S 79. *Techniques in Radiochemical Assay with Geiger–Müller Counters.*

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Some of the errors possible in measuring β-active material with Geiger–Müller counters are discussed. Emphasis has been laid on aspects not fully considered elsewhere, such as errors due to bad sample mounting; errors in liquid-immersion counting and in flow counters are also discussed.

THE general operation and use of Geiger–Müller counters has been given elsewhere (*e.g.*, Maddock, *Analyst*, 1948, **73**, 644; Kohman, *Analyt. Chem.*, 1949, **21**, 352), and in the present paper familiarity with a Geiger assembly will be assumed. Many workers concerned with the use of active material as tracers will be interested mainly in comparative measurements. In such cases the greater number of the errors mentioned below are small provided that measurements are made under *strictly comparable conditions*, namely, when such factors as the counter characteristics, the geometry of the system, the source mounting, the distribution of the source on the mounting, the weight of the source, and the proximity of other scattering material are maintained constant. The magnitude of the errors from some of these causes will be considered.

(1) *Errors Common to All Geiger Counters.*—Even if all associated electronic apparatus is in working order, errors may arise from causes inherent in the method.

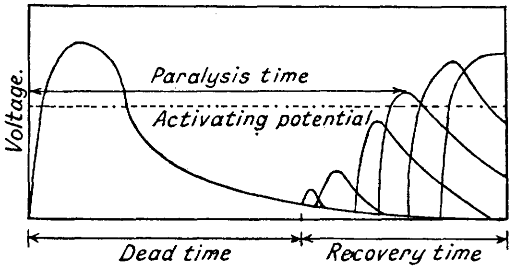
(i) *Statistical considerations.* Radioactive disintegrations are purely random in occurrence, and follow a Poisson distribution common to all random rare events (Udny, Yule and Kendall, " An Introduction to the Theory of Statistics," Griffin, 1947). One of the properties of a Poisson distribution is that the standard deviation (σ) is equal to the square-root of the arithmetic mean of the number of events. In radiochemical work it is customary to estimate the error in a given observation by taking the square-root of the total number of counts obtained. Thus the accuracy of an observation of, say, 2000 counts is usually written as 2000 ± 44·7. It is important to appreciate that the standard deviation (44·7) obtained in this way is not an error. By statistical theory it is that value of the deviation inside which there is about a 2 to 1 (68·27%) chance of the true result being found. There is also a 19 to 1 (95·45%) chance of the true result being within 2σ of the observed value. Thus, in the example given, there is about a 2 to 1 chance of the result being between 2044·7 and 1953·3 (*i.e.*, within 2·23%) and a 19 to 1 chance of the true result being between 2089 and 1911 (*i.e.*, within 4·47%). In order to maintain a reasonable (19 to 1) chance of obtaining a result within ±5% of the mean value it is necessary to make at least 2000 counts. Increasing the number of counts will correspondingly increase the accuracy of the measurement, and it is therefore important either to use a high counting rate or to count for long periods. In many counting experiments it is necessary to subtract the natural background (n) from the total number of counts (N). The standard deviation of the result is then $\sigma = (N + n)^{1/2}$. For example :

	Count in time, t.	Standard deviation, σ.	100σ/count.
(*a*) Source and background	225	15	6·65
(*b*) Background	64	8	12·50
(*c*) Activity of source (by diff.)	161	17	10·55
(*d*) Sum of (*a*) and (*b*)	289		

The standard deviation of the activity (161 counts) is 10·55% compared with 6·65% obtained for the standard deviation of the measurement of source and background together. From this it is clear that accuracy is increased by (*a*) reducing the uncertainty in the background by counting over a prolonged period and (*b*) reducing the background counting rate as far as possible (*e.g.*, by avoiding contamination, stray radiation, etc., and by keeping the counter in a lead castle to cut out almost all stray radiation except cosmic rays).

