Ionization Currents Produced by Radon, RaA and RaC′ in Cylindrical Ionization Chambers

ROBLEY D. EVANS, *Massachusetts Institute of Technology*

(Received May 21, 1935)

A general theoretical equation is derived for the ionization produced in cylindrical ionization chambers by Rn and its decay products. Where $dv/dt \equiv$volts per sec. discharge, $j \equiv$fractional saturation of the ion current, $C \equiv$electrostatic capacity in cm, $A \equiv$curies of Rn present at time $t=0$, $\lambda_1 \equiv$decay constant of Rn, $1/\mu \equiv$stopping power of the gas in the ionization chamber, $d \equiv$cm diameter and $h \equiv$cm height of the chamber, and $t \gtrsim 3$ hr.:

$$dv/dt = 5.3\times10^8(j/C)(3.57-5.00\mu/h - 3.47\mu/d+5.48\mu^2/hd)Ae^{-\lambda_1 t}$$

when d and h are greater than twice 6.60μ cm, the range of the RaC′ alpha-ray. Experimental confirmation employing standard radium solutions is described. Where either C or A is known, the other may be computed, therefore Rn ionization may be employed for absolute capacity measurements. General equations are also derived for alpha-ray ionization (1) above a plane surface due to a homogeneous radioactive substance below the plane, and (2) a radioactive substance uniformly distributed on a wire normal to the plane, and (3) for the ionization outside a dihedral right angle enclosing a homogeneous radioactive substance.

I. INTRODUCTION

THE convenience and structural simplicity of the cylindrical ionization chamber has led to its general adoption for the measurement of the ionization produced by the alpha-radiation from radioactive gases. The mathematical problem of predicting the net observable ionization produced by alpha-emitters in a cylindrical ionization chamber has long defied exact analysis. This is partly because of the geometrical complications, such as the presence of sharp corners, the lack of symmetry and the inhomogeneous fields at the ends, and partly because of inadequacies in the theory of the ionization by alpha-particles in a confined region bounded by absorbing walls. The desired analytical expression relates dv/dt, the rate of voltage discharge of the ionization-chamber-electrometer system, to its total electrostatic capacity, C; the height, h, and diameter, d, of the ionization chamber; the stopping power, $1/\mu$, of the gas in the chamber, and the amount, A, of radioactive material present. Thus:

$$dv/dt = (A/C)f(h, d, \mu) \qquad (1)$$

and the mathematical problem is the determination of $f(h, d, \mu)$.

An expression such as Eq. (1) is useful as (a) giving confirmation of and confidence in emanation measurements, (b) providing an absolute radium or radon calibration for laboratories possessing no precision radium standard but capable of making electrostatic capacity measurements, (c) providing an easy and accurate means of measuring electrostatic capacity in laboratories possessing a standard radium solution (d) providing a means of making accurate quantitative determinations of yields in atomic disintegration experiments if the products are alpha-active.

Equations analogous to Eq. (1), for the case of Rn and its alpha-emitting decay products RaA and RaC′ in a parallel plate condenser have been given by Flamm and Mache,[1] Siegl[2] and others, while the case of the spherical ionization chamber has been treated by Lind,[3] Mund[4] and Glockler and Heisig.[5]

Empirical expressions for Rn, and for Rn, RaA and RaC′ in equilibrium in cylindrical ionization chambers were given by Duane[6] and Duane and Laborde,[7] and these have been widely used.[8] Evans[9] combined some of Duane and Laborde's data with a preliminary theory to

[1] Flamm and Mache, Akad. Wiss., Wien, Ber. [II *a*], **121**, 227 (1912).
[2] Siegl, Akad. Wiss., Wien, Ber. [II *a*] **134**, 11 (1925).
[3] Lind, J. Phys. Chem. **16**, 564 (1912).
[4] Mund, J. Phys. Chem. **30**, 890 (1926).
[5] Glockler and Heisig, J. Phys. Chem. **35**, 2478 (1931).
[6] Duane, Comptes rendus **1**, 581 (1905); J. de phys. et rad. **4**, 605 (1905).
[7] Duane and Laborde, Comptes rendus **1**, 1421 (1910).
[8] Schmidt and Nick, Physik. Zeits. **14**, 199 (1912); Mache and Meyer, Physik. Zeits. **13**, 320 (1912); Berndt, Ann. d. Physik **38**, 958 (1912); Ramsey, Am. J. Sci. **40**, 309 (1915); Gockel, *Die Radioaktivität von Boden und Quellen*, Vieweg (1914), p. 102; Lester, Am. J. Sci. **44**, 225 (1917); Zlatarovic, Akad. Wiss., Wien, Ber. [II *a*] **129**, 59 (1920), etc.
[9] Evans, Thesis, Calif. Institute of Tech. (1932), p. 62.

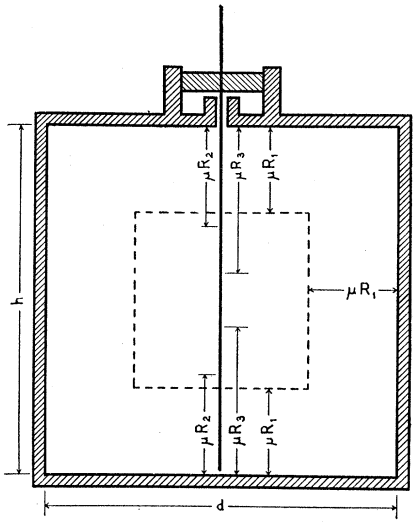

FIG. 1. Schematic cylindrical ionization chamber.

derive a semi-empirical formula for cylindrical ionization chambers in which the ionization from the solids, RaA and RaC′, and the gaseous Rn were treated independently. In the treatment given below, $f(h, d, \mu)$ is evaluated entirely analytically, and the results are confirmed by direct experiments.

