A Balance Method for Comparison of Quantities of Radium and Some of its Applications

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XX. *A Balance Method for Comparison of Quantities of Radium and Some of its Applications.* *By Prof.* E. RUTHERFORD, *F.R.S., and* J. CHADWICK, *B.Sc., University of Manchester.*

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IN many radio-active measurements it is now of great importance to determine with accuracy the quantities of radium or of radium emanation employed in various investigations. The International Committee appointed by the Congress of Radiology at Brussels, in 1910, have now in preparation an International Radium Standard, which will contain a known weight of pure radium salt. All quantities of radium will ultimately be expressed in terms of this International Standard. It is consequently of great importance to develop reliable methods of comparing quantities of radium accurately in terms of the primary standard. The most suitable method for this purpose is to compare the γ-ray activities of the two specimens. If the radium is enclosed in a sealed tube, the γ-ray activity reaches a practical maximum after two months, and the intensity of the penetrating γ-rays which are emitted serves as a definite measure of the quantity of radium present. The greater part of the γ-rays are emitted not by radium itself but by radium C, and recent investigations by Moseley and Makower * have shown that under ordinary conditions about 11·5 per cent. of the total γ-ray activity of radium is to be ascribed to radium B. The γ-rays from the latter are on the average much less penetrating than those from radium C, and are completely absorbed by a lead screen 2 cm. thick.

In making a comparison, it is essential to know that the radium compound contains only radium and its products and no meso-thorium or radio-thorium. Both the latter substances emit γ-rays of about the same penetrating power as those given out by radium, and without a special examination it is difficult to tell whether the thorium products are absent. Since meso-thorium and radium are always separated together, and are chemically closely allied, it is impossible to isolate pure radium compounds from minerals containing both uranium and thorium. Fortunately the uraninite deposits at Jochimstahl contain only a trace of thorium, so that the radium separated

* Moseley and Makower, "Phil. Mag.," January, 1912.

from this ore can be obtained practically free from meso-thorium.

The γ-ray activity of radium preparations has been usually compared by electroscopic methods. The electroscope is either composed of lead plate about 3 mm. thick or is completely surrounded by a lead cover of this thickness. Under such conditions the primary β-rays are completely stopped by the lead and the ionisation in the electroscope is due to the more penetrating γ-rays and to the β-radiation to which they give rise. The rate of movement of the gold leaf of the electroscope between two fixed points of the scale of the observing microscope is taken as proportional to the intensity of the γ-radiation. With a well-constructed electroscope it is possible to compare approximately equal quantities of radium with a probable error of not more than $\frac{1}{2}$ to $\frac{1}{3}$ per cent. If,

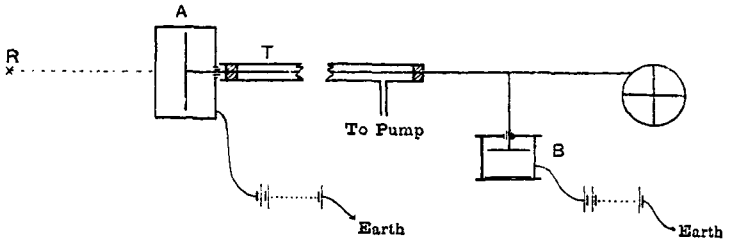

FIG. 1.

however, the quantities of radium differ largely in amount—for example, in the ratio of 10 to 1—the ionisation currents in the electroscope for a given distance of the radium from the electroscope differ in this ratio, and the comparison cannot be made with the same accuracy as in the case of nearly equal quantities. The accuracy of the method obviously depends upon the certainty of complete saturation of the ionisation in the electroscope in the two cases. Unless a special investigation be made to determine the variation of current with voltage under the experimental conditions it is difficult to be certain that saturation has been reached. Any method of comparison of the intensity of the γ-rays from two preparations by the direct measurement of the ionisation they produce is subject to the same limitations.

In order to avoid this difficulty the writers have developed a balance method for accurately comparing quantities of radium by their γ-ray effects. The general principle of the method is clearly seen from Fig. 1. The γ-rays from the radium prepara-

tion at R enter the ionisation chamber A. The saturation ionisation current in A is balanced against an equal but opposite ionisation current due to a standard preparation of uranium oxide in the vessel B. The accuracy of the balance is tested by a Dolezalek electrometer. In practice, the ionisation current due to B is kept fixed, and the standard radium preparation is moved until a balance is obtained. The standard is then removed to a distance, and a balance is again obtained for the second preparation. Since the intensity of the γ-rays from a point source of radium fall off nearly inversely proportional to the square of the distance, the relative γ-ray activity can be deduced from the distances of the radium from the vessel A in the two cases.

