

Radioactive Decay

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SECTION 1. INTRODUCTION

Here we shall treat the laws of spontaneous radioactive decay independently of the emission accompanying the transformation. This can be done because it happens that the law of decay is independent of the mechanism of the transformation. The results thus acquired can be applied to a great variety of cases, and they give a phenomenological explanation of a vast category of facts.

First, in Section 2 we shall neglect the fact that every substance contains an integral number of atoms and treat this number as a continuous variable. This procedure is legitimate if we deal with a great number of atoms and with processes in which a great number of atoms are involved. This continuum theory of the radioactive decay is exact for the average numbers of atoms in the sense that, if we treat many systems of radioactive atoms initially identical, the average number of atoms contained in the various systems at any subsequent time is given exactly by the continuum theory; on the other hand, each system may depart from the average, and for the study of these departures it is necessary to take into account the discontinuous, atomic nature of matter.

The effect of the discontinuous nature of matter is described in Section 3 together with some of its consequences and applications. Section 4 contains experimental methods for the determination of the decay constants of radioactive substances.

The decay law of radioactive substances was first clearly formulated and applied by Rutherford and Soddy (R8) as a result of their studies on the radioactivity of various substances, notably thorium, thorium X, and the emanations. In more or less explicit form it was known also to earlier investigators; the early work in this field is summarized in books by Curie (C7) and Rutherford (R6). Books by St. Meyer and Schweidler (M6) and Kohlrausch (K2) and an article by Bothe (B8) give complete information up to the date of their publication.

SECTION 2. CONTINUUM THEORY

A. One Substance

The fundamental law of radioactive decay can be formulated as follows: Given an atom; the probability that it decays during the time dt is λdt . The constant λ is called the decay constant. Dimensionally it is a reciprocal time, and it is characteristic of the given substance and of the mode of the decay. It is independent of the age of the atom considered and, as we shall see later, being a nuclear property is unaffected by all usual physical agents. This type of law is characteristic of "casual" events and is found in all types of radioactive decay: α , β , γ , orbital electron capture, spontaneous fission; and also in the atomic process of light emission by excited atoms.

The simplest application of this law is to a single radioactive substance of which we suppose we have initially $N(0)$ atoms. $N(0)$ is a large number, by hypothesis, so that we may consider $N(t)$, the number of atoms at time t , a continuously variable quantity. We can then write, according to our fundamental law, that $-dN$, the decrease in time dt of the number of atoms, is given by

$$-dN = \lambda N dt \quad (1)$$

which, integrated with the condition that initially we have $N(0)$ atoms, gives

$$N(t) = N(0)e^{-\lambda t} \quad (2)$$

Equation (2) is another formulation of the fundamental law of radioactive decay.

In practice one uses in addition to the decay constant its reciprocal $\tau = 1/\lambda$, called the mean life, and also the time T in which the number of atoms initially present is reduced by a factor 2. T is often called the "period" or "half-life" of the substance. The period is connected to the decay constant and to the mean life by

$$e^{-\lambda T} = e^{-T/\tau} = \frac{1}{2} \quad \text{or} \quad \lambda T = \log_e 2 = 0.6931472 \quad (3)$$

The name of mean life is given to τ because τ gives the average lifetime of the atoms. In fact, if we have initially $N(0)$ atoms, we will have $N(t) = N(0)e^{-\lambda t}$ at time t , according to Eq. (2). Of these, $N(t)\lambda dt$ will decay between times t and $t + dt$. The mean life is obtained by multiplying the last number by t , integrating with respect to dt between

0 and infinity, and dividing by the initial number of atoms present, $N(0)$:

$$\tau = \frac{1}{N(0)} \int_0^{\infty} \lambda t N(t) dt = \frac{1}{N(0)} \int_0^{\infty} N(0) e^{-\lambda t} \lambda t dt = \frac{1}{\lambda} \quad (4)$$

It is also easily seen from Eq. (4) that, if a radioactive substance kept on decaying at its initial rate $N(0)\lambda$, it would all disappear in a time τ .

Graphically, if we plot $N(t)$ versus t we obtain an exponential, and the tangent to the curve at $t = 0$ intercepts the t axis at a time τ . If we plot $\log N(t)$ versus t , we obtain a straight line: $\log N(t) = \log N(0) - \lambda t$. The slope of the straight line gives $-\lambda$. This second type of plot made directly on semilogarithmic paper is the most convenient and most commonly used.

B. More than One Substance

Very often one radioactive substance decays into another which is also radioactive. The two substances are then said to be genetically related; the first is called the parent or mother substance, the second the daughter. This relation is not limited to parent and daughter but extends sometimes over many "generations." Sometimes one substance can decay by two processes, e.g., alpha- and beta-emission giving rise to two different daughter substances; this occurrence is called dual decay or branching. We shall not consider branching now, but we will solve the following problem (B2). Given that at time $t = 0$ we have $N_1(0)$, $N_2(0)$ atoms of the radioactive substances 1, 2, etc., which are genetically related, find the number $N_1(t)$, $N_2(t)$, ... of atoms present at any subsequent time.

Substance 1 decays according to the law expressed in Eq. (1): $dN_1 = -N_1\lambda_1 dt$. For every atom of substance 1 disintegrating, an atom of substance 2 is formed; hence the number of atoms of substance 2 varies for two reasons: it decreases because substance 2 decays, but it increases because the decay of substance 1 continuously furnishes atoms of substance 2. The net change is given by

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad (5)$$

where λ_1 and λ_2 are the decay constants of the first and of the second substance, respectively. For a third substance we have, in a similar way,

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3, \text{ etc.} \quad (6)$$

This system of differential equations can be solved by putting

$$\begin{aligned} N_1 &= A_{11}e^{-\lambda_1 t} \\ N_2 &= A_{21}e^{-\lambda_1 t} + A_{22}e^{-\lambda_2 t} \\ N_3 &= A_{31}e^{-\lambda_1 t} + A_{32}e^{-\lambda_2 t} + A_{33}e^{-\lambda_3 t} \\ &\dots \\ N_k &= A_{k1}e^{-\lambda_1 t} + A_{k2}e^{-\lambda_2 t} + \dots + A_{kk}e^{-\lambda_k t} \end{aligned} \quad (7)$$

The constants A_{ki} are to be determined in such a way that expressions (7) satisfy the differential equations and the $N_k(0)$ have the prescribed initial values. Substituting expressions (7) into the differential equations (1), (5), (6), etc., we have

$$A_{ki} = A_{k-1,i} \frac{\lambda_{k-1}}{\lambda_k - \lambda_i} \quad (8)$$

This recursion formula is sufficient to determine all the A_{ki} with the exceptions of those with equal indexes.¹ These are determined by the initial conditions

$$N_k(0) = A_{k1} + A_{k2} + \dots + A_{kk} \quad (9)$$

It will be noticed that in each of the expressions (7) we have a sum of exponentials containing the decay constants of all the substances in the family preceding the one considered.

Some special cases of initial conditions are practically very important, notably that in which only substance 1 is initially present with $N_1(0)$ atoms. We have then, by a direct application of Eqs. (7), (8), and (9),

$$\begin{aligned} N_1(t) &= N_1(0)e^{-\lambda_1 t} \\ N_2(t) &= N_1(0) \frac{\lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \\ N_3(t) &= N_1(0) \lambda_1 \lambda_2 \left(\frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_3 - \lambda_2)(\lambda_1 - \lambda_2)} \right. \\ &\quad \left. + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right) \\ &\text{etc.} \end{aligned} \quad (10)$$

This case occurs quite often, e.g., in the active deposit of radium where

¹ If two or more of the decay constants involved should accidentally be equal, Eq. (8) cannot be applied and Eq. (7) has to be modified by replacing the exponentials with equal λ with terms of the form $t e^{-\lambda t}$, $t^2 e^{-\lambda t}$, etc.

N_1, N_2, N_3 represent, respectively, the number of atoms of $\text{RaA}, \text{RaB}, \text{RaC}$.

It is also important to see what happens to a mixture of radioactive substances left undisturbed for a long time. In Eqs. (7) there will be terms containing the exponential with the smallest decay constant λ_s of the mixture. All atoms of species 1, 2, \dots , s become in a relatively short time atoms of species s , and N_s itself is given by $N_s(t) = \mathfrak{N}e^{-\lambda_s t}$, where $\mathfrak{N} = N_1 + \dots + N_s$. If t is large enough ($t \gg 1/\lambda_s$), we will have

$$\begin{aligned} N_1(t) &= N_2(t) = \dots = N_{s-1}(t) = 0 \\ N_s(t) &= \mathfrak{N}e^{-\lambda_s t} \\ N_{s+1}(t) &= \mathfrak{N} \frac{\lambda_s}{\lambda_{s+1} - \lambda_s} e^{-\lambda_s t} \\ &\text{etc.} \end{aligned} \quad (11)$$

The ratio between the amount present of each of the substances, following substance s and substance s itself, is independent of time and is given by

$$\frac{N_i}{N_s} = \frac{\lambda_s \lambda_{s+1} \dots \lambda_{i-1}}{(\lambda_{s+1} - \lambda_s)(\lambda_{s+2} - \lambda_s) \dots (\lambda_i - \lambda_s)} \quad (12)$$

All substances decay with the same time law, $e^{-\lambda_s t}$. When this situation obtains, we speak of "transient equilibrium."

It happens sometimes that λ_s is very small compared with all the λ_i of the substances following the s th in the radioactive family and that during the time considered $e^{-\lambda_s t} \simeq 1$. Then we write

$$\frac{N_i}{N_s} = \frac{\lambda_s}{\lambda_i} \quad (13)$$

and we speak of "secular equilibrium." The interpretation of Eq. (13) is immediate: The number of atoms of substances $s, s+1$, etc., disintegrating per unit time is the same, and hence the number of atoms present is inversely proportional to the decay constant. This situation occurs in ores containing uranium. The decay constant of U^{238} is extremely small compared with those of the following products in the radioactive families, and the ores have been undisturbed for a time long enough to attain radioactive equilibrium. For all practical purposes the number of atoms in such an ore is independent of time as long as we limit ourselves to times up to a few thousand years, and Eq. (13) obtains for the radioactive families. However, it must be pointed out that the inactive lead isotopes which terminate the radioactive families

are not necessarily present in the amount corresponding to the age of the mineral because geochemical processes may remove either the lead or the uranium isotopes in a different way.