(ii) *Paralysis time.* The shape of the pulse from the anode of a counter is as shown in Fig. 1. After the sharp initial rise in potential (of the order of 1 volt) there is a more gradual fall as the positive ions liberated in the discharge diffuse towards the cathode (much more slowly than the electrons travel to the anode); simultaneously the potential of the central wire returns to its initial value. When the positive ions have migrated sufficiently from the anode for the field to be more or less restored to its initial state, a second discharge may occur even though the first pulse has not completely decayed. The time necessary for full recovery of the counter is called the recovery time. If a second pulse is obtained, within the recovery time, it will not be of full pulse height (as has been shown by Stever, *Physical Rev.*, 1942, **61**, 40), and will be counted only if the pulse potential is sufficiently high to operate the auxiliary electronic equipment. The time required for this, called the paralysis time of the circuit, determines the rate at which counts can be observed without appreciable loss. The paralysis time may be reduced by adjustment of the sensitivity of the associated scaling unit and preamplifier or by reducing the dead time of the counter (the time after commencement of a pulse during which an ionising event is undetected by the counter). The dead time may be kept small by preventing the formation of secondary electrons either by external resistance quenching or by introducing into the counter gas an organic molecule such as alcohol (see Korff, " Electrical and Nuclear Counters," Van Nostrand, 1946). The paralysis time of a counting assembly, usually of the order of a few hundred microseconds, sets a practical limit to the maximum counting rate possible without the necessity of very large corrections.

FIG. 1.*



Shape of pulse from Geiger counter.

Several authors have suggested corrections for losses at high counting rates, occasioned by two pulses arriving within the paralysis time. If the paralysis time is the same for every pulse recorded, the counting rate n_0 is related to the observed counting rate n_1 by the relation

$$n_1 = n_0/(1 + n_0\tau) \qquad (1)$$

where τ is the paralysis time. Equation (1) holds for Geiger counters operated with a fixed paralysis time preset electronically. For counters which can be re-excited during their period of recovery by unrecorded ionising events, the equation

$$n_1 = n_0 e^{-n_0\tau} \qquad (2)$$

has been shown to hold. Other equations have been given for operation under different conditions, but in general, for slow counting rates, these equations degenerate to equation (1), which may be used up to about 5000 counts per minute with fair accuracy (see Lifschutz and Duffendack, *Physical Rev.*, 1938, **54**, 714; Schiff, *ibid.*, 1936, **50**, 88; Kurbatov and Mann, *ibid.*, 1945, **68**, 40; Beers, *Rev. Sci. Instr.*, 1942, **13**, 72).

Unless the auxiliary electronic circuit is set to have a paralysis time greater than the sum of the recovery time and dead time of the counter, the paralysis time of the counting assembly will to some extent be dependent on the counting rate. Although in some arrangements the paralysis time of a circuit may be set to a known value electronically, it is better to determine it

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* This diagram is based upon Fig. 3 of Stever, *Physical Rev.* 1942, **61**, 40.

accurately by some suitable experimental method (see Kamen, " Radioactive Tracers in Biology," Academic Press Inc., 1947). The magnitude of the paralysis-time correction is small for counting rates below 2000/minute, but increases rapidly for faster counting rates; *e.g.*, for counting rates of 1000, 5000, and 10,000 and a preset paralysis time of 300 microseconds, the corrections are respectively about 0·5%, 2·7%, and 8%.

(iii) *Spurious counts.* A large number of factors may result in erratic behaviour such as an ionising event being recorded as more than one count. The associated electronic equipment may well produce erroneous results of such a nature, and in this connection reference should be made to a full discussion of the errors caused by faults in scaling units, power packs, etc. (see, *e.g.*, Lewis, " Electronic Counting," Cambridge, 1942; Korff, *op. cit.*).

The most likely cause of spurious counts from the counter itself is the decomposition of the organic vapour used for quenching the discharge. Most counters have a lifetime of the order of 10^8 counts, after which they begin first to give multiple counts (owing to incomplete quenching of the discharge), then a continuous discharge, and finally they cease to function altogether. The efficiency of a counter can often be restored by refilling with a mixture of alcohol (1 cm. pressure) and argon (9 cm. pressure). Application of a negative polarity to the central electrode instead of the normal positive polarity also destroys the alcohol filling and renders the counter useless. After application of the wrong polarity, recovery sometimes occurs if the counter is left idle for about a week, but it is usually necessary to refill it.

Some counters are photosensitive, electrons being emitted by the gas, or from the electrodes. If made of glass, the counter can be covered externally with a thin film of opaque wax. Sometimes this is not possible, and the counter must be kept in a light-proof lead castle, as, for instance, if it has a very thin mica window. Changes in counting rate are also obtained with small temperature variations. The cause is not thoroughly understood, although it has been suggested (Korff, Spatz, and Hilberry, *Rev. Sci. Instr.*, 1942, **13**, 127) that it is due to desorption of the alcohol-quenching agent from the walls of the counter. Counters and lead castles should, therefore, never be placed in direct sunlight or overheated parts of the laboratory. In order to overcome errors due to these effects it is customary to check the efficiency of the counter by measuring a standard source of constant activity every time measurements are made. The following sources form convenient standards : UX_2 (natural uranium), Ra-D and -E, and ^{60}Co.

