</sub>, grounded shield separated from ion chamber by insulators.

ber is made. In this work copper was found to be a little better than brass in this respect. Steel may be even better, but this point was not investigated.

The amplifier used is a resistance capacity coupled in four stages, with inverse feedback. Essentially it is constructed according to the specifications given by Waddell.<sup>7</sup> To eliminate the storage battery the heaters are connected in series, including a dropping resistor of 250 ohms and plugged into a 110-volt d-c line. Using 20  $\mu$ f across the heaters was adequate to remove a-c signal and commutator ripple for 38 tubes of one manufacturer, although some tubes of other manufacture require much more filtering. This amplifier is very stable and is linear over a wide range of amplification. In this labora-

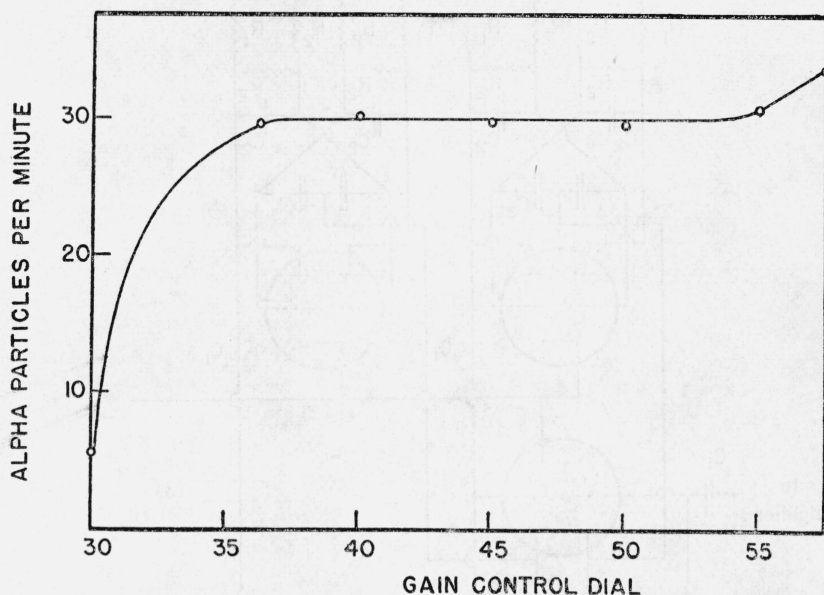


FIGURE 3.—Curve obtained by plotting counting rate for various settings of the gain-control of the amplifier.

tory, amplifiers of this type have been used with little trouble for more than 2 years. One of the most convenient features of this amplifier, when used with an ion chamber, as described in this paper is the ease with which the amplifier gain may be set at the proper point. This is achieved by plotting the counting rate for a definite quantity of radon in the chamber versus the settings on the dial of the gain control. Such a curve is shown in figure 3. There is a considerable range of amplification over which the counting rate is nearly constant. At higher values of gain the amplifier noise begins to affect the scaling circuit and the curve breaks sharply upward. By setting the gain near the middle of this plateau, one is assured of counting all the usable pulses from the chamber and can be fairly confident of counting the same proportion of the total number produced in the chamber under all circumstances.

<sup>7</sup> R. C. Waddell, *Rev. Sci. Instr.* **10**, 311 (1939).

It should be emphasized, however, that such a plateau can only be obtained in an atmosphere of practically pure nitrogen. If a small percentage of oxygen is present, the current pulses are reduced by recombination to such an extent that some disappear completely and others are reduced in size by varying amounts. Under such conditions an increase in counting rate is observed as the gain is increased right up to the "noise level" of the amplifier. This makes it impossible to be certain that a definite percentage of all pulses formed are counted.