The construction of the ionisation chamber is shown in Fig. 2A. It consists of a lead cylinder 2 cm. thick 10 cm. long

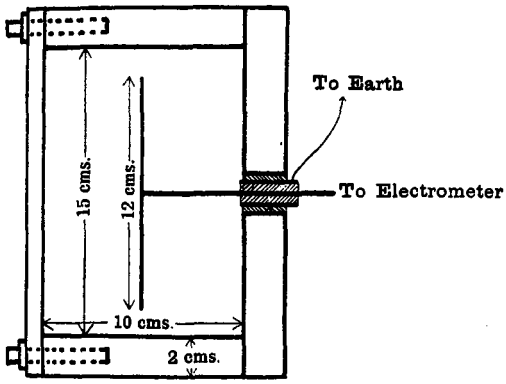

FIG. 2A.

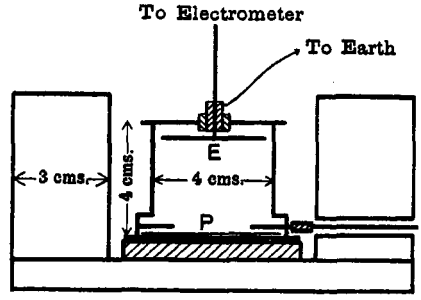

FIG. 2B.

and 15 cm. internal diameter. One end of the cylinder is closed by a lead plate 2 cm. thick, the other, where the γ-rays enter, by a lead plate whose thickness can be varied between 1 mm. and 2 cm. of lead. The plate ordinarily employed is of thickness 1 cm. The interior electrode consists of a circular aluminium plate 1 mm. thick and 12 cm. diameter. This is insulated from the cylinder by a sulphur stopper, provided with a guard ring in the usual manner.

The construction of the uranium vessel B is shown in Fig. 2B. It consists essentially of a brass cylinder 4 cm. high and 4 cm. diameter. On the bottom is placed a metal plate, P, which is covered with a uniform film of uranium oxide prepared by McCoy's method. Above the plate is an iris

diaphragm of the type used in microscopes. The aperture of the diaphragm could be varied by means of an attachment between 0·5 cm. to 3 cm. By this means the ionisation in B can be varied over a considerable range. The base of the vessel is screwed on to an ebonite block which is fastened to a lead plate 1 cm. thick. The vessel is protected from external γ-rays by surrounding it by a lead cylinder 6 cm. high and 3 cm. thick. The interior electrode E is a circular copper plate insulated by a sulphur stopper provided with a guard ring. The electrode in A (Fig. 1) is connected with the electrometer by a wire passing through a thick lead tube, T, exhausted by means of a water pump to a pressure of about 2 cm. In this way the ionisation of the air surrounding the wire due to an external source of γ-rays was made very small. A was kept charged to -120 volts and B to $+200$ volts. The electrometer needle was charged through a phosphor bronze suspension to 200 volts.

The radium preparation was mounted on the carriage of a photometer bench, symmetrically in regard to the face of the ionisation chamber, and was moved along the graduated bench until a balance was obtained. Suppose r_1 is the distance of the balance point from the front surface of the air in A, and $r+a$ the distance from the surface of the back plate, a being the depth of the ionisation chamber. The intensity of the γ-rays from the radium at a distance, r, from the source within the ionisation chamber is proportional to $\frac{e^{-\mu_1 d} \cdot e^{-\mu r}}{r^2}$, where μ_1 is the average coefficient of absorption of the γ-rays in the lead plate of thickness d and μ the absorption of the γ-rays by the air. The actual ionisation observed in the chamber A arises in all probability mainly from the β-rays set up by the γ-rays on the front and back plates. Our knowledge of the amounts and distribution of these excited β-rays is at present too inadequate to calculate the intensity of the radiation at each point of the ionisation chamber with certainty. If, however, the distance a is small compared with r it is to be expected that the ionisation in the chamber of depth a should be very nearly proportional to $1/r(r+a)$. It will be seen later that this relation is borne out by experiment. Assuming this formula, it is a simple matter to compare the γ-ray activities of two quantities of radium by determining the distances r_1, r_2 required for a balance. Suppose the standard preparation containing S milligrammes of radium balances at a distance r_1 from the inner surface of the front plate, and the test preparation containing R milligrammes