Electrical disturbances, such as the use of Tesla coils in close proximity to the counter, result in bursts of counts, which may also be caused by bad electrical connections.

(2) *Errors Arising in the Measurement of* β*-Particles from Solids.*—Even in the simple case of a comparison of the β-activity of two solids containing the same radioactive isotope using an end-windowed Geiger counter in a conventional lead housing, a number of additional errors must be also considered. By maintaining the same arrangement for the sample and counter in all cases, errors due to such factors as scattering from the surroundings and absorption in the counter window are eliminated. However, even under these conditions, at least four variables must be considered, namely, (*a*) back-scattering, (*b*) size of sample, (*c*) reproducibility of positioning, and (*d*) self-absorption.

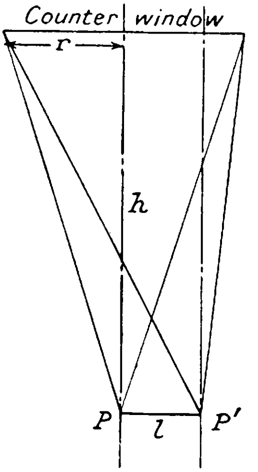
(a) *Back-scattering.* When a β-active sample is mounted on, say, a flat stainless-steel support and placed under a Geiger counter, those β-particles emitted in a direction away from the counter enter the support, where they are scattered and absorbed. Many are deflected through large angles, and some leave the support in the direction of the counter, with sufficient energy to reach it. Thus the recorded β-activity will be greater, owing to the back-scattering in this case, than if the backing were absent.

Yaffe *et al.* (this vol., p. S 341) have systematically investigated the conditions affecting back-scattering, and has shown that the amount of back-scatter depends on the thickness and atomic number of the mount, and on the energy of the β-particles. As the thickness of backing is increased from zero, the amount of back-scatter for a given maximum β-energy increases to a constant value at approximately $\frac{1}{5}$ of the range of the β-particles in the mount. The minimum thickness for constant back-scattering is to a first approximation independent of the atomic number of the support, but the percentage increase in recorded β-activity, from the value for zero weight of support, does depend on the atomic number.

To reduce errors from this cause in comparative measurements, it is best always to use the same type of support, preferably thick enough for saturation back-scatter; where this is not possible an experimentally determined correction factor must be applied. The back-scatter correction depends on the geometry of the system (Yankwich and Weigl, *Science*, 1948, **107**, 651).

(b) *Effect of source sizes.* In Fig. 2, if one considers a point source (*P*) of β-activity on the axis of a bell-type Geiger counter, then the angle subtended by the counter window at *P* is larger than that subtended at a point (P') displaced at a distance l from the axis, but at the same distance h from the counter window of radius r. Consequently, the observed β-count will be larger in the first case. Since, in practice, samples can rarely be regarded as point sources, the efficiency of measurement will vary with sample dimensions. An estimate of the magnitude of the effect may be obtained from Putman's work (*Brit. J. Radiol.*, 1947, **20**, 190), where figures are given of the activity observed from a point source at various distances from the counter window and for different displacements from the axis.

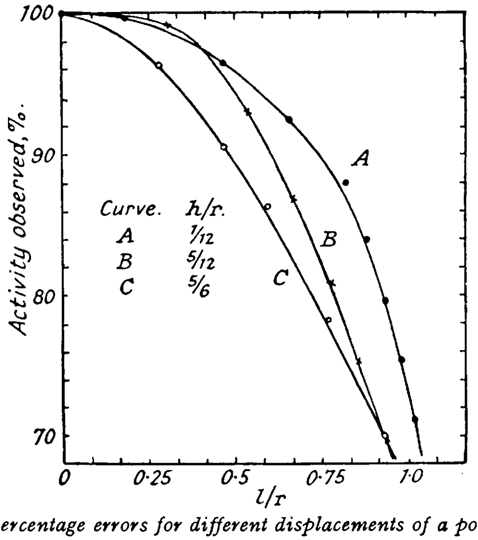
FIG. 2.



Angle subtended at a point source displaced from the axis of the counter.