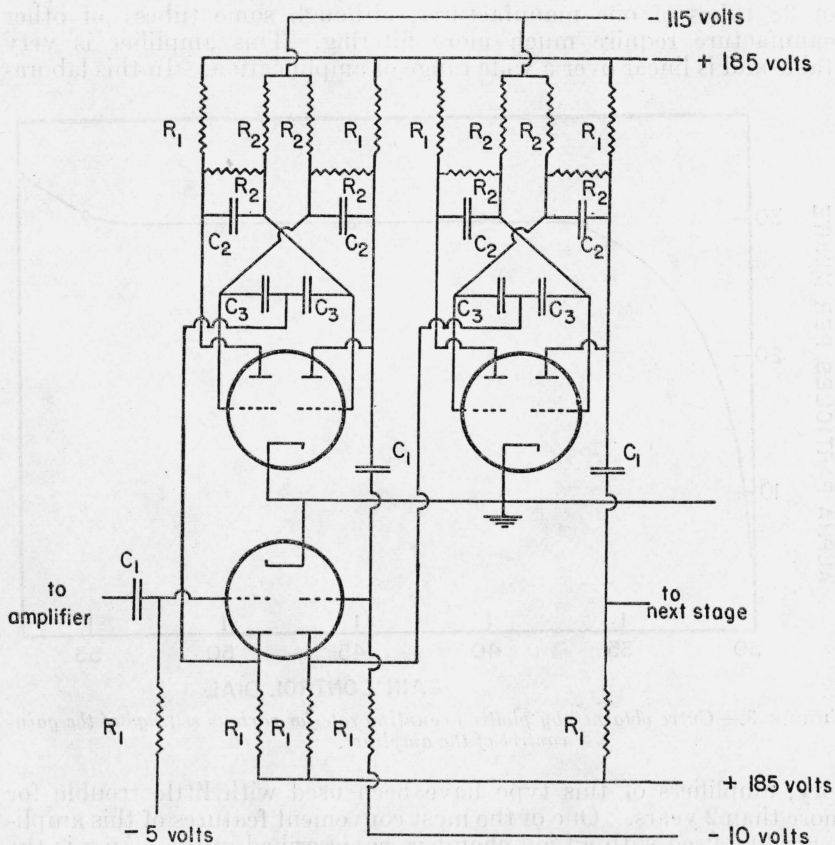


FIGURE 4.—Wiring diagram of the first two stages of the scaling circuit.

$C_1$ , 50  $\mu\text{f}$ ;  $C_2$ , 100  $\mu\text{f}$ ;  $C_3$ , 25  $\mu\text{f}$ ;  $R_1$ , 50,000 ohms;  $R_2$ , 250,000 ohms. Twin triodes, such as 6SC7 or 7F7 have been found satisfactory.

The scaling circuit is composed of twin triode units assembled as shown in figure 4. It has the advantage that it is relatively insensitive, is fairly easy to balance, and once adjusted it continues to operate reliably over long periods of constant use.

The recording unit found most satisfactory is the timing and printing mechanism of a photoelectric-type recorder used to count high-way traffic. It was used without the photocell and amplifier. The mechanism of such a recorder is shown in figure 5. This device prints precisely on the hour the accumulated count from a set of