A brilliant experiment relative to radioactive equilibrium is the following: Suppose that we have a solution containing U^{238} in equilibrium with its daughter product UX_1 (Th^{234}), which has a period of 24.1 days. By a chemical procedure we separate, practically instantaneously, UX_1 from U . The UX_1 fraction then decays with the period 24.1 days. On the other hand, since this fraction and the mother solution, when considered together, must remain in equilibrium, an amount of UX_1 must grow in the mother solution exactly equal to the amount disappearing from the separated fraction. This example was studied quantitatively by Rutherford and Soddy, and it helped to elucidate the theory of radioactive decay. The decay and growth curves were incorporated in Lord Rutherford's escutcheon (E3).

Another important case found in practice is that of a radioactive substance formed at a constant rate and initially absent. The differential equation for this process is

$$\frac{dN}{dt} = Q - \lambda N \quad (14)$$

where Q is the number of atoms formed per unit time. Its solution with the condition $N(0) = 0$ is

$$N(t) = \frac{Q}{\lambda} (1 - e^{-\lambda t}) \quad (15)$$

Q/λ represents the number of atoms that one would obtain for $t \rightarrow \infty$; it is sometimes called the saturation number of atoms and it is indicated by N_∞ .

Sometimes a substance 1 decays into a substance 2 and then into a substance 3, but the radiations accompanying the transformation of 2 into 3 are undetectable. It is still possible to deduce, from the way in which substance 3 grows, that it does not derive directly from 1 but through an intermediate product. By direct development of the exponentials in powers of t in Eqs. (10) it will be noticed that substance 2 grows initially linearly with the time:

$$\begin{aligned} N_2(t) &= N_1(0)\lambda_1 t & N_2 \text{ grows linearly} \\ N_3(t) &= N_1(0)\lambda_1\lambda_2(t^2/2) & N_3 \text{ grows quadratically} \\ N_4(t) &= N_1(0)\lambda_1\lambda_2\lambda_3(t^3/2 \cdot 3) & N_4 \text{ grows cubically} \\ & \text{etc.} \end{aligned} \quad (16)$$

Applications of Eqs. (10) and (14) to various types of initial conditions of practical importance are given explicitly in (R7).

In nuclear reactors substances are created and then reirradiated by the neutrons of the reactor. This type of process is also governed by a generalization of Eqs. (4), (5), and (6). Thus, for instance, the amount of a substance of atomic number Z and mass number A in a pile varies according to the equation

$$\frac{dN_{Z,A}}{dt} = -N_{Z,A} \{ \lambda_{Z,A} + \varphi[\sigma_c(Z,A) + \sigma_f(Z,A)] \} + N_{Z,A-1}\varphi\sigma_c(Z,A-1) + N_{Z-1,A}\lambda_{Z-1,A} \quad (17)$$

where φ is the neutron flux, which might be a function of time, and the σ 's are capture and fission cross sections of the various substances. A study of these equations is to be found in (V1) and (B4).

An important practical case is the one in which the λ can be neglected compared to the $\varphi\sigma$ products. The initial approximate solution with only $N_{Z,A}$ present at time zero gives

$$\begin{aligned} N_{Z,A+1} &= N_{Z,A}(0)\sigma_c(A,Z)t\varphi \\ N_{Z,A+2} &= N_{Z,A}(0)\sigma_c(A,Z)\sigma_c(A+1,Z)\frac{t^2}{2}\varphi^2 \\ N_{Z,A+3} &= N_{Z,A}(0)\sigma_c(A,Z)\sigma_c(A+1,Z)\sigma_c(A+2,Z)\frac{t^3}{3!}\varphi^3 \end{aligned} \quad (18)$$

For more complicated cases the equations are easily treated with analogue computers.

C. Branching

Several radioactive substances decay by more than one mechanism, e.g., β^- and β^+ emission, alpha- and beta-emission. Let us consider alpha- and beta-branching and call the probability of alpha-emission by one atom, in time dt , $\lambda_\alpha dt$ and the probability of beta-emission by the same atom $\lambda_\beta dt$. Then the total probability of decay of the atom in time dt by either alpha- or beta-disintegration is $(\lambda_\alpha + \lambda_\beta) dt$. Hence

$$\frac{dN}{dt} = -(\lambda_\alpha + \lambda_\beta)N \quad (19)$$

and the mean life of the substance, defined as the time in which the amount initially present is reduced from 1 to $1/e$, is

$$\tau = \frac{1}{\lambda_\alpha + \lambda_\beta} \quad (20)$$

The ratio of the number of alpha- to beta-particles emitted during a certain time is called the branching ratio and is equal to $\lambda_\alpha/\lambda_\beta$. Sometimes the quantities $\tau_\alpha = 1/\lambda_\alpha$ and $\tau_\beta = 1/\lambda_\beta$ are used and are called, not too properly, the mean life for alpha- and for beta-decay, respectively. From Eq. (20) it follows immediately that

$$\frac{1}{\tau} = \frac{1}{\tau_\alpha} + \frac{1}{\tau_\beta} \quad (21)$$

A similar terminology is used in other types of branching.

D. Units and Standards in Radioactive Measurements

Almost all radioactive measurements are made for the purpose of establishing the number of atoms present at a certain time. This is done according to Eq. (1) by determining the number decaying per unit time. The number of particles emitted can be determined by measuring the charge accumulating on the sample as a consequence of this emission, and, if one knows that one particle is emitted per disintegration, the charge accumulated gives directly the number of atoms disintegrated.

This direct method originated with Wien and Rutherford. It has been recently revived by Clark (C3) but is seldom used, and almost always the detecting apparatus actually measures either the ionization produced by the radiations emitted at the instant of decay or some other effect of these radiations. It is then necessary to establish the effect on the detecting apparatus of the radiations accompanying the decay of one atom. This operation is called the determination of the efficiency of the detecting apparatus. In some cases it is relatively simple. Thus it is sometimes possible to obtain standards containing a known amount of a radioactive substance from the National Bureau of Standards or a similar agency.

Given a standard of a particular substance, the determination of the efficiency of the detecting apparatus is very simple. All one needs to do is to measure the standard with the detecting apparatus. The limits to the method come primarily from the fact that calibrated standards are available only for relatively few substances.

A primary standard of radium was prepared in 1911 by Mme. M. Curie. It originally contained 21.99 mg of radium, and it was deposited at the Bureau des Poids et Mesures at Sèvres (W2). Another primary standard, prepared by O. Hoenigschmid and containing 31.17 mg of radium, was deposited at the Radium Institute of Vienna. The original primary standards have now been replaced by new ones deposited

at the same places (F1, M1). Secondary standards have been prepared by comparison of their gamma-radiation and that of these standards and are deposited in the corresponding laboratories of various countries.

For substances other than radium, standards are available from several sources at present. In the United States the National Bureau of Standards is prepared to supply the standards indicated in Table 1. The methods used for calibration of the single nuclides are indicated in (M1) and also in (M2). Oak Ridge National Laboratory can also supply calibrated standards as indicated in Table 2. The intercalibration of standards proceeds continuously, and a recent report is given in (M1).

Under favorable conditions the determination of the efficiency of the detecting apparatus, even without a standard, may be relatively simple; e.g., for an alpha-emitter, deposited as a plane thin layer in an ionization chamber connected to a linear amplifier, the number of pulses registered is practically equal to half the number of particles emitted, apart from a small correction due to back scattering by the support of the sample (N5, F0, G1a), and hence to half the number of atoms decayed. On the other hand, the determination of the efficiency of a Geiger-Müller counter for a substance decaying by orbital electron capture may be a matter of serious difficulty. It is impossible to give general methods for the determination of the efficiency of detecting apparatus. A few examples, however, will give an idea of some of the procedures used. Since counting of alpha-particles is one of the easiest and most reliable methods, it is used to determine the efficiency of beta-counters as well, if the beta-emitter decays into an alpha-emitter as in the succession $\text{RaD} \rightarrow \text{RaE} \rightarrow \text{Po(N2)}$. If the RaD , RaE , and Po are in equilibrium, the number of atoms of RaE disintegrating per unit time is practically equal to the number of Po atoms disintegrating per unit time. The latter can be counted by their alpha-emission, and hence the number of RaE atoms disintegrating is also known. The efficiency of the detector for RaE , e.g., a Geiger-Müller counter of wall thickness sufficient to stop the alpha-particles of polonium and the weak radiations of RaD , is then determined by dividing the number of pulses registered in a certain time by the number of alphas emitted during the same time.

Another method, which is applicable to substances of which the decay scheme is known and which contains a beta-emission followed by a single gamma-emission, involves the following procedures. Two counters are used, the first being insensitive to beta-rays and having an efficiency η_γ for gamma-rays, and the second being sensitive to beta-rays only and having an efficiency η_β . If N disintegrations have oc-

TABLE 1

NBS RADIOACTIVITY STANDARDS—ALPHA-, BETA-, AND GAMMA-STANDARDS

Sample No.	Radiation	Nuclide	Nominal Activity ^a	Volume	Chemical Form of Standard
4900	α	^b Po ²¹⁰	200 dps	(c)	Plated from polonium chloride
4901	α	^b Po ²¹⁰	500 dps	(c)	" " " "
4902	α	^b Po ²¹⁰	1000 dps	(c)	" " " "
4903	α	^d U ₃ O ₈	15 dps	(e)	" " " "
4910	β (α)	^f RaD + E	200 dps	(c)	Lead peroxide
4911	β (α)	^f RaD + E	500 dps	(c)	" " " "
4912	β (α)	^f RaD + E	1000 dps	(c)	" " " "
4913	β (γ)	Co ⁶⁰	10 ⁴ dps/ml	(g)	Chloride in HCl solution
4914	γ (β)	Co ⁶⁰	10 ⁴ dps	5.0 ml	" " " "
4915	γ (β)	Co ⁶⁰	10 ⁴ dps	5.0 ml	" " " "
4916	β	^h P-32	10 ⁴ dps/ml	(g)	Phosphoric acid solution
4917	β (γ)	^h I-131	10 ⁴ dps/ml	(g)	Sodium iodide solution
4918	β (γ)	^h Au-198	10 ⁴ dps/ml	(g)	Auric cyanide solution
4919	β	^h Sr-90	10 ⁴ dps/ml	(g)	Chloride in HCl solution
4920	β	^h Y-90	10 ⁴ dps/ml	(g)	Thallic nitrate in HNO ₃ solution
4921	β (γ)	^h Tl-204	10 ⁴ dps/ml	(g)	Chloride in HCl solution
4922	γ (β)	Na ²²	10 ⁴ dps	5.0 ml	" " " "
4923	β (γ)	^h Na-24	10 ⁴ dps/ml	(g)	" " " "
4924	β	C ¹⁴	10 ⁴ dps/ml	25.0 ml	Sodium carbonate solution
4925	β	C ¹⁴	10 ⁴ dps/g	"	Benzoic acid
4926	β	H ³	10 ⁴ dps/ml	25.0 ml	Tritiated water solution
4927	β	H ³	10 ⁴ dps/ml	(g)	" " " "
4928	β	S ³⁵	10 ⁴ dps/ml	(g)	Sodium sulfate solution
4929	K	Fe ⁵⁵	10 ⁴ dps/ml	(g)	Ferric chloride in HCl solution
4930	K (γ)	Zn ⁶⁵	10 ⁴ dps/ml	(g)	Zinc chloride in HCl solution
4931	γ (β)	Cd ¹⁰⁹	10 ⁴ dps/ml	5.0 ml	Chloride in HCl solution
4932	γ (β)	Hg ²⁰³	10 ⁴ dps/ml	5.0 ml	Mercurous nitrate in solution