Fig. 3, calculated from Putman's results, shows the activity observed from a source at a point P' expressed as a percentage of the count obtained when the source is at P, for different distances from the counter. These curves are a direct measure of the percentage error obtained when a source is mounted in such a way that it is deposited entirely round the rim of a circular mounting tray. In practice, this seldom happens, but it is very easy for active material to spread non-uniformly on the mount with a predominance round the edge, particularly if the mounting-tray has any kind of lip. It will be seen that the errors from this cause may be quite serious if a large tray is used. Putman's data are available for distances only up to 1 cm. from the window, but it is possible to estimate geometrically the error involved for other distances.

FIG. 3.



Observed percentage errors for different displacements of a point source.

To do this on a three-dimensional model involves an elliptical integral of the third degree. The magnitude of the variation to be expected, however, can be roughly estimated on a two-dimensional model.[*] Thus, in Fig. 2 we have :

Angle subtended by the window at the point P

$$= \tan^{-1}\frac{2h/r}{(h/r)^2 - 1} \qquad \ldots\ldots\ldots \quad (3)$$

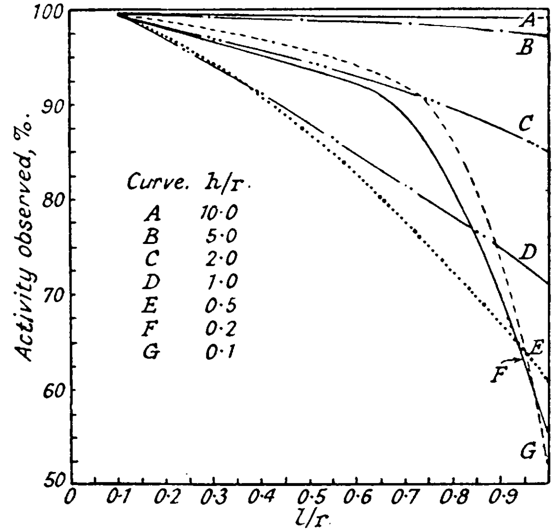
and the angle subtended by the window at the point P'

$$= \tan^{-1}(r - l)/h + \tan^{-1}(r + l)/h$$

$$= \tan^{-1}\frac{2h/r}{(h/r)^2 - 1 + (l/r)^2} \qquad \ldots\ldots\ldots \quad (4)$$

By inserting values of h/r and l/r into equations (3) and (4) the percentage difference in the angles subtended at P and P' may be determined. Fig. 4 shows the angle subtended at P' expressed as a percentage of that subtended at P and is plotted against l/r for different values of h/r. The form of the curves is very similar to that obtained experimentally (see Fig. 3) and it will be seen that for low values of l/r the error is smallest when the source is close to, or very far away from, the window. In practice it is best to keep the source diameter below half the diameter of the window (since errors get very large towards the extremities) and to present it as close as possible to the counter window. If it is not possible to place the source very near the window, then errors will be reduced by keeping it well away, although the source has to be considerably more active to maintain good statistical accuracy. In between these two extremes, there is a position of maximum error, which can be shown to obtain when the angle subtended by the window at the point P' is a right angle.

FIG. 4.



Calculated percentage errors for displacement of a point source.

The decreases in counting rate obtained when the activity is uniformly spread over a circular area are not so great as those obtained with sources concentrated at the edges. The errors have been estimated from Putman's experimental results by the following expression :

Observed activity of a uniform circular source of radius l, expressed as a percentage of the activity observed if all the source were concentrated on the axis of the counter

$$\left\{ \frac{\sum_{l=0}^{l=l} (\text{Percentage activity of point source at a distance } l) \times 2\pi l}{\sum_{l=0}^{l=l} \pi l^2} \right.$$

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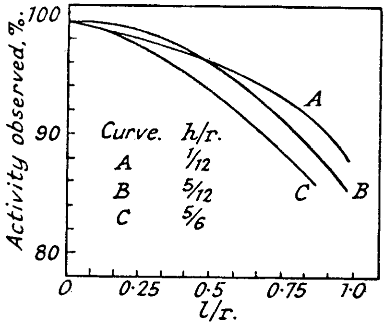
[*] *Added in proof.* The three-dimensional calculation has now been made. The results are similar, but the durations are larger.

Fig. 5 shows this quantity calculated for intervals in l of 0·5 mm. and different source diameters. It will be seen that when the source size is greater than three-quarters of the diameter of the counter window, reduced counting rates of the order of 10% or more can occur.

FIG. 5.