Fig. 1 illustrates the typical cylindrical ionization chamber considered in the following analysis. The center electrode is a wire of sufficiently small diameter to occupy a volume which is negligible in comparison with the rest of the chamber. The center wire is charged negatively, hence collects[10] the positively charged[11] atoms of RaA as they are formed by the decay of Rn in the chamber.

R_1, R_2, R_3 are the ranges of the Rn, RaA and RaC′ alpha-particles in air at 0°C and 760 mm Hg, and μR_1, μR_2, μR_3 are their ranges in the gas filling the ionization chamber. An alpha-particle from Rn emitted within a distance of μR_1 from any wall of the chamber and directed toward that wall will lose some of its energy by absorption in the wall. To determine what ionization is produced inside the chamber by Rn alpha-particles we must evaluate the ionization lost in the wall, and deduct this from the total maximum ionization which would be produced if all the energy were dissipated in ionizing the gas in the chamber. Similar corrections must be derived for those RaA and RaC′ alpha-particles which are emitted from the lower or upper portion of the center wire and strike an end wall. To avoid overlapping, the theory is here derived only for chambers in which the diameter and height exceed twice the range of the RaC′ alpha-particle.

For generality, we derive the dependence of the ionization current on time elapsed (a) after introducing Rn into a cylindrical ionization chamber, and (b) after withdrawing Rn from a chamber in which it had previously come to transient equilibrium with its decay products.

II. Accumulation of RaA and RaC′

When Rn is introduced into an ionization chamber its disintegration gives rise to the successive solid decay products RaA, RaB, RaC, RaC′, RaD, etc. For the present work the 0.04 percent of RaC traversing the RaC″ branch is entirely negligible. Of these decay products only RaA and RaC′ emit alpha-particles, and the half-life of RaD is so long (25 yrs.) that its decay products are negligible. We have therefore to consider the number of alpha-particles per sec., N_1, N_2, N_3, from Rn, RaA, RaC′ respectively, as a function of time, where, at $t=0$, $N_1=Z$ and $N_2=N_3=0$. The general expressions for the number of atoms of any of the decay products as a function of time constitute the familiar Case I of Rutherford's treatises, and the general equations,[12] given by Bateman,[13] need only to be multiplied by the decay constants of the products involved to give N_1, N_2, N_3. In this way, substituting for the decay constants: $\lambda_{Rn}=2.097\times10^{-6}$ sec.$^{-1}$, $\lambda_{RaA}=3.79\times10^{-3}$ sec.$^{-1}$, $\lambda_{RaB}=4.31\times10^{-4}$ sec.$^{-1}$, $\lambda_{RaC}=5.86\times10^{-4}$ sec.$^{-1}$, $\lambda_{RaC'}=ca.10^{6}$ sec.$^{-1}$, as given by Rutherford,[12] we obtain:

$$N_1=Ze^{-\lambda_{Rn}t} \tag{2}$$

$$N_2=1.00054Z(e^{-\lambda_{Rn}t}-e^{-\lambda_{RaA}t}) \tag{3}$$

$$N_3=1.0089Z(e^{-\lambda_{Rn}t}-0.0232e^{-\lambda_{RaA}t}-4.26e^{-\lambda_{RaB}t}+3.28e^{-\lambda_{RaC}t}). \tag{4}$$

[10] Dadourian, Am. J. Sci. **19**, 16 (1905); Kovarik and McKeehan, Bull. Nat. Research Council **10**, 140 (1925).
[11] McGee, Phil. Mag. **13**, 1 (1932).
[12] Rutherford, *Radioactivity* (1905), p. 332; Rutherford, Chadwick and Ellis, *Radiations from Radioactive Substances* (1930), p. 11.
[13] Bateman, Proc. Camb. Phil. Soc. **15**, 423 (1910).

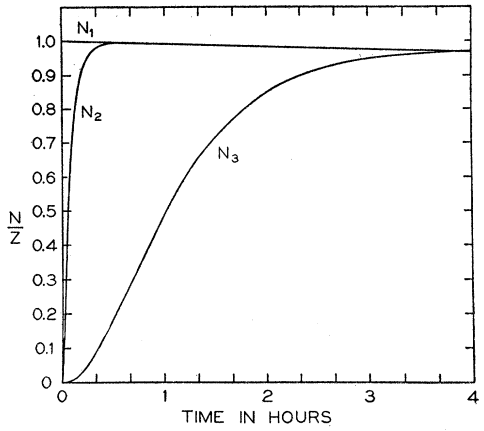

FIG. 2. Rate of emission of alpha-particles from Rn, (N_1), and accumulating decay products, RaA, (N_2), and RaC', (N_3), in terms of emission from Rn at $t=0$. See Eqs. (2), (3), (4).

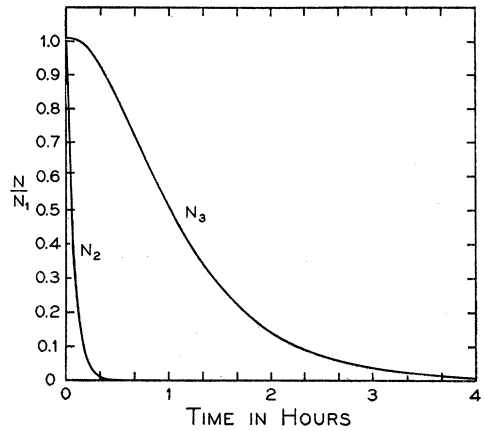

FIG. 3. Rate of emission of alpha-particles from RaA, (N_2), and RaC', (N_3) after removing a Rn source giving N_1 alpha-particles per sec. at $t=0$.