Radium Standards (for Radon Analysis)

Sample No.	Radium Content (grams)	Volume (ml) ^f	Sample No.	Description	Radium Content (grams)	Volume (ml) ^g
4950	10 ⁻⁸	100	4955	Radium	0.1 × 10 ⁻⁶	5
4951	10 ⁻¹¹	100	4956	Radium	0.2	5
4952	Blank solution	100	4957	Radium	0.5	5
			4958	Radium	1.0	5
			4959	Radium	2.0	5
			4960	Radium	5.0	5
			4961	Radium	10	5
			4962	Radium	50	5
			4963	Radium	50	5
			4964	Radium	100	5

^a The disintegration rate as of the reference date is given on a certificate accompanying the standard.
^b Samples consist of Po²¹⁰, deposited on a silver disk 1 in. in diameter, 1/4 in. thick, and faced with 0.002 in. of palladium.
^c Deposited source.

^d Deposited source.

^e Deposited source.

^f Deposited source.

^g Deposited source.

TABLE 1 (Continued)

NBS RADIOACTIVITY STANDARDS—ALPHA-, BETA-, AND GAMMA-STANDARDS

Sample No.	Method of Primary Standardization	Method of Secondary Standardization
4900	2 π prop. counting	
4901	" " "	
4902	" " "	
4903	" " "	
4910	Quantitative extraction from ditchblende	Defined-geometry G.-M. counter
4911	" " "	" " "
4912	" " "	" " "
4913	4 π prop. counting; coin. counting	4 π ion. chamber; formamide counting
4914	" " "	" " "
4915	" " "	" " "
4916	4 π prop. counting	2 π ion. chamber; formamide counting
4917	4 π prop. counting; coin. counting	2 π , 4 π ion. chambers; formamide counting
4918	" " "	" " "
4919	4 π prop. counting	2 π ion. chamber; formamide counting
4920	" " "	" " "
4921	4 π prop. counting; coin. counting; Co ⁶⁰ comparison	4 π ion. chamber
4922	4 π prop. counting; coin. counting; Co ⁶⁰ comparison	" " "
4923	4 π prop. counting; coin. counting	4 π ion. chamber
4924	Compensated gas counting	Formamide counting; CO ₂ ion. chamber; liq. scint. counting
4925	" " "	Formamide counting; CO ₂ ion. chamber; liq. scint. counting
4926	Calorimeter; compensated gas counting	Liq. scint. counting; H ³ ion. chamber
4927	" " "	" " "
4928	Calorimeter; 4 π prop. counting	Liq. scint. counting; formamide counting
4929	Liq. scint. counting	Liq. scint. counting; x-ray counting
4930	Liq. scint. counting; coin. counting	" " "
4931	4 π prop. counting; coin. counting	4 π ion. chamber
4932	" " "	" " "

^d Samples consist of U₃O₈ deposited on a 0.1-mm platinum foil and mounted on an aluminum disk 1/2 in. in diameter and 1/2 in. thick. The alpha-ray disintegration rate as of the date of calibration is indicated on the certificate accompanying the standard.

^e Evaporated source.

^f Standards consist of Pb²¹⁰-Bi²¹⁰ in equilibrium, deposited on a silver disk 1 in. in diameter, 1/4 in. thick, and faced with 0.002 in. of palladium.

^g Approximately 3 ml of low-solids carrier solution containing the active nuclide in a flame-sealed ampoule.

^h Distributed periodically at announced intervals.

ⁱ Benzoic acid for use in liquid scintillation counters.

^j Samples are sealed in glass containers. They were prepared by determining the radium content of a purified sample of radium bromide by direct comparison with the U. S. primary radium standard with a gold leaf electroscop. The radium bromide was then transferred quantitatively to a carrier solution consisting of 0.2% by weight BaCl₂·2H₂O in a 5% by weight solution of HCl.

^k Samples are contained in flame-sealed glass ampoules. They were prepared by determining the radium content of a purified sample of radium bromide by direct comparison with the U. S. primary radium standard 25 by means of the gold leaf electroscop. The radium bromide was then dissolved quantitatively in a 5% by weight solution of HNO₃.

current in the sample, the counting of the two counters will be

$$\begin{aligned} C_\gamma &= N\eta_\gamma \\ C_\beta &= N\eta_\beta \end{aligned} \quad (22)$$

where C_γ and C_β are the numbers of counts registered by the two counters respectively. If we count also the number of counts registered simultaneously (coincidences) by the two counters, we have

$$C = N\eta_\beta\eta_\gamma \quad (23)$$

From these three equations we find N , η_β , and η_γ . This method is susceptible to many variations and is finding ever wider applications.

Particularly susceptible to coincidence methods of counting are the positron emitters. Each positron on annihilation emits two gamma-rays in opposite directions and with the same energy. With modern scintillation counters the gamma-rays can be detected with good efficiency, and the geometrical correlation ensures that, if one gamma-ray goes through one (defining) counter subtending a measured solid angle Ω , the other goes through the opposite one subtending a slightly larger solid angle Ω' .

If η is the probability of detecting one gamma-ray crossing a detector, Eqs. (22) and (23) are replaced by Eqs. (24):

$$\begin{aligned} C_1 &= N\Omega\eta \\ C_2 &= N\Omega'\eta \\ C &= N\Omega\eta^2 \end{aligned} \quad (24)$$

where η is the same for both counters.

Another method of absolute counting involves a counter subtending a complete solid angle (4π). Such counters are discussed in Section 1C of Part XI.

Finally, rather crude results can be obtained for beta-emission by determining the counting rate of a sample by a Geiger-Müller counter, measuring the absorption curve of the radiation, and extrapolating to zero thickness. This method becomes unreliable if conversion electrons are present. However, since it has very wide application, it has been studied in detail and, with standardized geometries, types of counters and absorbers, is capable of fair precision. Detailed procedures for commercial counters are given in references (N6), (S4), and (Z1).

The efficiency of the counting system is especially difficult to determine in cases of orbital electron capture, and one often has to use artifices suited to the special problem. We cite as an example Be^7 , in which the x-rays are extremely difficult to detect and a gamma-ray is only occasionally emitted. Nevertheless, it has been possible to deter-

mine what fraction f of the decaying atoms emits the characteristic gamma-ray of 478 kev. This was done by preparing the Be^7 by a $\text{Li}^7(p, n)$ reaction and counting the neutrons formed. For each neutron an atom of Be^7 is generated, and in this way it is possible to know how many of them are present (T2). Calling this number N , we have $\nu_\gamma = N\lambda f$, where ν_γ is the number of gamma-rays emitted per second by the source and λ is the decay constant of Be^7 . For the use of x-rays to standardize orbital electron capturers see (A0).

It is expedient to define a unit of "activity," i.e., to give a special name to the amount of substance that undergoes a certain number of disintegrations per second. In the early days of radioactivity when the main substances available were the members of the uranium-radium family, it was decided to define the unit of activity (the curie) as the activity of a product in the Ra family in equilibrium with 1 gram of Ra(II). This definition clearly does not give explicitly the number of disintegrations per second until the decay constant of Ra is known; once this constant is known, the number of disintegrations per second corresponding to 1 curie is given by

$$6.023 \times 10^{23} \frac{\lambda}{A} = C \quad (25)$$

in which A is the atomic weight of Ra, λ is its decay constant in sec^{-1} , and the numerical constant is Avogadro's number. This definition is directly applicable only to the radium family, but, since measurements have given values of C around 3.7×10^{10} , it has become customary to use the word curie for the activity of a sample undergoing 3.7×10^{10} disintegrations per second, even for substances not in the radium family, and to assume as the definition of the curie the amount of any radioactive substance undergoing 3.7×10^{10} disintegrations per second (C8).

Actually, in 1948 the International Council of Scientific Unions asked the International Union of Pure and Applied Chemistry and the International Union of Pure and Applied Physics to nominate representatives to a new Joint Commission on Standards, Units, and Constants of Radioactivity. The new Joint Commission, the official successor of the International Radium Standard Commission, invited the surviving members of the former commission to serve as advisory councilors. At its meeting in Paris in July, 1950, the Joint Commission officially redefined the curie unit. The new definition of the curie is: "The curie is a unit of radioactivity defined as the quantity of any radioactive nuclide in which the number of disintegrations per second is 3.700×10^{10} ." This new definition divorces the curie unit from its former special association with radon and radium. For example, if it should be later agreed that 1 gram of radium has, say, 3.63×10^{10} disintegrations per second, then

1 gram of radium would have an activity of $3.63/3.700 = 0.98$ curie. The arbitrary coefficient 3.700 (instead of, say, unity) was chosen so that the new curie will be substantially equivalent to the old curie when radon is involved, and will be identical with the unofficial curie which was previously used widely by many workers in artificial radioactivity. The curie has now become an official unit of disintegration rate (P1).