of radium balances at a distance r_2. Since when a balance is obtained, the ionisation in the chamber is equal for both preparations, and the absorption by the lead is the same in both cases—

$$\frac{Re^{-\mu r_2}}{r_2(r_2+a)} = \frac{Se^{-\mu r_1}}{r_1(r_1+a)}$$

or

$$R/S = \frac{r_2}{r_1} \cdot \frac{r_2+a}{r_1+a} e^{-\mu(r_1-r_2)}.$$

For an accurate determination it is necessary to know the absorption of the γ-rays by air. This has been determined as accurately as possible by one of the authors, and an account of the experiments is given in an accompanying paper. If the γ-rays pass through 3 mm. of lead before entering the ionisation chamber the value of μ for air at normal temperature and pressure is given by $\mu=0{\cdot}0000624$ (cm.)$^{-1}$, and $\mu=0{\cdot}000059$ (cm.)$^{-1}$ at standard pressure and 15°C. Suppose, for example, the preparation R balances at a distance 1 metre beyond the standard preparation S at a temperature 15°C., then $e^{-\mu(r_1-r_2)}=e^{0{\cdot}0059}$ $=1{\cdot}0059$. The correction for absorption is thus 0·59 per cent.

Since the absorption is certainly correct within 5 per cent., the maximum error introduced by the correction in the case considered is not greater than 3 parts in 10,000.

If the standard and test preparations are contained in glass tubes of unequal thickness, it is, of course, necessary to correct for the absorption of the γ-rays in glass, and in some cases by the radio-active preparation itself. This latter correction is in general small, and can easily be applied if the weight and nature of the material are known.

It has been found that when the γ-rays have passed through 1 cm. of lead the coefficient of absorption of the γ-rays in ordinary soda glass is given by 0·117 (cm.)$^{-1}$. The correction to be applied for $\frac{1}{10}$ mm. extra thickness of wall is consequently 0·12 per cent., and for an extra thickness of 1 mm. = 1·2 per cent. In properly constructed standards contained in thin-walled glass tubes the correction is in general a fraction of a per cent.

It is, of course, necessary that the ionisation in the smal vessel B, due to the γ-rays from the radium, should be negligibly small in comparison with the ionisation due to the uranium. This can readily be arranged by making the volume of B small compared with that of A. The ratio of volume of

B to A used was 1 to 35. In addition, the γ-rays had to penetrate 5 cm. of lead before entering the ionisation vessel B. The relative effect of the γ-rays on the ionisation of B is clearly greater the larger the quantity of radium under comparison. In the experiments the greatest distance of the balance point from A was about 3 metres, and the distance between A and B was 4 metres. Taking the various factors into consideration, it can be simply calculated that the maximum ionisation due to the γ-rays in B is not more than $\frac{1}{3000}$ of the ionisation due to the uranium, and may consequently be neglected.

Test of Distance Law.—We have seen that the ionisation in the vessel A should vary approximately as $\frac{e^{-\mu r}}{r(r+a)}$, where r and a have been already defined. The accuracy of this relation has been tested experimentally in the following way : Four small glass tubes were each filled with about 20 millicuries of emanation. After radio-active equilibrium was reached, the balance was obtained for each of the four tubes separately and then all together. Correction was made for the decay of the emanation, and consequently of the γ-ray activity during the interval of measurement. Since the time for the five observations was about one hour, the correction for the decay of the emanation was small, and could be made with accuracy. If r_1, r_2, &c., be the distance of balance for the four tubes separately, and r for the four tubes together, the values $\Sigma r_1(r_1+a)e^{\mu r_1}$ for the four tubes should equal $r(r+a)e^{\mu r}$, if the distance law assumed is correct.

The agreement between the values is shown in the following table for a number of separate experiments. In some cases the ionisation in B was varied ; in others, the ionisation was kept constant, but the distance of balance from A became smaller owing to decay of the emanation with time.

Approximate distance of single tubes from ionisation chamber.	$\Sigma r_1(r_1+a)e^{\mu r_1}$ for four tubes.	$r(r+a)e^{\mu r}$ for four tubes together.
120 cm.	66,860	66,740
,,	66,090	65,900
,,	65,550	65,400
110 cm.	52,140	52,170
,,	51,210	51,170
70 cm.	22,910	22,860
,,	19,550	19,580
62 cm.	18,490	18,440

At 45 cm. from A the value corresponding to the second column was found to be 0·4 per cent. less than in the third. At this distance, therefore, the deviation from the formula becomes appreciable. Provided the balance is not obtained for distances less than 60 cm., for the particular apparatus employed, the formula is seen to agree with experiment to 1 part in 400.