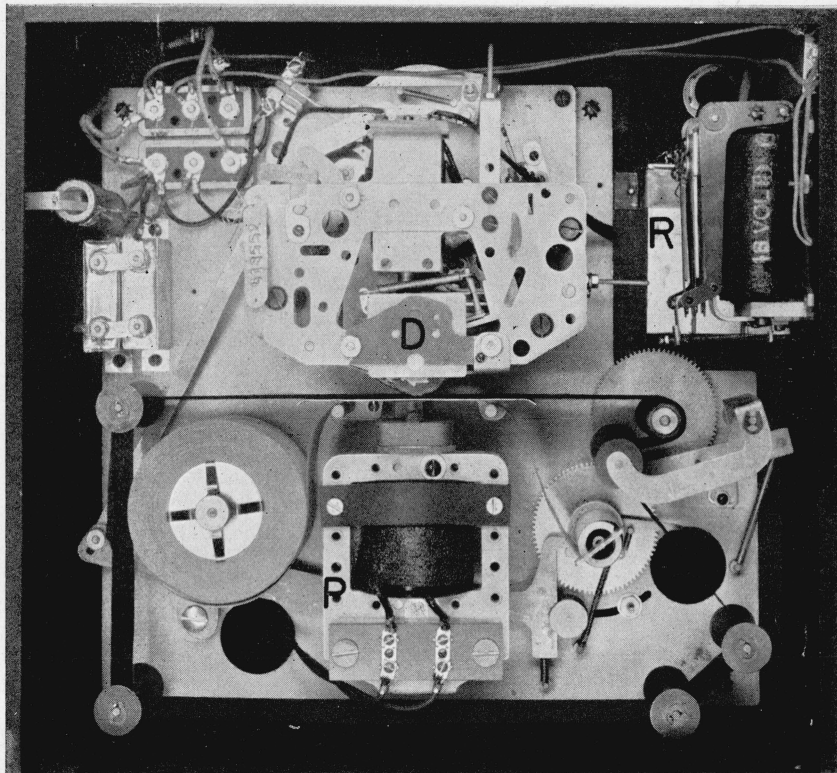


FIGURE 5.—*Interior of traffic recorder.*

*P*, Printing magnet; *D*, counting dials; *R*, relay controlling counting dials actuated by strobotron circuit.

decade dials with raised type faces, recording at the same time the day, the hour, and a. m. or p. m. It uses adding-machine paper and ribbon. It can be made to count on the half-hour or quarter-hour, if desired, but this is not required for the usual radon measurements. It is obvious that this method of recording is far superior to any

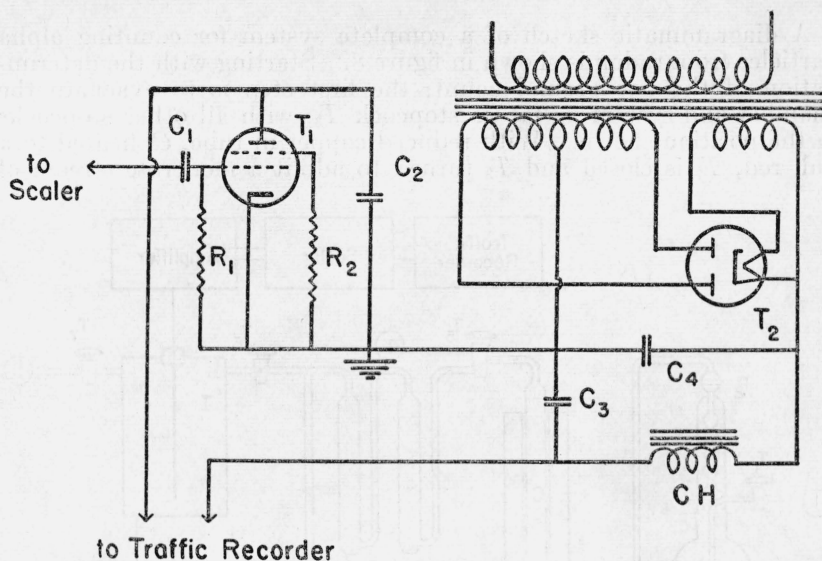


FIGURE 6.—Wiring diagram of strobotron circuit for actuating traffic-recorder counting dials.

$C_1$ , 0.05  $\mu$ f;  $R_1$ ,  $R_2$ , 100,000 ohms;  $C_2$ , 6 to 12  $\mu$ f;  $C_3$ ,  $C_4$ , 4  $\mu$ f;  $C_{11}$ , 12-henry choke;  $T_1$ , strobotron;  $T_2$ , 5Z3.