Percentage errors for uniformly distributed circular sources.

(c) *Reproducibility of positioning.* Reproducibility of positioning of a source is very important when working very close to the counter window. From Putman's results it can be shown that lowering a point source on the axis from 2 mm. to 2½ mm. from the window may cause a difference in count of 4% for a counter window about 22 mm. in diameter. Poor reproducibility of positioning may be reduced by good mechanical design of the supporting structure. To test the accuracy of positioning, the following experiment may be carried out. Place a sample of constant activity in position and take counts of one minute duration at least 30 times without moving the sample in any way, and confirm that the variations in observed activity are within the expected statistical limits. (If variations which cannot be accounted for statistically are observed, the counter assembly may not be in working order.) Now repeat the series, withdrawing and replacing the sample after each one-minute count and again ascertain whether the deviations are in accordance with statistical theory. If so, then the reproducibility of positioning is probably satisfactory.

(d) *Thick samples.* When the thickness of the sample itself is great enough to absorb an appreciable fraction of its own β-particles, a " self-absorption " correction must be applied. The determination of this factor has been described elsewhere (see, *e.g.*, Libby, *Analyt. Chem.*, 1947, **19**, 2; Yankwich, Norris, and Huston, *ibid.*, p. 439; Solomon, Gould, and Anfinsen, *Physical Rev.*, 1947, **72**, 1097).

In practical cases, self-absorption curves should always be determined under identical conditions with those under which samples are measured. Since the degree of back-scattering varies with the geometry of the counting arrangement, and is appreciably affected by changes in the average atomic number of the carrier and mount, extrapolation of self-absorption curves to zero weight (for example from a graph obtained by plotting the observed activity against the thickness of the source) should only be undertaken with the greatest caution. Back-scattering may produce a maximum in the apparent specific activity at a point about 5% of the range of the β-particles in the carrier, owing to non-attainment of saturation back-scatter. As the thickness of carrier is increased, the apparent specific activity rises owing to the increased counts caused by the increasing back-scatter (Glendenin, quoted by Solomon, Gould, and Anfinsen, *loc. cit.*).

Another correction for self-absorption introduced by Reid (" The Preparation and Measurement of Isotopic Tracers," Edward Bros., Ann Arbor, Mich., 1946) takes account of the oblique travel of some of the β-particles through the thickness of the sample, thus increasing the apparent absorbing path. However, Yankwich, Norris, and Huston (*loc. cit.*) were unable to support this experimentally, and it may be that oblique travel of β-particles through an absorber is to some extent included in an assumption of the experimentally observed exponential absorption in such cases.

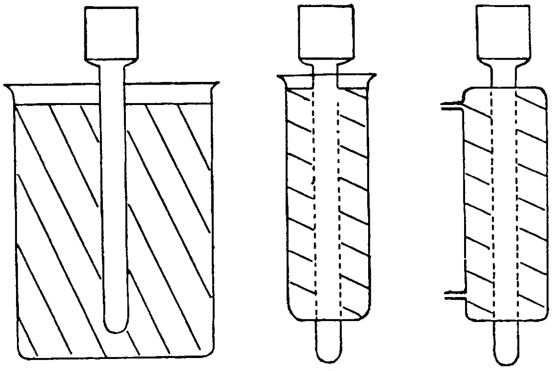
(e) *External absorption.* The experimental determination of the absorption curve of β-particles and the extrapolation to determine the effect of the window thickness is a common procedure. We will only comment on one factor that may affect results when working with very soft β-particles (*e.g.*, ^{14}C and ^{35}S). Assuming that a sample of ^{35}S is placed 1 cm. from the window of the counter, then a variation in atmospheric pressure of 730 to 770 mm., a temperature variation of 10° to 25° and a relative humidity varying from 30 to 80% could cause differences in count of about 4% owing to variations of β-absorption entirely caused by the change of density of the air. This is simply overcome by always placing sources of soft β-emitters as close as possible to the window. When this is not possible a standard source, whose β-energy and spectrum are as close to that of the sample as possible, should be used to check reproducibility of counting. It is good practice to do this as a routine with emitters of any energy, since scattering losses, etc., are also dependent on the energy of the β-particle.

(3) *Errors arising in the Measurement of β-Particles from Liquids.*—In estimating the radioactive content of liquids, absorption losses are very high. The contribution of most β-emitters below a depth of liquid of a few mm. is very small, and an infinitely thick layer may be assumed when the thickness equals the range of the β-particles. For the more penetrating γ-emitters, however, several metres of liquid may be necessary for this condition to obtain in practice.