Another simple method of testing the distance law is to compare the ratio of the γ-ray activity of two radium preparations which differ in considerable ratio—1 to 4, for example—for different values of the ionisation in B. If the distance law is correct the ratio of the γ-ray activities of the two preparations should agree whatever the distance of balance.

Advantages of this Method.—The balance method of comparing γ-ray activities has numerous advantages. In the first place, since the ionisation in the testing vessel is a constant when a balance is obtained, no error due to possible lack of saturation arises. There is no correction to be applied for natural leak provided it does not change during the time of observation. Since comparisons are made when the radium is at considerable distance from the ionisation chamber, the dimensions or shape of the tube containing the radium has little influence, and the correction for distribution is, in most cases, negligible.

In consequence of this the γ-ray activity of radium preparations contained in vessels of different size and shape can be readily compared. If the actual quantity of radium in the tube is to be determined in terms of the standard, correction, of course, must be made for the absorption of the γ-rays in the walls of the containing vessel and in the radio-active material itself.

The method is an ideal one for comparison of nearly equal quantities of radium, such as, for example, will be used in comparing duplicate standards in terms of the primary. Under such conditions, the balance distances are practically the same for the two preparations. Possible errors in the distance law assumed or in the amount of absorption of γ-rays by air do not appreciably affect the accuracy of the comparison. If the two standards to be compared are enclosed in tubes of equal thickness and contain radium of about equal purity, it will be a simple matter to determine the relative value of the standards to 1 part in 1,000 or even closer.

We have seen that the distance law breaks down for a dis-

tance smaller than 50 cm. If, however, the apparatus be required to determine small quantities of radium—*e.g.*, $\frac{1}{10}$ milligramme—it will be necessary either to reduce the ionisation in B or to calibrate the apparatus for smaller distances. The latter can be done by observing the alteration of distance of balance for a tube containing emanation as it decays with time. Assuming the period of decay of the emanation (3·85 days) a fairly accurate calibration curve can be obtained.

The investigations of J. A. Gray * have shown that γ-rays are excited by the passage of β-rays through matter. The source of β-rays in his experiments was radium D + radium E. The authors have found by the balance method that the β-rays from radium in falling on matter also give rise to γ-rays, but in amount small compared with the primary γ-rays from the active matter itself. Since the thickness of glass tubes enclosing the radium preparation is in general not sufficient to absorb all the β-rays, the escaping β-rays no doubt produce some γ-rays in the surrounding air. This possible error in the measurements can be corrected for by surrounding the radium tube with a sufficient thickness of a substance of small density, like glass or aluminium, to absorb all the β-rays. Gray has shown that the excitation of γ-rays is small for light substances, and is approximately proportional to the atomic weight of the material.

Determination of Quantities of Emanation.—The amount of emanation in equilibrium with 1 gramme of radium is called 1 curie; from 1 milligramme of radium, 1 millicurie. It is often necessary to determine the amount of emanation stored in a tube by itself. This is simply done by comparing the γ-ray activity of the emanation tube with that due to a standard quantity of radium. Knowing the period of decay of the emanation, the amount of emanation at any time can be deduced.

In comparisons of this kind it is necessary to apply a correction. The γ-rays do not arise from the emanation itself, but from its subsequent products, radium B and radium C. Five or six hours after introduction of the emanation into a tube, a state of changing equilibrium exists between the emanation and its products, and the γ-ray activity then decays at the same rate as the emanation. It is clear, however, from general considerations that the amount of radium B or radium C pre-

* Gray, "Proc. Roy. Soc," A. **85**, p. 131, 1911.

sent at any time must be always greater than corresponds to true equilibrium with the emanation. The theoretical correction can be easily determined. Let λ_1, λ_2, λ_3, λ_4 be the radioactive constants of the emanation, radium A, B and C respectively. If P_0 atoms of emanation are initially present, the amount S of radium C at a time, t, later is given* by

$$S=c_1e^{-\lambda_1 t}+c_2e^{-\lambda_2 t}+c_3e^{-\lambda_3 t}+c_4e^{-\lambda_4 t},$$

where $c_1=\dfrac{\lambda_1\lambda_2\lambda_3 P_0}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)}$, $c_2=\dfrac{\lambda_1\lambda_2\lambda_3 P_0}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}$

and so on.