This definition of the curie avoids the most perplexing and inconvenient feature that would be introduced by the definition contained in Eq. (25), according to which a new and better determination of λ would change the numerical value of the disintegration per second corresponding to 1 curie. The number 3.7×10^{10} is the value recommended by the International Radium Commission in 1931 (I1) for the number of alpha-particles emitted per second by 1 gram of radium. The determinations of this number used by the Commission and some more recent ones have been discussed by Rutherford (R6). A more recent determination and a discussion of the subject up to 1947 will be found in (K3). In this paper Kohman suggests 3.62×10^{10} as the best value of the number of disintegrations per gram of radium per second. See also (K4).

Recently it has been also proposed to introduce a unit of activity corresponding to 10^6 disintegrations per second and to call it "Rutherford," abbreviated rd (C4), but with the new definition of the curie there is no need for such a unit, and it has been practically abandoned.

Other units have also been proposed for special purposes but are now rarely used; such are the eman, a unit of concentration of radon in air corresponding to 10^{-13} curie/cm³, and the Mache unit, also a unit of concentration defined as that quantity of radon per liter which, without decay products and with complete utilization of the alpha-particles, can maintain by its ionization of air a saturation current of 10^{-13} esu (I1).

E. Dosimetry

Another approach to the definition of units of radioactivity is to consider the effect of the radiations instead of the number of disintegrations per unit time. We can speak, e.g., of 1 gram radium equivalent for a MsTh sample, meaning that under certain conditions of filtration of the gamma-rays (5 mm of lead) the ionization produced in a certain instrument by the two substances is the same (I1). It is clear that this type of definition is dependent on the filtration of the radiations emitted by the various substances and on the instrument used for detecting the radiations. For this reason it must be used cautiously and with proper specification of the experimental conditions. However, it is often the only usable definition because the efficiency of the measuring instru-

ments for the particular substance studied has not been determined. From this point of view it is possible to measure the ionizing action of a radioactive substance emitting gamma-rays contained in a source by specifying that it gives a certain number of "roentgens" per hour at 1 meter distance (rh) (C8). The roentgen is defined as "that quantity of X or gamma-radiation such that the associated corpuscular emission per 0.001293 gram of air produces in air ions carrying 1 esu of quantity of electricity of either sign" (R1). The mass of air referred to is the mass of 1 cm³ of dry air at 0° C and atmospheric pressure. If we assume that an average of 32.5 ev is expended to produce a pair of ions in air (G3)—but see also page 233 of Part II in Volume I—we find that 1 roentgen corresponds to the absorption of

$$\frac{32.5 \times 1.60 \times 10^{-12}}{4.80 \times 10^{-10}} = 0.108 \text{ erg cm}^{-3} \text{ of air}$$

or

$$\frac{0.108}{1.293 \times 10^{-3}} = 83.8 \text{ erg gm}^{-1} \text{ of air}$$

or 6.77×10^4 Mev cm⁻³ of air.

For several substances for which the decay scheme is known it is possible, using the data of Fig. 1, to calculate the rhm corresponding to one disintegration per second and hence to deduce from an ionization measurement the activity of the sample.

This has been done for the isotopes listed in Table 3, reported from Evans (E2), according to whom the figures should be accurate to ± 3 percent. Only the ionization produced by gamma-rays and annihilation radiation is considered. This corresponds to a filtration through a few hundred mg cm⁻² of a very light element. Isotopes that do not emit gamma-radiation have zero mrhm.

For radium in equilibrium with its short life products, which is particularly important because this substance is often used for calibration purposes, the effect of filtration of the gamma-rays is reported as follows (E2):

Filter (mm Pt)	0	0.25	0.5	2.5	5.0
rh/g Ra	0.96	0.88	0.84	0.63	0.46

Similar data are also reported in (V2).

For beta-activity the problem is complicated by the scattering and absorption of the beta-rays and by the continuous nature of the beta-spectrum. For several substances the average energy of the beta-rays has been computed (M1, C2, K1). This information allows one to calculate the disintegration rate from an ionization measurement made under suitable conditions, if it is assumed that the formation of a pair of ions in air requires 32.5 ev.

TABLE 3

MILLIROENTGENS PER HOUR PRODUCED AT 1 METER (MRHM) BY THE NUCLEAR GAMMA-RAYS AND THE ANNIHILATION RADIATION FROM 1 MILLICURIE ($\equiv 37.00 \times 10^6$ DISINTEGRATIONS PER SECOND) OF SEVERAL NUCLIDES

Small effects such as the bremsstrahlung from inelastic collisions of beta-rays with nuclei are neglected, as are also the soft characteristic x-rays emitted after orbital electron capture transitions, the small deficit of gamma-rays in some transitions where internal conversion has been measured (e.g., ca. 0.5 percent in I^{130}), and the absorption of the gamma-rays and annihilation radiation in 1 meter of standard air (~ 0.3 percent). The accuracy of the values for mrhm is the same as our knowledge of the nuclear gamma-ray energies, which is about $\pm 1-3$ percent.

Nuclide	mrhm/1 mc	Nuclide	mrhm/1 mc
Na ²²	1.29	Br ⁸²	1.50
Na ²⁴	1.92	I ¹²⁸	0.018
Mn ⁵²	1.93	I ¹³⁰	1.25
Mn ⁵⁴	0.49	I ¹³¹	0.24
Fe ⁵⁹	0.65	Ta ¹⁶²	0.6
Co ⁵⁸	0.56	Au ¹⁹⁸	0.58
Co ⁶⁰	1.31	RaB + C	
Cu ⁶⁴	0.12	with 0.5 mm Pt	
Zn ⁶⁵	0.30	filtration	0.84
As ⁷⁶	0.4		

In practice, measurements of roentgens are obtained by using small air ionization chambers having the walls of a substance which behaves with respect to beta- and gamma-rays not very differently from air at very high pressure. One can use then an approximate theorem of Gray (G3) and Bragg which states that the ionization produced by the gamma-rays in the chamber is unaffected by the walls and is the same as would be obtained in an equal volume of an extended gaseous medium. This theorem is valid if the linear dimensions of the cavity are small compared with the range, in the gas filling the cavity, of the secondary corpuscular radiation (electrons) produced in the wall of the cavity. Chambers satisfying this condition with walls made of suitable plastics are called air chambers and are commercially available, being extensively used for medical x-ray dosimetry.

The Gray-Bragg relation can be written in the form

$$J = \frac{E}{\rho w} \quad (26)$$

where J is the number of ion pairs produced per unit volume of the gas, ρ is the ratio of the stopping power of the walls to the stopping power of the gas for the equilibrium radiation; E is the energy absorbed per unit volume of the walls; for an ideal case of an air chamber ρ is simply

the ratio of the densities of the walls and gas and w is the energy spent per ion pair formed (see Volume I, Part II, page 233).

The energy flux required to produce one roentgen depends on the frequency of the radiation considered. It has been calculated for a large energy interval (F0a), and the results are summarized in Fig. 1.

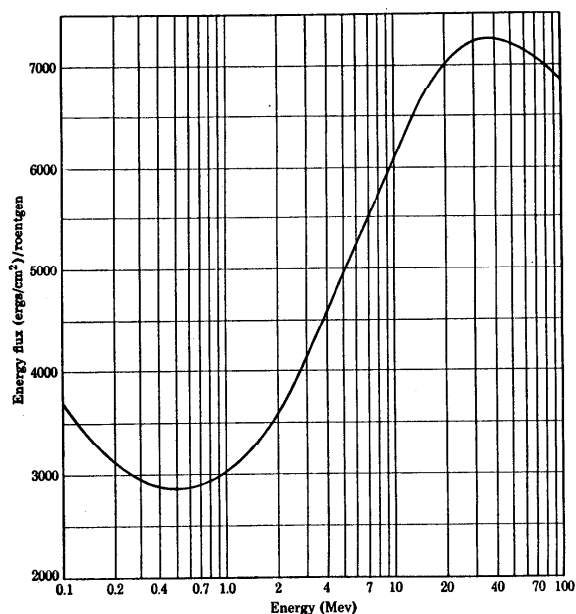


Fig. 1. Energy flux of gamma-rays required to produce 1 roentgen in an air cavity as a function of the energy of the quanta of the radiation (data from F0a).

It must be borne in mind that in order to use this curve correctly the thickness of the wall must be sufficient to establish equilibrium between the primary radiation and its secondary; thus for 10 Mev about 4 g cm^{-2} of wall thickness is required. The ionization depends on the chemical composition of the walls, and according to the definition of the roentgen the walls should be of air. The data are computed for $w = 32.5 \text{ ev}$ and, if one uses a different value of w , the ordinates vary proportionately. For data above 10 Mev compare (J3).

A discussion of the applicability of Eq. (26) is given in (M3) and (G1).

In addition to the methods of dosimetry based on the ionization produced in a gas by the radiation studied, the recent introduction of extremely powerful sources has required the development of measuring methods for doses previously unattained.

A useful method is based on the chemical reactions produced by radiations in solutions containing Fe^{2+} or Ce^{4+} ions. In the former case a solution of Fe_2SO_4 in 0.8N sulfuric acid, saturated with oxygen, is used, and calibrations (A1, M7) show that 20.3 ± 1 ferric ions are produced for every 100 ev spent by the radiation in the solution. Similar results have been obtained for the reduction of ceric sulfate. Some types of chemical dosimeters contain reagents and indicators contrived in such a way as to change color as a function of the irradiation suffered. The changes in color of specially sensitized alkali halide crystals have also been used (F2).

Photographic dosimetry has been in use for many years. It is based on the blackening of photographic film under the action of radiations. For x-rays it has been shown that the density obtained, i.e., $\log(I_0/I)$, where I_0/I is the ratio between the incident and transmitted light for the film in question, is a function of the total dose to which the film has been exposed and not of the time distribution of the dose (reciprocity law). Photographic film is valuable in the dose range from 10^{-2} r to 10^4 r approximately; it is available in a variety of emulsions and can be used for beta- and gamma-rays (D3, E1, C5).

Biological Aspects of Dosimetry. Dosimetry of radiation is of great importance in biological work, and it is not yet in a completely satisfactory state.

The roentgen as defined above is a perfectly well-defined unit, but it is not always very convenient to use. It is specifically referred to air, and in most cases the objects irradiated are not air.