Three methods of measuring the radioactive content of liquids will be considered and attention will be confined to aqueous solutions only; the conclusions are similar for organic solvents except that the depth of liquid required for a saturation counting rate depends on the density, but if the liquid depths are measured in mg./cm.^{2} the quantitative results are independent of the solvent to a first approximation.

(i) *Surface counting of liquids.* Probably the simplest means of " counting " liquids is to present the window of a bell-type Geiger counter to the surface. The problems of counting are then very similar to those obtaining for solids. Although it is desirable to use a thin layer of liquid, it is usually not possible to measure the depth accurately enough for allowance to be made for absorption losses, which are usually serious. Since the observed activity is greatest when the liquid can be regarded as infinitely thick, and in tracer experiments liquids are seldom of sufficient radioactive content to give a count greater than the maximum which the counter will accommodate, " infinitely-thick " layers are normally used. The depth necessary may be estimated from the range of the β-particle; for β-energies of less than 2·5 Mev., it is not necessary to use more than 10 mm. depth of liquid.

FIG. 6.


(a) (b) (c)

Three different techniques for measuring the radioactive content of liquids.

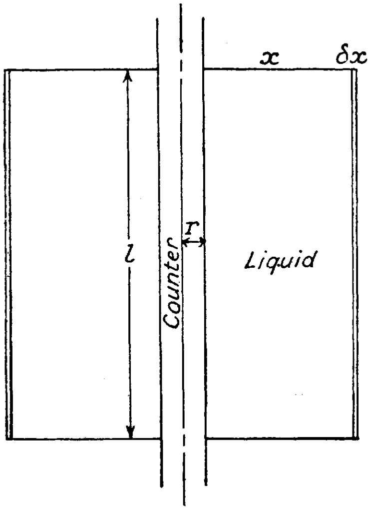
Errors due to scattering from the walls of vessels, etc., may be reduced by using containers of diameter several times greater than that of the counter window. This, however, is not always convenient, and if a smaller vessel is used it should be filled as completely as possible, to reduce scattering on the protruding walls. If the experimental arrangement is such that the source is not infinitely thick for the β-energy used, it is essential that the liquid should always be of the same depth and of the same distance from the counter window.

Losses due to evaporation of solvent, and adsorption of radio-isotopes on the walls of the vessel or on precipitates, may be serious, and care should be taken to ensure that the activity is homogeneously distributed throughout the liquid.

(ii) *Immersion counters.* Counters are available which can be immersed directly into a radioactive liquid, and three different ways in which this can be done are shown in Fig. 6. In methods (*b*) (Veall, *Brit. J. Radiol.*, 1948, **21**, 1347) and (*c*) the thickness of liquid round the counter is constant, but in (*a*) the counter may be placed either in a large boiling-tube of convenient size, or in a vessel large enough to enable counting to take place to infinite thickness.

Consider a thin concentric shell of liquid of thickness δx and length l at a distance x from a cylindrical counter of radius r (as shown in Fig. 7). Provided that x is more than two or three times the mean free path of a β-particle in the liquid, the absorption losses in the annular space may be approximately determined by applying the exponential absorption law. It is probable that this assumption is only approximate because of the cylindrical nature of the source and counter, and it is appreciated that a number of other factors should be introduced for a rigid treatment. Nevertheless, the expression derived below describes the behaviour of such counters with β-particles fairly reliably, although counter end effects, back-scattering, etc., are neglected.

FIG. 7.



Use of a liquid source.

Applying the exponential absorption law, the observed activity (δA) due to the concentric shell is given by

$$\delta A = \pi(x + r)lA_0 e^{-\mu x}\,\delta x$$

where A_0 is the number of disintegrations occurring per ml. per unit time. The observed activity A' due to liquid of thickness d round the counter is then