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TU 3 ..	1 9 8 5 4
TU 4 ..	1 9 8 8 3
TU 5 ..	1 9 9 1 1
TU 6 ..	1 9 9 3 8
TU 7 ..	1 9 9 6 4
TU 8 ..	1 9 9 9 2
TU 9 ..	2 0 0 1 8
TU 10 ..	2 0 0 4 9
TU 11 ..	2 0 0 7 9
W 12 ..	2 0 1 0 4
W 1 ..	2 0 1 3 0
W 2 ..	2 0 1 6 0
W 3 ..	2 0 1 8 6
W 4 ..	2 0 2 1 4
W 5 ..	2 0 2 3 7
W 6 ..	2 0 2 6 3
W 7 ..	2 0 2 9 0
W 8 ..	2 0 3 1 9

FIGURE 7.—Sample record from traffic recorder as printed on adding-machine paper

photographic method. The electric circuit by which the scaler trips the dials on the traffic recorder is shown in figure 6, and a reproduction of a typical record is shown in figure 7.

#### IV. PROCEDURE

A diagrammatic sketch of a complete system for counting alpha particles from radon is shown in figure 8. Starting with the determination of the background count, the first step is to evacuate the chamber and system as far as stopcock  $T_3$ , with all other stopcocks in the position shown. With reduced copper in tube,  $C$ , heated to a dull red,  $T_7$  is closed and  $T_3$  turned to admit a moderate stream of

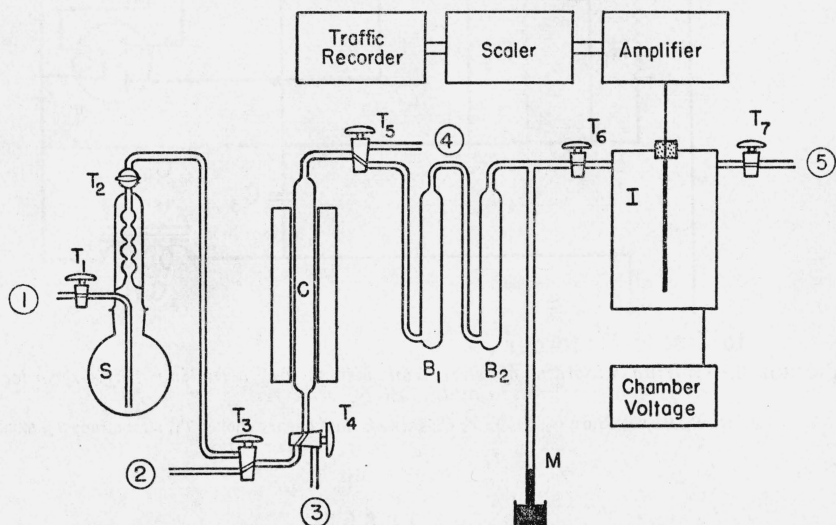


FIGURE 8.—Diagram of complete system for determination of radon.

$S$ , standard radium solution;  $C$ , reduced copper;  $B_1$ , Drierite;  $B_2$ ,  $P_2O_5$ ;  $M$ , open mercury manometer;  $I$ , ion-counting chamber.

nitrogen from the cylinder through the furnace and drying bulbs into the ion chamber. When the open manometer,  $M$ , shows that atmospheric pressure has been established in the ion chamber,  $T_6$  is closed. The voltage is then applied to the ion chamber and the counting process started.

When a sufficient number of pulses have been recorded for a satisfactory determination of the background, the chamber may be calibrated by admitting a known quantity of radon from the standard radium solution stored in bulb  $S$ . This is accomplished by again evacuating the system up to stopcock  $T_2$ , with the furnace hot. Then nitrogen from the cylinder is admitted slowly through  $T_1$  into the bulb,  $S$ , with  $T_2$  and  $T_3$  open to the chamber and  $T_7$  closed. At the same time sufficient heat is applied to bulb  $S$ , to cause the solution to boil. Nitrogen is admitted until the pressure in the ion chamber is again atmospheric. This transfers to the ion chamber the quantity of radon accumulated in the standard solution since it was