It is convenient for many biological problems to refer the dose to the number of ergs absorbed per gram of substance. In a beam delivering 1 r, 1 gram of soft tissue absorbs 93 ergs approximately. In the same beam a different substance might absorb a different amount of energy; for instance, air would absorb 83.8 ergs, as shown on page 17. The dose corresponding to the absorption of 93 ergs per gram of soft tissue is called "roentgen equivalent physical" and abbreviated r.e.p. There is also in use another unit of absorbed dose called "rad," which corresponds to 100 ergs per gram.

The biological effect of radiation is again a function not only of the dose and quality of the radiation, but also of the organ and tissue receiving the radiation.

Whereas the establishment of the energy spent by the radiation is a relatively simple problem of physics, the evaluation of its biological effect is a much more difficult one, and one which is not even very well defined, because two radiations equivalent for a given biological effect might not be equivalent for another one. Nevertheless, some tentative numbers, referring to the "relative biological effectiveness" (RBE), have been put forward by the National Committee on Radiation Protection for various types of radiations and are summarized in Table 4.

TABLE 4

RECOMMENDED VALUES OF THE RELATIVE BIOLOGICAL EFFECTIVENESS (RBE) OF RADIATION OF DIFFERENT SPECIFIC IONIZATIONS APPLICABLE TO EXPOSURE TO RADIATION FROM EXTERNAL SOURCES ^{a,b}

(X-rays, electrons, and positrons of any specific ionization: RBE = 1)

Heavy Ionizing Particles

Average Specific Ionization ^c (ion pairs per micron of water)	RBE ^d	Average Linear Energy Transfer (LET) to Water ^e (kev per micron of water)
100 or less	1	3.5 or less
100 to 200	1 to 2	3.5 to 7.0
200 to 650	2 to 5	7.0 to 23
650 to 1500	5 to 10	23 to 53
1500 to 5000	10 to 20	53 to 175

^a Present knowledge of the biological effectiveness of radiation of different specific ionizations does not warrant fine distinctions. Therefore ranges rather than individual figures are given in this table. For any range of specific ionization it is safer to use the higher of the two values of RBE given for that range, but a value obtained by linear interpolation is acceptable.

^b The critical organs and effects considered are: skin with respect to cancer, blood-forming organs with respect to leukemia, gonads with respect to impairment of fertility, and lenses of the eyes with respect to cataracts.

^c Specific ionization is expressed in ion pairs per micron of water in terms of its air equivalent.

^d RBE is in terms of the permanent biological effectiveness of ordinary x-rays for which the average specific ionization in the tissue of interest is assumed to be 100 ion pairs per micron of water.

^e Linear energy transfer is given in kev per micron of water, using 35 ev per ion pair.

It has been found convenient in practical biological work to express doses of radiation of different specific ionizations in terms of a unit that embodies both the magnitude of the dose and its biological effectiveness. The unit has been called the "rem" and has been defined by the relation

$$\text{"rems"} = \text{"reps"} \times \text{RBE} \quad (27)$$

The problem of a safe maximum dose for radiation exposure in man is still far from being solved; however, the U. S. National Committee on Radiation Protection has given a set of protection rules (N4). The basic one is: "For adults under 45 years of age whose entire body, or major portion thereof, is exposed to ionizing radiation from external sources for an indefinite period of years, the maximum permissible weekly doses shall be 300 millirems in the bloodforming organs, the

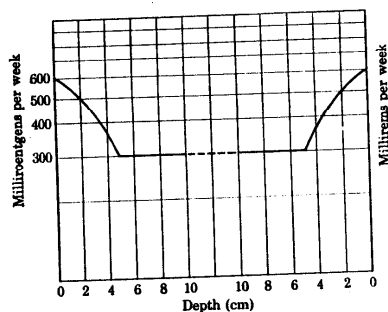


Fig. 2. Basic permissible dose distribution in the main portion of the body. The basic permissible weekly dose is 600 mr for the skin, 300 mr for the blood-forming organs at an assumed average depth of 5 cm, and according to the curve for the tissues at other depths from the surface of the main portions of the body. The basic permissible dose distribution remains the same when the basic permissible doses are expressed in rems (N4).

gonads, and the lenses of the eyes; 600 millirems in the skin; and the respective values of the weekly doses in millirems in all other organs and tissues of the body according to the basic permissible dose distribution. For persons 45 years of age or older similarly exposed, the corresponding maximum permissible total weekly doses shall be double the above stated values, provided that the portion of the weekly dose in the lenses of the eyes contributed by radiation of high specific ionization does not exceed 300 millirems." The basic permissible dose distribution mentioned above requires that the weekly dose at depths greater than 5 cm shall not exceed 300 millirems, as shown in Fig. 2, taken from (N4). Indispensable complements to this rule and the maximum permissible amount of radioisotopes in the human body and other safety rules will be found in (N1), (N2), (N3), and (N4).

The whole subject of radiation dosimetry is extensively treated in a recent book (H2).

SECTION 3. FLUCTUATIONS IN RADIOACTIVE DECAY

A. General Theory

In Section 1 we treated radioactive decay from the point of view of a continuous change of the number of atoms. Since this number is obviously an integer, it is clear that the theory previously given is only approximate, and, although it holds exactly for the average values in the limit of a very large number of atoms, we may expect departures from it in cases in which numbers of atoms or events are considered.

It is the purpose of this section to study the fluctuations or differences between the actual number of decaying atoms n and the average number \bar{n} .

The first problem to be treated is the following: We have a substance with an extremely long life (so long that we can neglect its average decay in the time considered) and emitting on the average \bar{n} particles per second. What is the probability $P(n)$ that in a given second it emits n particles?

This classical problem in the theory of probability is solved by Poisson's formula:

$$P(n) = \frac{\bar{n}^n}{n!} e^{-\bar{n}} \quad (28)$$

To prove this equation we divide our time interval of 1 sec into K equal parts. K is an arbitrary large number.

The probability of a disintegration occurring in any one of the K subintervals is then \bar{n}/K , and the probability that no disintegration occurs in any specified interval is $1 - \bar{n}/K$. The probability that we have a disintegration in n , and only n , specified subintervals is

$$\left(\frac{\bar{n}}{K}\right)^n \left(1 - \frac{\bar{n}}{K}\right)^{K-n} \quad (29)$$

The first factor in Eq. (29) comes from the requirement that n specified subintervals contain a disintegration, and the second from the requirement that the remaining $K - n$ do not contain a disintegration. If we now abandon the condition that the disintegrations must occur in specified subintervals and retain only the requirement that they must occur, we have to sum Eq. (29) for all possible choices of the specified subintervals. These are equal in number to the combinations of K

objects taken n at a time:

$$\binom{K}{n} = \frac{K(K-1)\cdots(K-n+1)}{n!} = \frac{K!}{n!(K-n)!} \quad (30)$$

We thus have

$$P_K(n) = \binom{K}{n} \left(\frac{\bar{n}}{K}\right)^n \left(1 - \frac{\bar{n}}{K}\right)^{K-n}$$

This formula can be reinterpreted in a more general way. We try repeatedly a certain experiment which must have one of two results, E and F , which are mutually exclusive. The probability of E occurring in each trial is p ; the probability of F occurring is $1 - p = q$. What is the probability, in K trials, that E occurs n times, and hence F occurs $K - n$ times? The answer, as seen above, is

$$P_K(n) = \binom{K}{n} p^n q^{K-n} \quad (31)$$

It will be noticed that $P_K(n)$ is the term containing $p^n q^{K-n}$ of the binomial development of $(p + q)^K$, and hence

$$\sum_{n=0}^K P_K(n) = (p + q)^K = 1 \quad (32)$$

Equation (32) verifies that the probabilities $P_K(n)$ are normalized correctly.

Before considering certain limiting cases of formula (30), we shall calculate the average value \bar{n} of the number of times that event E occurs in a series of K trials. Intuitively we expect it to be $\bar{n} = Kp$, and this is borne out by direct calculation. By definition,

$$\bar{n} = \sum_{n=0}^K n P_K(n) = \sum_{n=0}^K n \binom{K}{n} p^n q^{K-n} \quad (33)$$

On the other hand,

$$\frac{\partial}{\partial p} (p + q)^K = \frac{\partial}{\partial p} \sum_{n=0}^K \binom{K}{n} p^n q^{K-n} = \sum_{n=0}^K n \binom{K}{n} p^{n-1} q^{K-n}$$

Sec. 3A] Fluctuations in Radioactive Decay

and hence, using Eq. (33),

$$\bar{n} = p \frac{\partial}{\partial p} (p + q)^K = pK(p + q)^{K-1} = Kp \quad (34)$$

because $p + q = 1$.

We now calculate the average value of $(n - \bar{n})^2 = (n - Kp)^2$, which is called variance by modern statisticians. Its square root is the "standard deviation" and is indicated by σ . We have first

$$\sigma^2 = \overline{(n - Kp)^2} = \bar{n}^2 - 2pK\bar{n} + p^2K^2 = \bar{n}^2 - p^2K^2 = \bar{n}^2 - \bar{n}^2 \quad (35)$$

and \bar{n}^2 is evaluated, in a way similar to that used above for \bar{n} , as follows:

$$\begin{aligned} \bar{n}^2 &= \sum_{n=0}^K n^2 P_K(n) = \left(p^2 \frac{\partial^2}{\partial p^2} + p \frac{\partial}{\partial p}\right) \sum_{n=0}^K \binom{K}{n} p^n q^{K-n} \\ &= \left(p^2 \frac{\partial^2}{\partial p^2} + p \frac{\partial}{\partial p}\right) (p + q)^K = K(K-1)p^2 + Kp \end{aligned} \quad (36)$$

from which, remembering that $q = 1 - p$, we get

$$\sigma^2 = \bar{n}^2 - p^2K^2 = -Kp^2 + Kp = Kpq$$

or

$$\sigma = \sqrt{Kpq} \quad (37)$$

The last expression for the standard deviation is of considerable importance.