The numerical values for Eqs. (2), (3), (4) are given in Fig. 2.

III. Decay of RaA and RaC'

If, after transient equilibrium has been established between Rn and its decay products through RaC', the Rn is suddenly removed, then the number of alpha-particles per sec. N_2 and N_3, from RaA and RaC', respectively, may be expressed in terms of the number of Rn alpha-particles per sec., N_1, at the moment previous to withdrawing the Rn from the ionization chamber. N_2 and N_3 may be computed from Bateman's[14] equations or from Meyer and Schweidler's[15] table, by multiplying the amount of each element, as given by these equations or tables, by the decay constant of the element. It can be seen that the mean life of RaC' is so short that the rate of emission of RaC' alpha-particles is equal to that of RaC, which facilitates computation. Fig. 3 shows N_2 and N_3, the rate of emission of RaA and RaC' alpha-particles after the removal of a Rn source giving N_1 alpha-particles per sec. at $t=0$, transient equilibrium having been present.

[14] A serious misprint in the original general equations for decay (reference 13) has avoided correction for twenty years. In Rutherford, Chadwick and Ellis (reference 12) p. 14, the coefficient d of Eq. (13) should be multiplied by $1/\lambda_4$.

[15] Meyer and Schweidler, *Radioaktivität* (1927), p. 437.

IV. Ionization by Rn in Cylindrical Ionization Chambers

When a small amount of Rn is added to the gas in a cylindrical brass ionization chamber all the Rn remains uniformly distributed throughout the entire volume of the chamber. We have verified this by withdrawing samples of gas from a chamber and examining the resulting change in ionization.

The ionization losses due to absorption of alpha-particles in the walls can be computed by regarding the wall as the outer boundary of a homogeneous radioactive material and computing the ionization outside this boundary.[16] Exact integrals for the alpha-particle ionization, q, per cm^2 above the plane surface of a radioactive solid emitting N alpha-particles per sec. per cm^3 have been given by Evans.[17] If R is the range of the alpha-particle in air at 0°C and 760 mm Hg, $R\mu$ is its range in the radioactive source, k the total number of ion pairs per alpha-particle, then

$$q=\epsilon kNR\mu \qquad (5)$$

where ϵ is a dimensionless coefficient, increasing slightly with R, and having values between 0.114 and 0.150 for all known alpha-particles from Sm ($R=1.06$ cm) to ThC' ($R=8.17$ cm).

Considering, for the moment, all the walls of

[16] R. W. Raitt, personal communication, June, 1933.

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

the cylindrical chamber of volume, V cm^3, as plane surfaces of total area, S cm^2, and writing subscripts 1 to indicate numerical values for Rn alpha-particles, the total ionization absorbed by these walls is:

$$i_1 = \epsilon_1 k_1 N_1 R_1 \mu S/V \qquad (6)$$

where N_1 is the total number of Rn alpha-particles per sec. in the entire chamber, hence N_1/V represents the Rn alphas per cm^3.

The actual curvature of the walls leads to two small and oppositely directed correction terms which, for practical measurements, cancel each other. On the one hand the curvature decreases the effective fractional volume of radioactive source, which is within $R\mu$ cm of the wall, from $R\mu S/V = 2R\mu(1/h+2/d)$ to $2R\mu(1/h+2/d - 2R\mu/d^2)$. This correction has its maximum value for small chambers, where it corresponds to about 5 percent of the Rn ionization, or 2.5 percent of the ionization from Rn and its decay products. On the other hand the wall curvature increases the wall area which can be hit from any point radially distant less than $R\mu$ cm from the wall. This fractional increase is about $1-d\sin^{-1}(2(b^2-y^2)^{\frac{1}{2}}/d)/2(b^2-y^2)^{\frac{1}{2}}$ where d is the diameter of the chamber, b the length of track inside the chamber and y the perpendicular distance from the origin of b to the chord which defines the hypothetical plane replaced by the actual curved wall. Without practical error, we may say that these two effects of wall curvature exactly cancel each other, leaving Eq. (6) as the Rn ionization absorbed by the walls.

The corners of the chamber give rise to a small but appreciable correction. This can be evaluated by computing the ionization produced outside a dihedral right angle by a radioactive source within, and bounded by, the angle. Since this computation has general applications to all finite, bounded radioactive sources, where it is the "edge correction," its derivation will be given. In Fig. 4, an element of volume, in the radioactive source bounded by the dihedral right angle AOB', emits alpha-rays uniformly in all directions. Those which emerge above the plane AA' cause ionization above this plane which is exactly described by Eq. (5), where q is taken as the total ionization above AA' per cm of length (into the plane of the paper) of the dihedral edge, O, and where the effective cross sectional area of the source (in the plane of Fig. 4) for emission of alpha-rays capable of emerging from AA' is $(R\mu)^2$. Consideration of this ionization is indicated in Fig. 4 by circles on the alpha-ray tracks. Similarly those which emerge to the right of the plane BB' are described by Eq. (5) and are indicated in Fig. 4 by dots on the alpha-ray tracks. The ionization from these two plane edges is already included in Eq. (6), because S is taken as the total inside surface of the ionization chamber.

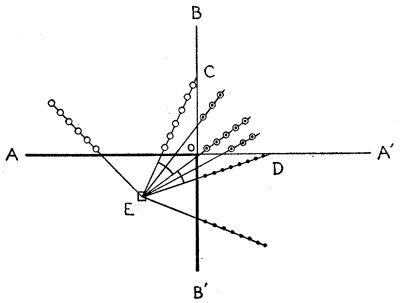

FIG. 4. Schematic cross section describing corrections for ionization outside a dihedral right angle AOB' bounding a homogeneous radioactive source.