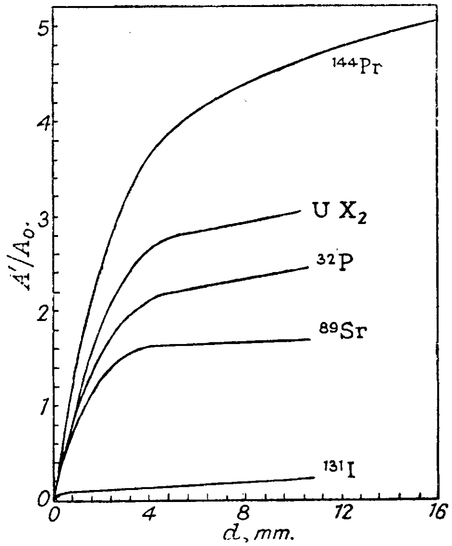
$$\begin{aligned} A' &= \int_0^d \pi(x + r)lA_0 e^{-\mu x}\,\mathrm{d}x \\ &= \left[-\frac{\pi l A_0}{\mu^2} e^{-\mu x}(1 + \mu x) - \frac{\pi r l A_0 e^{-\mu x}}{\mu} \right]_0^d \\ &= \frac{\pi l r A_0}{\mu}(1 - e^{-\mu d}) + \frac{\pi l A_0}{\mu^2}[1 - \mu d e^{-\mu d} - e^{-\mu d}] \quad . \quad . \quad . \quad . \quad (5) \end{aligned}$$

From equation (5) it will be seen that the observed activity increases as an exponential function of the thickness of liquid. Fig. 8 shows some experimental curves by Walton and Thom (private communication) for five β-emitters in which A'/A_0 is plotted against d. If d becomes infinite, then

$$\begin{aligned} A'_\infty &= \pi l r A_0/\mu + \pi l A_0/\mu^2 \\ &= (\pi l r A_0/\mu)(1 + 1/\mu r) \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6) \end{aligned}$$

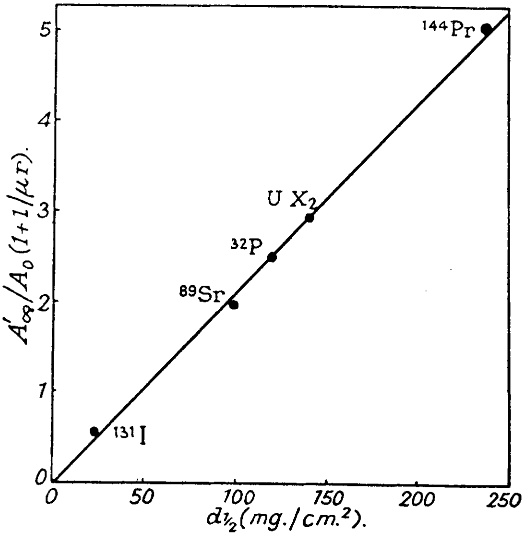
Hence it will be seen that if $A'_\infty/A_0(1 + 1/\mu r)$ is plotted against the absorption half-thickness $d_{\frac{1}{2}} = 0{\cdot}693/\mu$ (determined from the initial portion of a semi-logarithmic absorption plot), a straight line will be obtained as shown in Fig. 9; this was calculated from the values given by Walton and Thom for A'_∞/A_0, which had been corrected for the absorption of β-particles by the counter window and, in the case of ^{131}I and ^{144}Pr, for the small contributions of the γ-activities of these two emitters. The experimentally observed value of A'/A_0 at infinite thickness for ^{144}Pr given by Walton and Thom was 5·70. Since the measurements of half-thicknesses depend on experimental conditions, they cannot be regarded as a reliable index of the β-energies, but within the limits of error Fig. 9 is in accordance with equation (5).

FIG. 8.



Walton and Thom's experimental curves.

FIG. 9.



Plot of $A'_\infty/A_0(1+1/\mu r)$ *against* $d_{\frac{1}{2}}$.

For β-emitters of low energy, μ is very large, and equation (6) becomes

$$A'_\infty \doteq \pi l r A_0/\mu = \pi l r A_0 \times d_{\frac{1}{2}}/0{\cdot}693.$$

Hence the plot of A'_∞/A_0 is linear with respect to $d_{\frac{1}{2}}$, as has been shown by Walton and Thom.