last removed. This can be computed from the known quantity of radium in the solution and the elapsed time.  $T_6$  is closed and the calibration count is taken. This calibration need not be made for every run, since in the absence of any alteration of the chamber the rate of emission per curie of radon will remain constant. However, occasionally checks are desirable. The measurement of an unknown may now be made by evacuating the chamber as before and attaching the flask containing the unknown at  $T_3$ , as indicated.  $T_3$  is turned to connect the flask to the furnace and a flushing tube is connected to the supply of nitrogen from the cylinder. By closing  $T_7$  and opening  $T_6$ , the ion chamber is filled to atmospheric pressure with oxygen-free nitrogen, since the oxygen originally in the sample flask is removed in the furnace. At the same time the radon in the sample flask is also transferred to the ion chamber. Then  $T_6$  is closed and the alpha particles from the sample may be counted.

A combustion tube about 30 millimeters in diameter and containing a column of reduced copper 50 centimeters high will completely remove the oxygen from 200 to 300 liters of air before it needs reduction. To rejuvenate the copper, stopcocks  $T_5$  and  $T_4$  are turned to permit hydrogen from the cylinder to circulate through the combustion tube with the furnace hot. The water generated will be discharged through the open branch of  $T_4$ , chiefly as steam. As soon as the copper is completely reduced the hydrogen is cut off, and the furnace is ready for further use. When in daily use the copper requires reduction about every 3 or 4 weeks.

Although the number of alpha particles per curie will remain fairly constant for a given chamber, the background count slowly changes. This change, when in continuous use, is an increase, depending on the amounts of radon admitted to the chamber and the length of time it remains. For instance, a  $10^{-9}$  curie radon sample in the chamber for 24 hours would eventually raise the background about 5 counts per hour, but it would grow according to the growth curve of RaF, that is gain only about  $2\frac{1}{2}$  counts per hour after 140 days. Under certain circumstances it may be a decrease. Therefore, frequent counts of the background are desirable. Immediately after running a heavy sample, if a background is run, a gradual rise in background rate during the run may be noticed, possibly due to radon coming from the walls, which may have been absorbed there during the time the sample was in the chamber. This amount, however, is a small fraction of a percent of the sample, so that if separate chambers are reserved for small and large samples, no appreciable error due to this effect is encountered.

The method of computation is quite simple. The usual procedure is to disregard about the first 3 to 5 hours of the record after filling the chamber to permit RaA and RaC to grow to equilibrium and the active deposit of the previous sample to decay. In 3 hours the activity of the deposit will fall to about 1 percent of the sample value, in 5 hours, less than 0.05 percent. On the other hand, in 3 hours the growth curve of radon plus the products of disintegration will be within 1 percent of equilibrium. It is not always necessary to disregard the first part of the record, and in some cases with large samples where a short run may be desirable, the result can be calculated an hour after filling the chamber.



If we disregard the first 3 or so hours, it is only necessary to calculate the average count per hour, subtract the background, and divide by  $250 \times 10^{12}$ , giving the average amount of radon in curies during the run. Usually what is desired, however, is the initial amount of radon. If we let  $A_t$  equal the average of an exponential decay function,  $e^{-\lambda t}$  of duration,  $t$ , then

$$A_t = \frac{1}{t} \int_0^t e^{-\lambda t} dt = \frac{1 - e^{-\lambda t}}{\lambda t},$$

and to correct for decay during the run, divide the average radon by  $A_t$ , and we have the initial amount at the time of the first reading. Since most runs are overnight and for an integral number of hours, a short table of correction factors,  $A_t$ , can be quickly computed with a slide rule.

On a background run the standard deviation of the count per hour from the record is

$$\sigma = \sqrt{\frac{\sum_{i=1}^n \delta_i^2}{n-1} - 10.50},$$

where  $\delta_i$  is the deviation of the  $i_{th}$  hour count from the mean and  $n$  the number of hours. The correction term 10.5 is entered to take care of the variation of the count due to the scaler, which only counts intervals of 8. The difference between this calculated value and the theoretical value  $\sigma_{th}$  (equal to the square root of the average count per hour) should, on the average, be less than  $\sigma/\sqrt{2n}$ , which is the standard deviation of  $\sigma$ .