Inspection of Fig. 4 shows that alpha-particles emergent above AA' and to the right of BB' have been counted twice, as indicated by the dotted circles. We must therefore compute the ionization represented by these track elements and deduct it from the ionization described by Eq. (6).

As shown in the next section, the general expression for the corner correction is $i' = \psi N k R^2 \mu^2$ ion pairs per sec. per cm of edge where ψ is a dimensionless coefficient. As in the case of the side walls, the curvature of the corner exerts two oppositely directed influences, one due to reduction of effective volume, the other to increased effective surface exposed to each volume element, which cancel each other. Each of these two corrections represents, in this case, a maximum of only about 0.6 percent of the total Rn ionization in small chambers, where curvature effects are most pronounced. The total effective length of the corners at the top and bottom of the chamber is $2\pi d$, therefore the Rn ionization absorbed by the walls is:

$$i_1 - i_1' = \epsilon_1 k_1 N_1 R_1 \mu (S/V) - \psi_1 k_1 N_1 R_1^2 \mu^2 (2\pi d/V) \qquad (7)$$

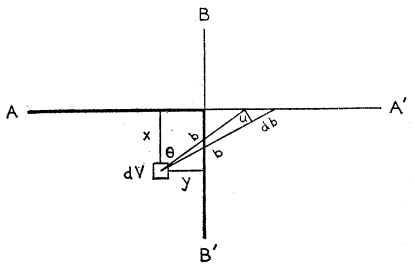

FIG. 5. Geometry for Eqs. (10) to (15) giving ionization outside a dihedral right angle.

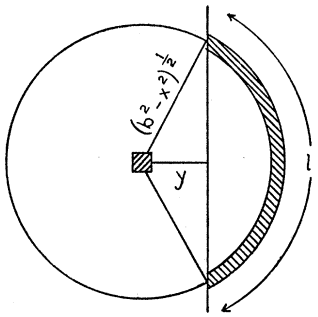

FIG. 6. Geometry for Eqs. (10) to (15), in the AA' plane of Fig. 5.

and the total net ionization by Rn is $I_1=N_1k_1-(i_1-i_1')$ ion pairs per sec. Substituting $S=\pi d^2/2+\pi hd$, and $V=\pi hd^2/4$, we have for the Rn ionization:

$$I_1=N_1k_1\{1-2\epsilon_1R_1\mu(1/h+2/d)+8\psi_1R_1^2\mu^2/hd\}. \quad (8)$$

As shown in the next section ψ for Rn $(=\psi_1)=0.0208$, and from Table I of reference 17, $\epsilon_1=0.143$, $R_1=3.91$ cm, $k_1=1.55\times10^5$ ion pairs per Rn alpha-particle. Hence, in general, for Rn ionization in a cylindrical ionization chamber:

$$I_1=1.55\times10^5N_1\{1-1.12\mu(1/h+2/d)+3.54\mu^2/hd\}\text{ ion pairs per sec.} \quad (9)$$

V. Alpha-Particle Ionization Outside a Dihedral Right Angle

As shown in Section IV, the ionization from the dotted and circled alpha-ray track ends in Fig. 4 constitutes the corner correction required for all cases involving a homogeneous radioactive source confined within the dihedral right angle AOB'. This ionization may be computed by adding the contribution from alpha-rays emerging from the plane of AA' to the right of BB' (those within the angle OED) to those alpha-rays emerging from the plane of BB' above AA' (those within the angle OEC). From symmetry these two are equal, hence, for analysis, rays emerging within the angle OED will be considered.

In Fig. 5, take the x and y coordinates of the volume element $dV=dxdydz$, as shown, with dz normally into the plane of the page. We first find the total number, dn, of alpha-rays emerging in the angle OED, of Fig. 4, which have ranges below AA' of between b and $b+db$ cm. Since the stopping power of the source is $1/\mu$, and since the stopping power may be taken as constant over the entire region $AA'BB'$ without loss of generality, the ionization produced above AA' by each of these rays is the same as that produced along the path of an alpha-ray in air at 0°C and 760 mm Hg between a point b/μ cm from the origin of the ray and the end of the ray. If $\varphi(r)$ is the specific ionization in ion pairs per cm of path in air at 0°C and 760 mm Hg at a distance r from the origin of the "standard" alpha-ray, then the ionization above AA', per cm of depth in z, equals $dn\int_{b/\mu}^{R}\varphi(r)dr$, and the total ionization, i', for all values of b from 0 to $R\mu$, and for all rays emergent in both the angles OED and OEC, i. e., for all dot-circled rays in Fig. 4, is:

$$i'=2\int_{b=0}^{b=R\mu}dn\int_{b/\mu}^{R}\varphi(r)dr. \quad (10)$$

The evaluation of dn follows the geometry of Figs. 5 and 6. The number of alpha-rays per sec. emitted in all directions from the volume element dV is $Ndxdydz$, where N is the number of alpha-rays per sec. per cm^3. The fraction of these rays which emerge from the plane AA' to the right of BB' is $ul/4\pi b^2$, where l is the length of the arc of emergence of rays which have ranges below AA' lying between b and $b+db$ cm. From geometry, $u=db\cot\theta=\{x/(b^2-x^2)^{\frac{1}{2}}\}db$, and $l=2(b^2-x^2)^{\frac{1}{2}}\cos^{-1}\{y/(b^2-x^2)^{\frac{1}{2}}\}$. Hence the number of rays originating in dV, and emerging above AA' and to the right of BB' (i. e., within the angle OED) with ranges between b and $b+db$ below AA' is $(ul/4\pi b^2)NdV=(Nx/2\pi b^2)\cos^{-1}\{y/(b^2-x^2)^{\frac{1}{2}}\}dbdxdydz$, and the total number of such rays, per cm depth in z, is:

$$dn=\frac{Ndb}{2\pi}\int_0^1 dz\int_b^0 dy\int_{(b^2-y^2)^{\frac{1}{2}}}^0 \frac{x}{b^2}\cos^{-1}\frac{y}{(b^2-x^2)^{\frac{1}{2}}}dx \quad (11)$$

$$=(N/6\pi)bdb. \quad (12)$$

Substituting Eq. (12) in Eq. (10) we have for the ionization dot-circled in Fig. 4:

$$i'=(N/3\pi)\int_0^{R\mu} bdb\int_{b/\mu}^R \varphi(r)dr. \quad (13)$$

The point b along the path of the actual alpha-ray, in a medium of uniform (i.e., independent of r) stopping power $1/\mu$, corresponds in specific ionization to the point r along the path of a standard alpha-ray in air at 0°C and 760 mm Hg when $r=b/\mu$. Making this substitution in Eq. (13), we have:

$$i'=(N\mu^2/3\pi)\int_0^R rdr\int_r^R \varphi(r)dr, \quad (14)$$

which is valid for media of any μ.

Lemma. By partial integration of the left side of Eq. (15) and integration by parts of the right side, it may be shown that if n and φ are any functions of r, $n=n(r)$, $\varphi=\varphi(r)$, then, provided that $n(a)=0$,

$$\int_a^R dn\int_r^R \varphi dr=\int_a^R n\varphi dr. \quad (15)$$

Eq. (14) may therefore be transformed with the aid of the lemma of Eq. (15), resulting in:

$$i'=(N\mu^2/6\pi)\int_0^R r^2\varphi(r)dr, \quad (16)$$

which may be written:

$$i'=\psi NkR^2\mu^2, \quad (17)$$

where ψ is a dimensionless coefficient and, like ϵ of Eq. (5), depends slightly on R. For $R=3.91$ cm (Rn), graphical integration employing $\varphi(r)$ as found by Henderson[17, 18] given $\psi_1=0.0208$. If the approximate analytical expression[17] $(\frac{2}{3})kR^{-\frac{2}{3}}(R-r)^{-\frac{1}{2}}$ is substituted for $\varphi(r)$, the integration may be carried out analytically, and leads to the approximate value, $\psi_\infty=3/40\pi=0.0238$, which is the limiting value for very long alpha-rays.

[18] G. H. Henderson, Phil. Mag. **42**, 538 (1921).

Eq. (15), with suitable values for ψ, describes completely the edge or corner corrections applicable to all finite radioactive sources of the type considered here. In practical cases it must be combined with Eq. (5), as was done in deriving Eq. (7).

VI. Ionization Due to RaA and RaC′

As pointed out in Section I the negatively charged center wire of the cylindrical ionization chamber of Fig. 1 is assumed to collect all the RaA atoms resulting from the disintegration of Rn. Since the mean life of RaA is very long compared with the time required for collecting this ion from its place of origin in the gas, all the RaA alpha-rays, as well as those from RaC′, are regarded as emitted from the surface of the center wire. If a small fraction of the RaA should be collected on the walls of the chamber, the difference in the energy absorbed by the end wall would be negligible, as the equations have the same form, differing only by a term representing the wall curvature. Because the emission is random in direction, half of the RaA and RaC′ alpha-rays will be absorbed in the center wire. Of the remaining rays, those emitted within a distance $R_2\mu$ for RaA, and $R_3\mu$ for RaC′, of an end wall, and directed toward that wall, will lose some of their energy in the wall.

The method of analytical evaluation of this energy is identical for the RaA and RaC′ rays. Assuming that the radioactive source (say RaA) is uniformly distributed along the surface of a wire normal to, and just reaching the plane DD' of Fig. 7, we need to evaluate the ionization appearing above the plane DD', which, in this case, is the energy absorbed by the wall. If the wire is h cm long and emits N_2 alpha-rays per sec. in all directions, hence $N_2/2$ which can be effective in producing ionization below the plane

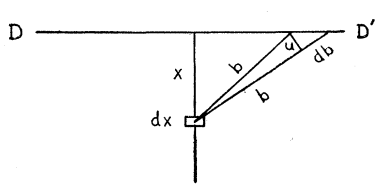

Fig. 7. Geometry for Eqs. (18) to (24) describing ionization above DD' due to alpha-particles emitted from a wire normal to DD'.

DD', then in an element of length, dx, there will be $(N_2/2h)dx$ alpha-rays effectively emitted. The fraction of those emitted from a depth, x, below DD' which have the range element between b and $b+db$ below DD' is $2\pi(b^2-x^2)^{\frac{1}{2}}u/4\pi b^2 = (x/2b^2)db$, and the number of such rays is $(N_2/2h)(x/2b^2)dbdx$ from the depth x, or $(N_2/8h)db$ from all depths between $x=0$ and $x=b$. Every ray in such a group produces ionization above DD' equal to $\int_{b/\mu}^{R}\varphi(r)dr$, where r, as in Section V is measured on the path of a standard alpha-ray in air at 0°C and 760 mm, and $1/\mu$ is the stopping power of the material in the region below DD'. The total ionization above DD', due to rays having all possible values of b from $b=0$ to $b=R\mu$ is

$$i=(N_2/8h)\int_0^{R\mu}db\int_{b/\mu}^{R}\varphi(r)dr \qquad (18)$$

and substituting $r=b/\mu$ as for Eq. (14), and applying the lemma of Eq. (15) we find