We now make use of the fact that K is an arbitrarily large number, and we pass to the limit for K tending to infinity. We make use of the well-known formulas

$$\lim_{K \rightarrow \infty} \binom{K}{n} = \frac{K^n}{n!} \quad \text{and} \quad \lim_{K \rightarrow \infty} \left(1 - \frac{\bar{n}}{K}\right)^{K-n} = e^{-\bar{n}}$$

and, substituting these values in Eq. (30), we have

$$P(n) = \frac{\bar{n}^n}{n!} e^{-\bar{n}} \quad (38)$$

where we have dropped for $K \rightarrow \infty$ the index K . This is the famous

Poisson formula; it is illustrated in Fig. 3. From Eq. (38) remembering the development in power series of e^x , we also see immediately that

$$\sum_{n=0}^{\infty} P(n) = e^{-\bar{n}} \sum_{n=0}^{\infty} \frac{\bar{n}^n}{n!} = 1 \quad (39)$$

which verifies that the sum of the probabilities for all possible numbers of disintegrations in a given time interval is 1

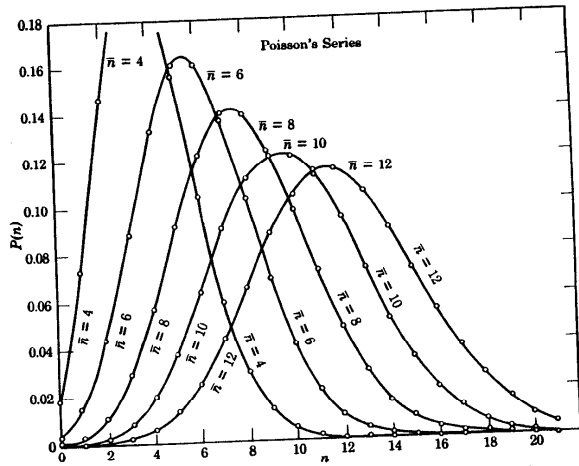


Fig. 3. Poisson's formula: $P(n) = (\bar{n}^n/n!)e^{-\bar{n}}$ for different values of \bar{n} .

If \bar{n} is a large number, $P(n)$ has a sharp maximum in the vicinity of $n = \bar{n}$, and we may develop $\log P(n)$ in a power series of $n - \bar{n}$. Using Stirling's asymptotic formula, $\log(x!) = x \log x - x + \frac{1}{2} \log 2\pi x$, we obtain for Eq. (38)

$$\log P(n) = n \log \bar{n} - \bar{n} - n \log n + n - \frac{1}{2} \log 2\pi n \quad (40)$$

and, taking the first and second derivatives with respect to n ,

$$\frac{d \log P}{dn} = \log \bar{n} - \log n - \frac{1}{2n} \quad (41)$$

$$\frac{d^2 \log P}{dn^2} = -\frac{1}{n} + \frac{1}{2n^2} \quad (42)$$

The zero of the first derivative confirms the existence of the maximum¹ near $n = \bar{n}$ (we neglect terms in $1/n$ with respect to $\log n$, and terms in $1/n^2$ with respect to terms in $1/n$); the second derivative can

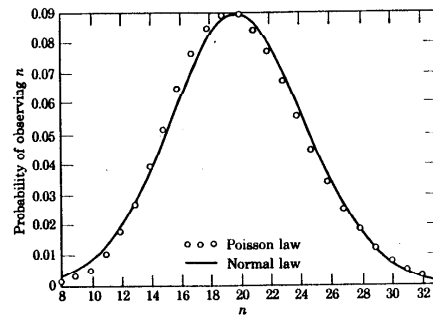


Fig. 4. Comparison of Poisson's law for $\bar{n} = 20$ with Gauss's law.

be used to write the first terms of the power series for $\log P(n)$:

$$\begin{aligned} \log P(n) &= \log P(\bar{n}) + \frac{(n - \bar{n})^2}{2!} \left[\frac{d^2}{dn^2} \log P(n) \right]_{n=\bar{n}} \\ &= -\frac{1}{2} \log 2\pi \bar{n} - \frac{(n - \bar{n})^2}{2\bar{n}} \end{aligned}$$

from which, passing from logarithms to numbers, we immediately get the famous Gauss formula:

$$P(n) = \frac{e^{-(n-\bar{n})^2/2\bar{n}}}{(2\pi\bar{n})^{1/2}} \quad (43)$$

(Examples of Poisson and Gauss distributions are shown in Figs. 3 and 4.) Remembering that

$$\int_{-\infty}^{\infty} e^{-x^2} dx = \sqrt{\pi}$$

we readily see that

$$\int_0^{\infty} P(n) dn = 1 \quad (44)$$

which expresses again the normalization of the probability. It must

¹ In a better approximation the maximum is near $n = \bar{n} - \frac{1}{4}$.

be remembered that $\bar{n} \gg 1$, and hence the contributions to the integral for $n < 0$, are negligible.

From Eq. (37), (38), or (43) it is possible to compute the root mean square (rms) deviation or standard deviation defined by Eq. (35). From Eq. (37), if K tends to infinity, p tends to 0 and q to 1; Eqs. (37) and (34) give directly

$$\sigma^2 - Kp = \bar{n} \quad (45)$$

The same result is obtained if we insert for $P(n)$ the Poisson formula and use the identity

$$\sum_{n=0}^{\infty} n^2 \frac{\bar{n}^n}{n!} = \bar{n}(\bar{n} + 1)e^{\bar{n}}$$

which can be easily proved by comparing coefficients of equal powers of \bar{n} . Finally, the same result comes from the definition of σ^2 contained in Eqs. (35) and (36) and the Gauss distribution (43) if we remember that

$$\int_{-\infty}^{\infty} x^2 e^{-x^2} dx = \frac{1}{2} \pi^{1/2}$$

These expressions say that the average value of the square of the deviation from the average number of disintegrations to be expected in a certain time interval is equal to this last number, and hence

$$\sigma = \bar{n}^{1/2} \quad (46)$$

σ is known as the standard deviation of the distribution in n . Thus, if we have on the average 100 disintegrations per minute, we may expect deviations from this average, in every single minute, such that their mean square is again 100. Very often it is of great interest to know the relative standard deviation which results from Eq. (46):

$$\frac{\sigma}{\bar{n}} = \frac{1}{\bar{n}^{1/2}} \quad (47)$$

The relative standard deviation is inversely proportional to the square root of the number of counts used and is a measure of the precision of the determination.

Another approach to an estimate of σ can be obtained by starting from its definition, Eq. (35), in which, however, we have not distin-

guished \bar{n} from the "true" value of \bar{n} , which would be obtained, for instance, with an infinite number of observations. Let us indicate the "true" value of \bar{n} by \bar{n}_t . In a sample giving n_i counts in each N equally long time intervals, we should actually form

$$\frac{1}{N} \sum_{i=1}^N (n_i - \bar{n}_t)^2 \quad (48)$$

Since this value \bar{n}_t is actually unobtainable, we replace it with

$$\bar{n} = \frac{1}{N} \sum_{i=1}^N n_i$$

and we assume, for the square of the standard deviation of the single measurement,

$$\sigma^2(n) = \frac{1}{N-1} \sum_{i=1}^N (n_i - \bar{n})^2 \quad (49)$$

and, for the square of the standard deviation of the average,

$$\sigma^2(\bar{n}) = \frac{1}{N(N-1)} \sum_{i=1}^N (n_i - \bar{n})^2 \quad (50)$$

The replacement of N in Eq. (48) by $N-1$ in Eq. (49) compensates the fact that \bar{n}_t , the true value, has been replaced by \bar{n} . For large N , Eqs. (49) and (48) tend to coincide, and it is not practically important to distinguish between them because, as a matter of fact, the difference between them is of the order of magnitude of the error in σ^2 . Justification of Eq. (49) and of the last statement will be found in standard books on probability and statistics.

Of course, the values of σ^2 thus obtained are themselves only approximate, and we may have examples in which σ^2 as calculated by Eq. (49) comes out anomalously small. In fact, σ cannot be lower, for our case, than $\sqrt{\bar{n}}$, and, if our series of measures has given a value lower than this, we still must assume that the standard deviation is $\sqrt{\bar{n}}$ and that we have obtained the lower value by accident. On the other hand, if we have found a standard deviation much larger than $\sqrt{\bar{n}}$, we may suspect the existence of some other cause of accidental errors.

We shall now illustrate these concepts with an actual numerical example. A uranium sample was counted, and the number of alpha-particles emitted in a minute was recorded in the next 10 intervals each

of 1-minute duration:

n	$n - \bar{n}$	$(n - \bar{n})^2$
36,076	130	16,900
35,753	-193	37,249
35,907	30	1,521
36,116	170	28,900
35,884	-62	3,844
36,136	190	36,100
35,741	-205	42,025
35,640	-306	93,636
36,124	178	31,684
36,087	141	19,881
$\bar{n} = 35,946.4$	$\Sigma 4$	$\Sigma 311,740$

$$\sigma(n) = \sqrt{\frac{311,740}{9}} = 186$$

In the first column we report the actual number of counts observed, in the second column the difference between the average number of counts and the specific 1-minute count; in the third column we have the square of the numbers written in the second column. The average count for a 1-minute period is 35,946.4 with the standard deviation of any single measurement given by $(311,740/9)^{1/2} = 186$. If, according to Eq. (46), we put $\sigma = \sqrt{\bar{n}}$, we find $\sigma = 189$, a very satisfactory agreement. The average counting rate is 35,946.4 counts per minute, and the standard deviation of the average is given by $189/\sqrt{10} = 60$, where 10 is the number of observations and the standard deviation of one observation is 189. In conclusion, the average counting rate of our sample is $35,946 \pm 60$ counts per minute.

The question may be asked, "What is the probability that, having measured a counting rate A with a standard deviation σ , the deviation from the 'exact' result A^* will be larger than $\epsilon\sigma$?" The "exact" result is defined, as above, as the one obtainable by repeating the measurement of A a very large number of times. The answer to this problem in the case of a Gaussian distribution is given in regular tables (111). We quote here a few pertinent numbers.

The probability P of observing a counting rate differing from A^* by more than $\epsilon\sigma$ is

$\epsilon = 0$	0.6745	1	1.5	2.0	2.5	3	3.5	4
$P = 1$	0.5000	0.3173	0.1336	0.0455	0.0124	0.0027	0.00046	0.000063

It will be noted that there is the probability of 0.5 of observing a count-

ing rate differing from A^* by more than 0.6745σ . This quantity is called the probable error.

Connected with this type of question is a test called the χ^2 test, or test for the goodness of fit, which is sometimes used to ascertain whether the fluctuations observed in a finite series of observations are consistent with Poisson's law.