Since the record actually shows the scaled count, it is simpler to calculate the standard deviation of its scaled count per hour by the formula

$$\sigma_{sc} = \sqrt{\frac{\sum_{i=1}^n \Delta_i^2}{n-1} - 0.164},$$

where  $\Delta_i$  is the deviation of the  $i_{th}$  hour scaled count from the mean, and  $0.164 = 10.5/8^2$ . This calculated value should fall on the average within

$$\frac{\sigma_{sc}}{\sqrt{2n}} \text{ of } \frac{\sigma_{th}}{8}.$$

The hourly differences taken from three records of consecutive background runs are given in table 1 with the dates of the observations. In this table, columns marked (a) are the hourly differences taken from the traffic-recorder printed record; columns marked (b) are the deviations from the mean, taken as nearest whole number; and columns marked (c) are these deviations squared. In the corresponding horizontal rows the total scaled count (sums of columns (a)), the mean scaled count,  $M$ , and the total actual count are computed, with the corresponding background counts per hour,  $B_1$ ,  $B_2$ , and  $B_3$ . The figures indicating the errors are the theoretical standard deviations equal to the background divided by the square root of the total actual count. The sums of the squares of the deviations from the mean scaled count,  $M$ , are designated as  $\mu_2$ , and are computed from the formula  $\mu_2 = v_2 - nd^2$ , where  $v_2$  is the sum of the squares of the deviations about any convenient number  $A$  (taken as 13),  $n$  is the number of hours, and  $d = A - M$ . The subtracted quantity 0.16 is

the correction term for the variation of the count due to the scaler, which records only in units of 8. The standard deviation of the scaled count  $\sigma_{sc}$  ( $=1.89 \pm 0.37$  in the first set of observations) is computed in each case. The figure for the "error" is found by dividing the value obtained by  $\sqrt{2n}$ , i. e., ( $0.37=1.89/\sqrt{2n}$ ).

TABLE 1.—Data for determining uniformity of background

Hours	Date of determination								
	June 3			June 11			June 17		
	a	b	c	a	b	c	a	b	c
1.....	11	2	4	12	1	1	14	1	1
2.....	13			11	2	4	12	1	1
3.....	12	1	1	13			11	2	4
4.....	13			14	1	1	11	2	4
5.....	11	2	4	13			14	1	1
6.....	12	1	1	14	1	1	12	1	1
7.....	12	1	1	11	2	4	13		
8.....	16	3	9	13			12	1	1
9.....	9	4	16	14	1	1	13		
10.....	12	1	1	14	1	1	13		
11.....	14	1	1	13			16	3	9
12.....	13			13			12	1	1
13.....	16	3	9	12	1	1	13		
14.....							11	2	4
15.....							14	1	1
Total scaled count.....	164			167			191		
Mean (M).....	12.6			12.8			12.7		
Total actual count.....	1,312			1,336			1,528		
Background.....	{ $B_1$ $100.8 \pm 2.8$ }			{ $B_2$ $102.4 \pm 2.8$ }			{ $B_3$ $101.9 \pm 2.6$ }		
$v_2$ .....			47			14			28
$nd^2$ .....			2.18			0.52			1.50
$\mu_2$ .....			44.82			13.48			26.50
$\mu_2/n-1$ .....			3.73			1.12			1.89
Corr.....			0.16			0.16			0.16
$\sigma_{sc}^2$ .....			3.57			.96			1.73
$\sigma_{sc}$ .....			$1.89 \pm 0.37$			$0.98 \pm 0.19$			$1.31 \pm 0.24$
$\sigma_{th}$ 8.....		$\frac{\sqrt{100.8}}{8}=1.26$			$\frac{\sqrt{102.4}}{8}=1.26$			$\frac{\sqrt{101.9}}{8}=1.26$	

Although the standard deviations computed from the data fall near the theoretical values, only in one set of figures (June 17) do the values coincide. This is to be expected from the statistical definition of the standard deviation. On comparing the three values of the background,  $B_1=100.8 \pm 2.8$ ,  $B_2=102.4 \pm 2.8$ , and  $B_3=101.9 \pm 2.6$ , we find that the background does remain constant within the standard deviation over a reasonable length of time, and therefore the determinations of this quantity are subject to no greater fluctuations than those of the sample of radon.