$$i=(N_2\mu/8h)\int_0^{R}dr\int_r^{R}\varphi(r)dr$$

$$=(N_2\mu/8h)\int_0^{R}r\varphi(r)dr \qquad (19)$$

$$\equiv(N_2\mu/2h)\epsilon kR, \qquad (20)$$

where ϵ is the dimensionless coefficient defined by Eqs. (19) and (20), and appearing in Eq. (5), and is obtained by graphical integration[17] of Eq. (19). The RaA ionization lost by absorption in the two end walls is, therefore, $2i_2$, where the subscript 2 refers to RaA, and the ionization produced in the gas of the cylindrical ionization chamber by RaA is

$$I_2=(N_2k_2/2)-(N_2\mu/h)\epsilon_2k_2R_2$$
$$=\tfrac{1}{2}N_2k_2(1-2\epsilon_2\mu R_2/h) \qquad (21)$$

and, similarly, the ionization in the chamber, due to RaC′ is

$$I_3=\tfrac{1}{2}N_3k_3(1-2\epsilon_3\mu R_3/h), \qquad (22)$$

where the subscripts 3 refer to RaC′ values of N, R, k and ϵ. From Table I of reference 17, $\epsilon_2=0.144$, $R_2=4.48$ cm, $k_2=1.70\times10^5$ ion pairs per RaA alpha-ray; and $\epsilon_3=0.148$, $R_3=6.60$ cm, and $k_3=2.20\times10^5$ ion pairs per RaC′ alpha-ray. Substituting in Eqs. (21) and (22) we obtain:

$$I_2=0.85\times10^5N_2(1-1.29\mu/h) \qquad (23)$$

$$I_3=1.10\times10^5N_3(1-1.95\mu/h)$$

ion pairs per sec. (24)

VII. Total Ionization Due to Rn+RaA +RaC′

The ionization currents actually observed in the cylindrical ionization chamber are due to the combined effects of the alpha-rays from Rn and its alpha-emitting decay products RaA and RaC′. Other sources of ionization such as that due to beta-rays from RaB and RaC, and that due to electrons liberated from the walls by the alpha-rays which strike the walls, are negligible in comparison. The total alpha-ray ionization, I, is the sum of I_1, I_2 and I_3 of Eqs. (9), (23) and (24).

A small additive correction must be made for the ionization produced by the recoil atoms. For elements near RaA, momentum considerations show that the recoil atom has 2.0 percent as much energy as the associated alpha-ray. Most of this recoil energy produces ionization, but a small fraction of it may be dissipated in elastic molecular collisions without ionization. Bearing in mind that the density of ionization due to a recoil atom exceeds that due to an alpha-ray, it is probable that there is more recombination of ions, and a correspondingly lower saturation, in the ionization produced by recoil atoms. We may safely say that 1.5 ± 0.5 percent should be added to the alpha-ray ionization to account for recoil energy. Because the recoil tracks are very short there will be no significant wall correction in their case. Every Rn alpha-ray gives rise to an effective recoil atom, and those RaA and RaC′ alpha-rays which are directed into the center electrode each produce a recoil atom which can ionize the gas. The correction for recoil therefore increases the first term of Eqs. (9), (23) and (24) from 1 to 1.015 ± 0.005, as shown in Eq. (26).

With ordinary collecting potentials of several hundred volts across the chamber, some recombination of ions will occur in the dense columnar

ionization due to the alpha-rays. The relative saturation, j, discussed in Section VIII, is the fraction of the total ionization, I, which is actually observed. If C is the electrostatic capacity of the ionization-chamber-electrometer system in cm, e the electronic charge, 4.77×10^{-10} e. s. u., and dv/dt the discharge rate in volts per sec. then:

$$dv/dt=300jeI/C \tag{25}$$

and

$$\begin{aligned}I&\equiv I_1'+I_2'+I_3'\\&=1.55\times10^5N_1\{1.015-1.12\mu(1/h+2/d)\\&+3.54\mu^2/hd\}+0.85\times10^5N_2\{1.015-1.29\mu/h\}\\&+1.10\times10^5N_3\{1.015-1.95\mu/h\}.\end{aligned}\tag{26}$$

During the accumulation or decay periods discussed in Sections II and III, Rn is not in equilibrium with RaA nor RaC', and N_1, N_2 and N_3 are the functions of time described by Eqs. (2), (3), (4) and Figs. 2 and 3. For the brass ionization chambers used in the experimental phases of this investigation $h=d=6$ in. $=15.24$ cm. The chambers were filled with nitrogen at 22°C and 760 mm Hg, hence, from the Bragg-Kleeman rule, $\mu=1.09$. Substituting these values in Eq. (26) we have:

$$I=(1.24N_1+0.79N_2+0.97N_3)\times10^5. \tag{27}$$

Fig. 8 shows the dependence of I on time for both accumulation and decay. The experimental points, plotted on Fig. 8 for comparison with the theory, show statistical fluctuations about the mean value because of the small quantity of Rn employed in these tests (19.66×10^{-12} curie). Reliable ionization values for small values of t are difficult to observe because of the possibility of the presence of heavy ions in chamber, due to filling, which require several minutes for collection. The shape of these theoretical curves is not strongly dependent on h and d, since these must exceed $2R_3\mu$ for the cases treated by the theory. The agreement in Fig. 8 between theory and experiment is entirely satisfactory, but a more critical test is offered by observing the total equilibrium ionization due to a known amount of Rn, as is done in Section IX.