Suppose that a series of values n_i have been obtained for the numbers of counts, respectively, in N equal time intervals. The expression

$$\chi^2 = \frac{1}{\bar{n}} \sum_{i=1}^N (n_i - \bar{n})^2 \quad (51)$$

is formed. This is a function of the n_i ; in the case of a Poisson distribution for the n_i , χ^2 will also have a distribution. Table 5 gives the probability $P(\chi^2)$ that, if the N numbers n_i are the representatives of a Poisson distribution, χ^2 will exceed a certain number. The "number of degrees of freedom" mentioned in the first column is, in our case, $N - 1$. In our example on page 30, χ^2 is 8.67, and thus there is a probability of about 0.51 that, if 10 numbers are taken out of a Poisson distribution, χ^2 will exceed 8.67. As a working rule, values of χ^2 indicating a probability corresponding to $P(\chi^2) \geq 0.1$ in the table might be accepted, because, if $P(\chi^2) \geq 0.1$, we may say that there is no good reason to reject the hypothesis that the observed n_i numbers conform to a Poisson distribution.

If we want to know the standard deviation of a function f of two or more independently observed quantities x_1, x_2 affected by the standard deviation σ_1, σ_2 , we use the fundamental formula of the propagation of errors:

$$\sigma_f = \left[\left(\frac{\partial f}{\partial x_1} \right)^2 \sigma_1^2 + \left(\frac{\partial f}{\partial x_2} \right)^2 \sigma_2^2 \right]^{1/2} \quad (52)$$

Of frequent application are the formulas for the standard deviations of the sum, difference, product, and quotient of two quantities. If the observed quantities are numbers of counts, Eqs. (46) and (52) give

$$\sigma(n_1 \pm n_2) = (n_1 + n_2)^{1/2} \quad (53)$$

$$\sigma(n_1 n_2) = [n_1 n_2 (n_1 + n_2)]^{1/2} \quad (54)$$

$$\sigma \left(\frac{n_1}{n_2} \right) = \left(\frac{n_1}{n_2^2} + \frac{n_1^2}{n_2^3} \right)^{1/2} \quad (55)$$

and

$$\sigma \left(\frac{n_1 n_2}{n_1 n_2} \right) = \frac{\sigma(n_1/n_2)}{n_1/n_2} = \left(\frac{1}{n_1} + \frac{1}{n_2} \right)^{1/2} \quad (56)$$

TABLE 5
TABLE OF χ^2

Degrees of freedom	P = 0.99	0.98	0.95	0.90	0.80	0.70	0.50	0.30	0.20	0.10	0.05	0.02	0.01
1	0.000157	0.00028	0.00393	0.0138	0.0442	0.148	0.455	1.074	1.642	2.706	3.841	5.412	6.635
2	0.0201	0.0404	0.102	0.211	0.446	0.713	1.386	2.776	4.605	7.378	10.591	15.013	18.475
3	0.0159	0.0318	0.0771	0.161	0.315	0.509	0.935	1.928	3.219	5.024	7.378	10.591	13.801
4	0.0135	0.0270	0.0635	0.138	0.270	0.435	0.784	1.533	2.706	4.541	6.757	9.488	12.838
5	0.0125	0.0250	0.0600	0.132	0.260	0.420	0.764	1.514	2.675	4.502	6.708	9.488	12.838
6	0.0120	0.0243	0.0590	0.130	0.257	0.417	0.757	1.506	2.662	4.483	6.678	9.454	12.802
7	0.0118	0.0239	0.0585	0.129	0.255	0.415	0.753	1.501	2.655	4.473	6.665	9.448	12.797
8	0.0117	0.0237	0.0583	0.128	0.254	0.414	0.751	1.499	2.651	4.468	6.659	9.444	12.794
9	0.0116	0.0236	0.0582	0.128	0.253	0.413	0.750	1.498	2.649	4.465	6.656	9.442	12.792
10	0.0116	0.0235	0.0581	0.127	0.252	0.412	0.749	1.497	2.647	4.463	6.654	9.441	12.791
11	0.0115	0.0234	0.0580	0.127	0.251	0.411	0.748	1.496	2.645	4.461	6.652	9.440	12.790
12	0.0115	0.0233	0.0579	0.126	0.250	0.410	0.747	1.495	2.643	4.459	6.650	9.439	12.789
13	0.0114	0.0232	0.0578	0.126	0.249	0.409	0.746	1.494	2.641	4.457	6.648	9.438	12.788
14	0.0114	0.0231	0.0577	0.125	0.248	0.408	0.745	1.493	2.639	4.455	6.646	9.437	12.787
15	0.0113	0.0230	0.0576	0.125	0.247	0.407	0.744	1.492	2.637	4.453	6.644	9.436	12.786
16	0.0113	0.0229	0.0575	0.124	0.246	0.406	0.743	1.491	2.635	4.451	6.642	9.435	12.785
17	0.0112	0.0228	0.0574	0.124	0.245	0.405	0.742	1.490	2.633	4.449	6.640	9.434	12.784
18	0.0112	0.0227	0.0573	0.123	0.244	0.404	0.741	1.489	2.631	4.447	6.638	9.433	12.783
19	0.0111	0.0226	0.0572	0.123	0.243	0.403	0.740	1.488	2.629	4.445	6.636	9.432	12.782
20	0.0111	0.0225	0.0571	0.122	0.242	0.402	0.739	1.487	2.627	4.443	6.634	9.431	12.781
21	0.0110	0.0224	0.0570	0.122	0.241	0.401	0.738	1.486	2.625	4.441	6.632	9.430	12.780
22	0.0110	0.0223	0.0569	0.121	0.240	0.400	0.737	1.485	2.623	4.439	6.630	9.429	12.779
23	0.0109	0.0222	0.0568	0.121	0.239	0.399	0.736	1.484	2.621	4.437	6.628	9.428	12.778
24	0.0109	0.0221	0.0567	0.120	0.238	0.398	0.735	1.483	2.619	4.435	6.626	9.427	12.777
25	0.0108	0.0220	0.0566	0.120	0.237	0.397	0.734	1.482	2.617	4.433	6.624	9.426	12.776
26	0.0108	0.0219	0.0565	0.119	0.236	0.396	0.733	1.481	2.615	4.431	6.622	9.425	12.775
27	0.0107	0.0218	0.0564	0.119	0.235	0.395	0.732	1.480	2.613	4.429	6.620	9.424	12.774
28	0.0107	0.0217	0.0563	0.118	0.234	0.394	0.731	1.479	2.611	4.427	6.618	9.423	12.773
29	0.0106	0.0216	0.0562	0.118	0.233	0.393	0.730	1.478	2.609	4.425	6.616	9.422	12.772
30	0.0106	0.0215	0.0561	0.117	0.232	0.392	0.729	1.477	2.607	4.423	6.614	9.421	12.771

Sec. 3B) Fluctuations in Radioactive Decay

where n_1 and n_2 are the observed numbers of counts. Expressions of the type of Eq. (55) are most rapidly obtained by use of the relation

$$\sigma(\log f) = \frac{\sigma(f)}{f} \tag{57}$$

which is an immediate consequence of Eq. (52).

B. Applications

We shall now make some applications of the principles stated above, either in order to illustrate them or because the results are of practical importance.

1. Suppose that we have counted a sample which gives n_1 counts in a certain counter in time t_1 ; the background of the counter gave n_2 counts in time t_2 . We want to know the counting rate of the sample and the background counting rate of the counter. The times t_1, t_2 are known exactly. For the background we have

$$\nu_2 = \frac{n_2}{t_2} \pm \frac{n_2^{1/2}}{t_2} \tag{58}$$

(The expressions following the \pm sign are always standard deviations.)

We have, for the activity plus the background,

$$\nu_1 = \frac{n_1}{t_1} \pm \frac{n_1^{1/2}}{t_1}$$

for the net activity,

$$\nu = \nu_1 - \nu_2 \tag{59}$$

and, for its standard deviation according to Eq. (49),

$$\sigma(\nu) = [\sigma^2(\nu_1) + \sigma^2(\nu_2)]^{1/2} = \left(\frac{n_1}{t_1^2} + \frac{n_2}{t_2^2} \right)^{1/2} = \left(\frac{\nu_1}{t_1} + \frac{\nu_2}{t_2} \right)^{1/2} \tag{60}$$

Now we may ask what is the best way to apportion a fixed counting time $T = t_1 + t_2$ between counting of the sample and counting of the background. From Eq. (60) we obtain

$$\sigma^2(\nu) = \frac{\nu_1}{t_1} + \frac{\nu_2}{T - t_1}$$

and, minimizing with respect to t_1 ,

$$\frac{\nu_1}{t_1^2} = \frac{\nu_2}{(T - t_1)^2} \tag{61}$$

2. We want to check the existence of an effect which slightly changes the counting rate of a device. When can we reasonably conclude that the effect is real? We might require that the effect be, for example, 3 times greater than the standard deviation of the measurement.

Counts in conditions *a*, n_1 ; counts in condition *b*, n_2 ; counting time the same for both conditions and equal to t . We have then

$$\nu_1 - \nu_2 = \frac{n_1 - n_2}{t}$$

$$\sigma(\nu_1 - \nu_2) = \frac{(n_1 + n_2)^{1/2}}{t} \quad (62)$$

and we shall want to have $\nu_1 - \nu_2 > 3\sigma(\nu_1 - \nu_2)$ or

$$n_1 - n_2 > 3(n_1 + n_2)^{1/2} \quad (63)$$

3. A certain sample gives ν counts per second on the average. What is the probability of finding an interval of t seconds without counts? Formula (39) gives the answer if we put in it $n = 0$ and $\bar{n} = \nu t$:

$$P(0) = e^{-\nu t} \quad (64)$$

4. **Counting Loss Problem.** Assume that a counting device is unable to register a pulse for a time τ after having registered a previous pulse (dead time). If we try to measure a sample giving rise to an average true counting rate ν_0 , the device will show a counting rate $\nu < \nu_0$ because some of the pulses will occur during the dead time τ following each registered pulse and hence will not be registered. We want the relation between ν and ν_0 . The total dead time per second is $\nu\tau$; the counts that should occur during it but cannot be registered are $\nu_0\nu\tau$, and from this

$$\nu = \nu_0 - \nu_0\nu\tau$$

or, assuming $\nu_0 - \nu \ll \nu$,

$$\nu_0 = \nu(1 + \nu\tau) \quad (65)$$

The hypotheses under which (65) is derived are not always realized in practice, and very elaborate discussions on the subject exist in the literature (J4) for more general cases. On the other hand, the practical procedure for correcting for counting losses is to count separately two or more samples and then count them together. The count obtained from the sum of the samples is smaller than the sum of the counts from the single samples because of the counting losses, and, if Eq. (65) applies, the counting loss $\nu_0 - \nu$ is proportional to ν . This will always be true if $\nu_0 - \nu$ is small compared to ν and the propor-

tionality constant is determined empirically as stated above. It is not safe to correct for counting losses if these amount to more than about 0.2 of the counting rate, it is better to subdivide the samples, reduce the solid angle of the detecting device, or otherwise arrange to use lower counting rates. If we go to the other limiting case of a pulse rate $\nu_0 \gg 1/\tau$, the apparatus is blocked and the counting rate decreases with increasing ν_0 or it counts at a counting rate $1/\tau$ according to its detailed