For a sample run of appreciable size the calculation of the standard deviation should take into account the decay of radon. The square of the standard deviation due to exponential decay is

$$\sigma_d^2 = 1/t \int_0^t e^{-2\lambda t} dt - A_i^2 = \frac{1 - e^{-2\lambda t}}{2\lambda t} - A_i^2 = A_{2t} - A_i^2.$$

Therefore the calculated standard deviation of scaled count per hour should be

$$\sigma_{sc} = \sqrt{\frac{\sum_{i=1}^n \Delta_i^2 - \sigma_d^2 \left(\frac{R}{8}\right)^2}{n-1}} - 0.164,$$

where  $R$  is the average count per hour due to radon and its decay products. A table of  $\sigma_d^2$  can be quickly calculated from an accurate table of values of  $A_i$ . Since the half-life of RaA is short compared to an hour, its alpha particles cannot be treated as independent events; therefore, the theoretical standard deviation of the total count should be the square root of the sum of the background and  $4/3$  the radon count, and the theoretical standard deviation of the count per hour should be  $\sigma_{th} = \sqrt{B + 4/3R}$ , where  $B$  is the background count per hour. If the background is small compared to the radon count, this will amount to about a 15-percent increase in the standard deviation.

The record shown in figure 7 is for an air sample taken at 4:00 p. m., on April 27, 1943, and transferred to the chamber at 2:00 p. m. on May 4, 1943. If we start the computation at 5:00 p. m., the radon will be in equilibrium. The average count per hour from 5:00 p. m. to 8:00 a. m. is  $217.6 \pm 3.8$ . The background for this chamber is  $B = 125.0 \pm 2.9$ , giving a net count of  $R = 92.6 \pm 4.8$  per hour. The correction due to decay during the run is  $A_i = 0.945$ , and for the decay from time of collection to 5:00 p. m. on May 4, 1943, is 0.279. The volume of the flask is 2.10 liters. Therefore, the amount of radon at the time of sampling is

$$\frac{92.6 \times 10^{-12}}{250 \times 2.10 \times 0.945 \times 0.279} = 0.668 \pm 0.035 \times 10^{-12} \text{ curie/liter.}$$

If we wish to check the statistical distribution by comparing

$$\sigma_{sc} = \sqrt{\frac{\sum_{i=1}^n \Delta_i^2}{n-1} - \sigma_d^2 \left(\frac{R}{8}\right)^2} - 0.164.$$

with

$$\frac{\sigma_{th}}{8} = \frac{\sqrt{B + 4/3R}}{8},$$

the values are as follows:

$$\frac{\sum_{i=1}^n \Delta_i^2}{n-1} = \frac{64.4}{14} = 4.60$$

$$\sigma_d^2 \left(\frac{R}{8}\right)^2 = 0.000955 \left(\frac{92.6}{8}\right)^2 = 0.128.$$

Therefore

$$\sigma_{sc} = \sqrt{4.60 - 0.164 - 0.128} = 2.07 \pm \frac{2.07}{\sqrt{2n}} = 2.07 \pm 0.38$$

and

$$\frac{\sigma_{th}}{8} = \frac{\sqrt{B + 4/3R}}{8} = \frac{\sqrt{125 + 123}}{8} = 1.97,$$

showing that the two standard deviations compare within 0.38, as they should.

It is obvious that the system described may be modified to measure radon from solutions containing unknown amounts of radium or from solids by fusion in a vacuum furnace. It thus provides a simple and reliable method of making all types of determinations of radon. It is, moreover, particularly effective when the amounts are small.

WASHINGTON, June 1, 1943.





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[Continued on p. 4 of cover]

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