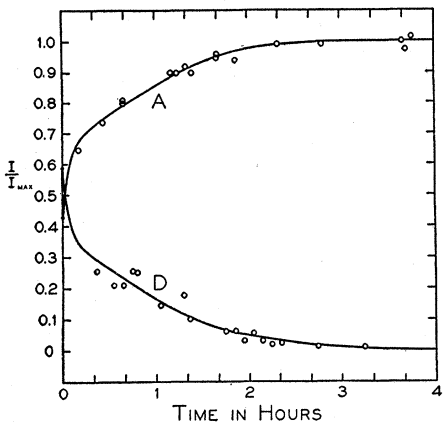

FIG. 8. Ionization in a brass chamber, $h=d=15.24$ cm, $\mu=1.09$, for (A) Rn plus accumulating RaA and RaC', and (D) decay of RaA and RaC' after withdrawing Rn.

VIII. Relative Saturation of Alpha-Ray Ionization Current in Cylindrical Ionization Chambers

Brössler[19] measured the degree of saturation achieved by various collecting potentials in cylindrical ionization chambers containing large amounts of Rn and its decay products, finding the saturation also dependent on the amount of Rn. If J is the saturated ion current in e. s. u. per sec., i the observed ion current in e. s. u. per sec., E the potential on the chamber in volts, and k, K, constants, Brössler found, for a chamber in which $h=30$ cm, $d=25$ cm, center electrode 0.6 cm diameter and RaA and RaC' collected on the wall:

$$i=J(1-e^{-Ek}) \tag{28}$$

$$J^2k^5=K \tag{29}$$

where $K=4.44\times10^{-11}$ e. s. u.^{2} sec.$^{-2}$ volt^{-5} for $J<100$ e. s. u. per sec., and $K=4.57\times10^{-11}$ for $J>100$. At $J=100$, Brössler gave $k=0.0013512$. The measurements were presumably made with air in the ionization chamber.

For small quantities of Rn, such as are met in the Rn analysis of ordinary materials, Brössler's data predict $j=i/J=1.000$, which is not in agreement with observations. Brössler's data should, therefore, not be extrapolated beyond the range of his measurements, i.e. $10\leq J\leq210$ e. s. u. per sec.

[19] Brössler, Akad. Wiss., Wien, Ber. [II a] **129**, 47 (1920).

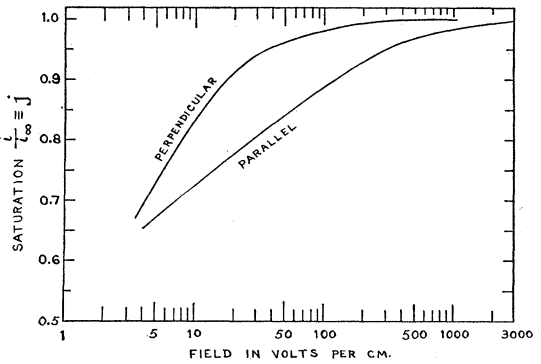

FIG. 9. Saturation of alpha-ray ionization in air at 760 mm Hg for rays (I) parallel to the collecting field, (II) perpendicular to the collecting field. (Moulin.[20])

Moulin[20] measured the relative saturation of alpha-particle ionization when the path of the ray is (a) parallel to the collecting field, (b) perpendicular to the collecting field. Measurements were made in air, CO_2 and H_2. Moulin's values for air are plotted in Fig. 9. The minimum value of the average collecting field in a cylindrical ionization chamber may be computed by neglecting end effects. Where E is the potential between the wall of diameter d and the center electrode of diameter a, and r is the radial distance from the axis of the chamber to a volume element $2\pi r dr$ in area and of unit length, the minimum volumetric average field, $(dE/dr)_{av}$, in the chamber is:

$$\left(\frac{dE}{dr}\right)_{av} = \left\{\int_{a/2}^{d/2} \frac{E}{\log_\epsilon d/a} \cdot \frac{1}{r} 2\pi r dr\right\} \Big/ \frac{\pi}{4}(d^2-a^2)$$

$$= 4E/(d+a)(\log_\epsilon d/a). \qquad (30)$$

From Eq. (30) and Fig. 9 the minimum value of the saturation, j, may be computed for any cylindrical chamber by taking a mean value from curves I and II of Fig. 9 as describing a uniform mixture of all possible angles between the alpha-rays and the collecting field gradient. For the present chambers, at $E=300$ volts, $(dE/dr)_{av} = 18.7$ volts/cm, hence $j>0.84$.

Since j also varies with the chemical nature of the gas in the chamber (for example, j is low[21] for CO_2, high for very pure N_2), it is preferable to make a direct measurement of j in any ionization-chamber-electrometer system which is being calibrated for precision measurements. This is best done by observing the ionization, I, at various values of E, and extrapolating a graph of I/E *vs.* I to $I/E=0$, $(E=\infty)$. For approximate use, j is usually between 0.87 and 0.93 in cylindrical chambers filled with CO_2, air or nitrogen at 760 mm Hg, where d and h are between 30 and 15 cm, and E is 300 volts. The saturation, j, at $E=300$ volts was found to be 90 ± 1 percent in the present chambers.

IX. Capacity Calibrations for Ionization Chambers

The ionization chambers used in the present experiments were connected in the double-chamber differential circuit regularly employed[22] for the measurement of small quantities of Rn. Direct capacity calibrations were made by adding small cylindrical condensers of identical cross section but of various lengths (and hence of known differences in capacity) to the central system and observing the fractional change in capacity. Quantitative observations of the ratios of the several capacities so produced were made by several independent methods.[22b] (A) By observing the potentials induced on the central system (center electrodes of ionization chambers and string electrometer fiber) when the potential on one ionization chamber is suddenly changed by a known voltage. (B) By observing the rates of discharge of the central system with one ionization chamber evacuated, the other radiated with gamma-rays. (C) By observing the rates of discharge of the central system when Rn plus decay products is used as the ionizing agent in one of the chambers.

The method (A) is a development of the inductive method, usually credited to Harms,[23] but used over a decade earlier by Lebedew.[24] It has been used by many subsequent observers[25]

[20] Moulin, Ann. de Chem. et Phys. **21**, 550 (1910); **22**, 26 (1911).