Since the values of λ_2, λ_3, λ_4 for radium A, B and C are large compared with λ_1, after five or six hours the terms in $e^{-\lambda_2 t}$, $e^{-\lambda_3 t}$, $e^{-\lambda_4 t}$ become vanishingly small, and $S=c_1e^{-\lambda_1 t}$.

At the instant t the amount S_0 of radium C which would be in equilibrium with the amount P of the emanation, kept constant by supply from radium, is given by $S_0=\lambda_1 P_0 e^{-\lambda_1 t}$.

$$\text{Consequently}\quad S/S_0=\frac{\lambda_2\lambda_3\lambda_4}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)},$$
$$=1{\cdot}0089,$$

when the half value periods of emanation radium A, B and C are taken as 3·85 days, 3 minutes, 26·8 minutes, 19·5 minutes respectively.

This shows that the amount of radium C is always 0·89 per cent. *greater* than corresponds to permanent equilibrium with an equal quantity of emanation.

After the γ-radiation has passed through 2 cm. of lead, the γ-rays arise entirely from radium C. If the γ-rays from the emanation tube are compared with the γ-radiation from a radium standard through 2 cm. of lead, the amount of emanation in the tube is 0·89 per cent. *less* than that deduced from direct comparison with the γ-ray effect for the radium standard.

If the γ-rays pass through 3 mm. of lead, Moseley and Makower have shown that about 11·5 per cent. of the γ-radiation arises from radium B and 88·5 per cent. from radium C. The correction in this case is somewhat smaller, but can readily be shown to be about 0·85 per cent.

The amount of radium C present in a vessel after removal of the emanation is often determined by comparison of its γ-ray

* *See* Bateman, "Proc. Camb. Phil. Soc.," 15, p. 423, 1910.

effect after a definite interval with that due to a radium standard. Correction in this case has to be made for the γ-rays emitted from radium B, unless the γ-rays from the latter are removed by using a lead screen 2 cm. thick. Numbers showing the decay with time of the γ-ray activity of the active deposit of radium have been given for one important case by Moseley and Makower (*loc. cit.*).

Experiments with Balance Method.—The decay of the γ-ray activity of a large quantity of emanation was determined over several weeks by the balance method. The decay was found to be exponential with a half value period of 3·854 days. This is in excellent agreement with the results of Mme. Curie and Rutherford, who found values of 3·846 and 3·85 days respectively.

Some experiments were also made to determine experimentally the time required for the γ-ray activity of a tube filled with emanation to attain a maximum. The exact time was noted when emanation was introduced into a small glass tube and sealed off. The amount of γ-ray activity was accurately measured at intervals between 2 and 6 hours later.

The activity rose rapidly at first and was within 1·5 per cent. of the maximum after 3 hours. Four separate determinations were made and the maximum through a lead screen 3 mm. thick was found to occur at 255, 254, 256 and 257 minutes respectively after filling the tube. In this case, about 11·5 per cent. of the γ-radiation was supplied by radium B. Taking this factor into account, it was calculated that the maximum should occur at 255 minutes, an interval agreeing with the mean experimental value.

Using this balance method experiments were made to test whether the emission of γ-rays was affected by placing a tube containing emanation between the pole pieces of a large electromagnet. No detectable effect was observed.

ABSTRACT.

A balance method is described for accurately comparing quantities of radium by their γ-radiation, in which the ionisation due to the γ-rays is balanced against the constant ionisation due to uranium oxide. By observing the distance of the radium preparation from the ionisation chamber when a balance is obtained, the relative γ-ray activities of the two preparations can be determined with an accuracy of at least one part in 400. A method of calibration is given, and the corrections necessary to deduce the quantities of radium present are considered.

Calculations are given showing the correction required to determine a quantity of emanation by comparison of its γ-ray activity with that due to a radium standard.

The balance method was employed to determine the period of transformation of the radium emanation. The half value period was found to be 3·854 days. It was found experimentally that the γ-ray activity due to the radium emanation and its products reaches a maximum 255 minutes after the introduction of the emanation into a sealed tube. Calculations showed that under the experimental conditions the theoretical maximum should be reached after 255 minutes.

It was found that the γ-ray activity of a radium preparation was not appreciably altered by exposure in a strong magnetic field.

UNIVERSITY OF MANCHESTER, FEBRUARY, 1912.