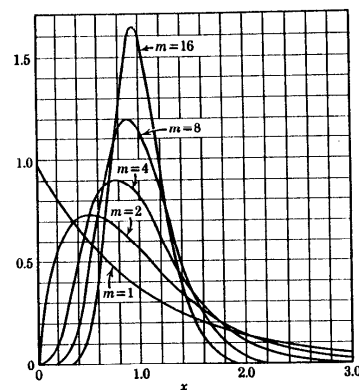


Fig. 5. The regularizing action of a scale-of- m circuit with a random input. The curves show the differential probability that the timing spacing between two successive output counts will lie between x and $x + dx$ times the average spacing (R2).

construction. Under these conditions the apparatus is obviously unsuitable for making measurements. For a detailed discussion of the practical methods of determining counting losses see (R2) and (B6).

5. **Scaling Circuit Regularizing Action.** In most pulse-detecting instruments, scaling circuits interposed between the output of the amplifiers and a mechanical counting device have the effect that the mechanical register is actuated only once for every m pulses. Almost always m is a power of 2, and we speak of scales of 2, 4, 8, 16, etc. Now a scale of m has two effects: it reduces the average rate of the mechanical register to ν_0/m , and it makes the spacing between pulses entering the mechanical register more regular than that corresponding to a random distribution of frequency ν_0 . The last effect in turn allows the mechanical register to register without appreciable loss a counting rate much

higher than it could if the pulses came at random. Using Poisson's or Gauss' distribution, we find that the standard deviation of the spacings of the pulses from their mean, which is equal to their mean spacing if no scaling circuit is used, is reduced to $1/\sqrt{m}$ of the mean spacing if a scale of m is used. Figure 5 represents graphically the regularizing action of a scaling circuit.

6. Fluctuations in Ionization Current. Let us consider an ionization chamber with a sample emitting alpha-particles each of which gives a potential v to the collecting electrode. On the average ν particles per second are emitted, and we take a reading of the potential of the electrode every T seconds. We compensate the average charging effect of our source by a leak resistor or by balancing it with another ionization chamber in opposition and an identical alpha-source. The average potential reading \bar{p} is then zero, but there are residual fluctuations of potential. We can calculate $\overline{p^2}$ from Eq. (45). Calling n the actual number of particles emitted in time T , and νT its average value, we have from Eq. (35)

$$\overline{(p - \bar{p})^2} = \overline{(n - \nu T)^2 v^2} = \nu^2 \nu T$$

or, since $\bar{p} = 0$ owing to the compensating device,

$$\overline{p^2} = \nu^2 \nu T \quad (66)$$

if the main effect is compensated with a resistor, or

$$\overline{p^2} = 2\nu^2 \nu T \quad (67)$$

if we have two balanced chambers. In this equation $\overline{p^2}$ and T are directly measurable, and an experiment of this type can be used to determine the quantity νv . For precision measurements it is well to check whether relation (66) or (67) is obeyed by the experimental data. If either relation obtains, the statistical accuracy is as good as it can be with the given experimental arrangement.

Equations (66) and (67) are mainly qualitative because they do not consider the differences between the amounts of ionization produced in the various ionization acts and the electrical characteristics of the measuring instrument.

The fluctuations of p have been the object of several studies (B8) in which the influence of the electrical characteristics of the detecting apparatus have also been considered. Although most of these studies were performed with long-period electrometers and experimental devices which are now obsolete, the method of evaluation of the results is quite

general. If a particle emitted at time zero produces in the instrument a deflection at time t indicated by $f(t)$, where the specific function depends on the characteristics of the apparatus, and if the apparatus is linear, the deflection at time T produced by k particles emitted at times t_1, t_2, t_3, \dots is given by

$$\phi = \sum_k f(T - t_k) \quad (68)$$

If we now think of performing many experiments and read for each at time T the deflection of our apparatus, we will obtain an average deflection $\bar{\phi}$. In order to calculate this quantity and its standard deviation, call ν the average rate of emission of the particles. In a small time interval τ the probability of emission of a particle is $\nu\tau$.

The contribution $\delta\bar{\phi}$ to the average deflection due to particles emitted in time τ is

$$\delta\bar{\phi} = f(T - t)\nu\tau \quad (69)$$

and, passing to the limit,

$$\bar{\phi} = \Sigma\delta\bar{\phi} = \int_0^T \nu f(T - t) dt \quad (70)$$

The standard deviation of $\bar{\phi}$ is obtained by using the theorem of the calculus of probability contained in Eq. (37), which states that the square of the standard deviation of the number $\nu\tau$ is $\nu\tau$ times the complementary probability $1 - \nu\tau$. Moreover, since the number of particles emitted in the single time intervals τ are statistically independent, the contributions to the square of standard deviation from the various time intervals are to be added. We have for the standard deviation of the deflection

$$(\phi - \bar{\phi})^2 = \Sigma\nu\tau(1 - \nu\tau)f^2(T - t)$$

or, passing to the limit for $\tau \rightarrow 0$,

$$\overline{(\phi - \bar{\phi})^2} = \int_0^T \nu f^2(T - t) dt \quad (71)$$

Equation (71) can be applied to specific cases, e.g., if $f(t) = 0$ for $t < 0$ and $f(t) = \nu e^{-\gamma t}$ for $t > 0$, we have for $T \gg 1/\gamma$

$$\overline{(\phi - \bar{\phi})^2} = \frac{\nu^2 \nu}{2\gamma} \quad (72)$$

which is similar to Eq. (66). More details on this subject are to be

found in an article by Bothe (B8), where other forms of $f(t)$ are discussed.

In the case of the $f(t)$ given above it is interesting to evaluate the limitations of a "rate meter" due to statistical considerations. Such an instrument requires a certain time to reach a reading corresponding to the average intensity; moreover, it will fluctuate around this average. Table 6, taken from Rainwater and Wu (R2), gives a quantitative measure of these effects.

TABLE 6
BASIC LIMITATIONS DUE ONLY TO STATISTICAL CONSIDERATIONS (R2)

e% (Standard deviation)	N = νt	t if $\nu = 1.5/\text{sec}$	Time to reach x% of equilibrium			
			80%	95%	99%	99.9%
20	25	17 sec	27 sec	51 sec	78 sec	117 sec
10	100	67 sec	104 sec	200 sec	5.1 min	7.7 min
5	400	267 sec	430 sec	13.3 min	20.6 min	30.9 min
2	2,500	27.8 min	45 min	83 min	128 min	192 min
1	10,000	111 min	3 hr	5.6 hr	8.5 hr	12.8 hr
0.1	1,000,000	7.7 days	12.4 days	23 days	35.4 days	53 days

The table is calculated for $\nu = 1.5 \text{ sec}^{-1}$. In the first column the desired standard deviation is given, in the second the number of counts necessary to attain the standard deviation is indicated, and in the third column the time necessary to accumulate the indicated number of counts if ν is 1.5 sec^{-1} . For an instrument in which each event gives the same deviation and the deviations cumulate (drift instrument, $\gamma = 0$) this is the time required in order to obtain the standard deviation indicated in the first column. For a steady deflection measurement it indicates the value of $1/\gamma$ required to limit the standard deviation to the value indicated in the first column. The remaining columns give the time necessary to reach a certain fraction of $\bar{\phi}$ if $1/\gamma$ has the value indicated in the third column.

It is also clear that, for a given time of measuring, greater statistical accuracy will always be obtained by a counting or drift method than by a rate meter.

Equation (72) is often applied in the determination of the conditions under which a heavily ionizing radiation, e.g., fission fragments, can be measured over a background of an intense radiation the single particles

of which ionize only slightly (beta-rays). If the resolving time of the instrument including the electronic part is τ and the number of ions obtainable from the heavily ionizing particle is F , whereas each of the weakly ionizing particles, occurring with the frequency ν , produces f ions, we must have

$$F \gg f(\nu\tau)^{1/2} \quad (73)$$

This relation, however, can be used only for qualitative estimates because a detailed study of the experimental set-up is required for more precise evaluations. For a more refined study of the statistical aspects of this problem see reference (S4).

7. Problem Involving Variable Counting Rate. In the problems treated above we have considered counting rates whose average value is constant in time. We shall consider now a problem where this condition does not obtain, i.e., the determination of the half-life of a radioactive substance decaying exponentially with time (P2).

The average counting rate is then given by $\nu(0)e^{-\lambda t}$, and we want to arrange the experiment in the most favorable way in order to determine λ . Similar considerations apply in the determination of the absorption coefficient of a radiation (if the absorption is exponential) and have been developed in detail for this case also (R3, R4).

Neglecting the background at first, we could count the particles emitted during fixed time intervals short with respect to τ and plot them on semi-logarithmic paper versus time. A straight line results, and its slope gives λ . This is the most common procedure, but it is not the most suitable for high accuracy.

Theoretically we obtain the optimum accuracy if we take the average time at which the particles have been counted starting from an arbitrary zero time and continuing until the sample is completely decayed. This average time is by definition τ , and its mean relative error is $\sqrt{1/N}$ if N particles have been observed. This result has mainly theoretical significance. A slightly more realistic procedure is to compare the number n_1 of counts in a suitably chosen time interval t with the total number of counts N obtainable in an infinite time. The ratio between the number $n_2 = N - n_1$ and N is on the average

$$r = n_2/N = e^{-\lambda t} \quad (74)$$

The most favorable length of the interval t can be chosen by the following considerations. From Eq. (74) we have approximately

$$\lambda = -\frac{1}{t} \log r$$