[21] Hess and Hornyak, Akad. Wiss., Wien, Ber. [II *a*] **129**, 661 (1920).

[22] Evans, (a) Phys. Rev. **39**, 1014 (1932); Rev. Sci. Inst. **4**, 223 (1933); (b) **6**, 99 (1935).

[23] F. Harms, Physik. Zeits. **5**, 47 (1904).

[24] Lebedew, Wied. Ann. **44**, 289 (1891). Kohlrausch, *Physical Measurements* (1894), p. 380.

[25] Siegl, Akad. Wiss., Wien, Ber. [II *a*] **134**, 11 (1925); Halledauer, Akad. Wiss., Wien, Ber. [II *a*] **134**, 39 (1925); Steinke, Zeits. f. Physik **48**, 647 (1928); Schindler, Zeits. f. Physik, **72**, 625 (1931); J. Clay, Zeits. f. Physik **78**, 250 (1932); H. Lampe, Zeits. f. Physik **79**, 254 (1932).

and has been shown to agree with electrodynamic methods to 1 part in 1000 by Lampe.[25] Using the inductive principle, a null method can be employed by which capacities may be compared with a standard capacity to 1 part in 10^6.

The numerical results from the three methods (A)(B)(C) are in agreement, within the respective probable errors of measurement, and indicate a total capacity of 16.4 cm for the double ionization-chamber string-electrometer apparatus, when the electrometer is at zero sensitivity. As was shown independently by Raitt[26] and by Sienkiewicz[27] and Benndorf[28] the effective capacity of a string electrometer rises with sensitivity. In the present apparatus, when v volts are applied to the electrometer plates, and s is the resulting sensitivity of the electrometer in divisions per volt, the effective capacity, C, is given by:

$$C=16.4(1+1.0\times10^{-5}\,vs). \qquad (31)$$

An analogous equation holds for all similar electrometer systems.

There is a fourth method for measuring the capacity, C, of the electrometer system. Assuming the validity of the present theoretical treatment of the ionization in cylindrical chambers by Rn and its decay products, we measure (a) the equilibrium ionization, dv/dt, due to a known amount of Rn, and (b) the relative saturation, j, of this ionization current. Then employing Eqs. (25) and (27), or Eq. (33) we can compute C. The Rn for these tests was released from a portion of a standard radium solution, most generously furnished by Professor S. C. Lind, which was prepared[29] from pure $RaCl_2$ which had been compared, by the gamma-ray method, with the international radium standard by the U. S. Bureau of Standards. The Rn was released from a solution containing 19.66×10^{-12} g Ra by boiling in a glass apparatus[30] developed for the routine analyses of liquid specimens having very low Ra content. The mean of a large number of such Rn calibrations, made over a period of two years in connection with routine analyses of feebly radioactive materials, leads to a value for C which is in complete agreement with the direct measurements by the three methods of the preceding paragraph. Because of the normal statistical fluctuations resulting from the use of a feeble Ra solution,[31] the probable error is about ±1 percent.

X. General Equation for Radon Calibration of a Cylindrical Ionization Chamber

Eqs. (25) and (26) may now be combined and simplified to describe the ionization current produced by Rn and RaA+RaC′ in transient equilibrium. If A curies of Rn are introduced into the cylindrical ionization chamber at time $t=0$, and the rate of discharge, dv/dt volts per sec., is observed at time $t\geq3$ hr., when transient equilibrium is practically established, then $N_1=3.7\times10^{10}Ae^{-\lambda_1 t}$, where λ_1 is the decay constant[32] of Rn, 7.551×10^{-3} hr.$^{-1}$, and $N_2=1.00054N_1$, $N_3=1.0089N_1$, and

$$I=10^5N_1(3.57-5.00\mu/h-3.47\mu/d+5.48\mu^2/hd) \qquad (32)$$

$$dv/dt=300jeI/C=5.3\times10^8(j/C)(3.57-5.00\mu/h-3.47\mu/d+5.48\mu^2/hd)Ae^{-\lambda_1 t}. \qquad (33)$$

Eq. (33) is the general equation for the Rn calibration of any cylindrical ionization chamber having $d\geq2\mu R_3$ and $h\geq2\mu R_3$, and is the solution of Eq. (1).

Acknowledgments

The assistance of Russell W. Raitt and Ben Holzman, of the California Institute of Technology, and of Arthur Kip, Fred Ludecke and Raymond Mugele of the University of California in various experimental phases of this investigation is deeply appreciated. Grateful acknowledgment for hospitality and support is also made to the Physics Department of the University of California, where the present theoretical treatment was begun during the author's tenure as National Research Council Fellow.

[25] R. W. Raitt, personal communication, Dec. 16, 1932.
[27] Sienkiewicz, Physik. Zeits. **34**, 376 (1933).
[28] Benndorf, Physik. Zeits. **34**, 335 (1933); Akad. Wiss., Wien, Ber. [II a] **142**, 175 (1933).
[29] Lind and Roberts, J. Am. Chem. Soc. **42**, 1170 (1920).
[30] Evans, Phys. Rev. **46**, 328 (1934); Rev. Sci. Inst. **6**, 99 (1935).
[31] v. Schweidler, Physik. Zeits. **14**, 198 (1931); T. Ehrenfest, Physik. Zeits. **14**, 675 (1913).
[32] A useful table of $e^{-\lambda_1 t}$ is given in Meyer and Schweidler, *Radioaktivität* (1927), p. 419. Because of a misprint the value of $e^{-\lambda_1 t}$ for $t=3$ hr. should be 0.9776, instead of 0.9786, as there given.