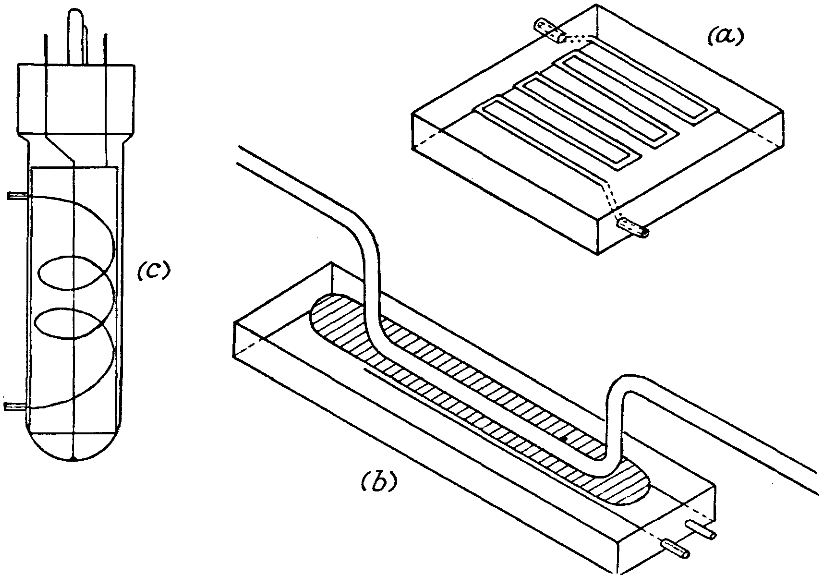
Most types of liquid immersion counters have fixed jackets to contain the liquid, and the volume of liquid assayed is therefore constant. Provided that measurements are made on one emitter only, the ratio of the counts obtained will be a direct measure of the ratio of the concentrations of radio-tracer, and fortunately the majority of measurements are of this nature.

Precautions should be taken, however, to ensure that the densities of compared liquids are not widely different, as absorption losses will not otherwise be comparable.

If it is desired to compare the activities of solutions of two different emitters, corrections must be applied for the different absorption coefficients by use of equation (5). Since this is only an approximate equation, the corrections are not very accurate, and in such cases, or whenever absolute measurements of activity are being made, the use of immersion counters is not recommended.

Liquid immersion counters may be made either of glass or of metal. If made of metal, it is desirable for the counter to have a thin window, usually made of aluminium. A thickness of less than 20 mg./cm.^{2} is seldom practicable because of the hydrostatic pressure exerted by the liquid. Most commercial glass immersion counters have a window thickness of about 35 mg./cm.^{2}, which is satisfactory for all except the softest β-emitters. For β-particles of less than about 0·6 Mev. maximum energy, at least 75% of the activity is absorbed by the counter window. In addition, absorption of β-particles of such low energy in the liquid surrounding the counter is great enough for a depth of 2 mm. of water to be regarded as of infinite thickness, so that only very small counting rates are obtained unless the specific activity is extremely high.

FIG. 10.



Three techniques for measuring the radioactive content of flowing liquids.

Two essentially practical points must be mentioned, both designed to ensure that strictly comparable conditions are used when two different activities are measured. The first is that any liquid jacket round the counter must be rigidly fixed, so that the thickness of liquid counted is always the same, particularly if an infinitely thick source is not used. Secondly, it is essential in all experiments in which counting rates are compared that the counter be immersed to the same depth in the liquid.

(iii) *Flow counters.* For some purposes it is convenient to measure continuously the radioactivity content of a flowing liquid. Three possible techniques are shown in Fig. 10. Method (*a*) is for measuring soft β-emitters. The channel ($\frac{1}{2}$ mm. deep) cut in a block of " Perspex " is sealed over with a thin sheet of " Distrene " of 2 mg./cm.^{2} thickness by means of a suitable cement. This method also has the advantage that a standard bell-jar type of counter can be presented to the surface of the " Distrene " window. The other two techniques require special counters. The counter shown in (*b*) is simply made of metal, but is not so efficient as that shown in (*c*), which is made of glass and allows the liquid to pass through the sensitive volume of the counter.

The dimensions of the counter shown in (*c*) depend on the type of radiation to be measured. For β-emitters of energy greater than about 1 Mev. counters are available in which the spiral is 1 mm. in diameter, with liquid capacity 0·3 ml. With γ-emitters, larger diameters and capacities may be used. During operation, the accumulation of charge arising by collision of gaseous ions with the non-conducting spiral, in the active space of the counter, may cause erratic behaviour. This is overcome by deposition on the spiral of " Aquadag " or colloidal graphite, so as to remove the charge as it is formed.

Flow counters are often used for determining the changes in radioactivity of an eluate from an experiment in which flowing liquids are used. If such changes are to be measured accurately, mixing of the liquid by turbulent flow, etc., should be avoided. Similarly the flow rate should not be slow enough for the concentration gradients to be disturbed by diffusion of ions in solution. The ideal conditions can be best estimated by a few trial displacements of an inactive by an active solution (and *vice versa*) in such a way that a sharp boundary exists between the two.

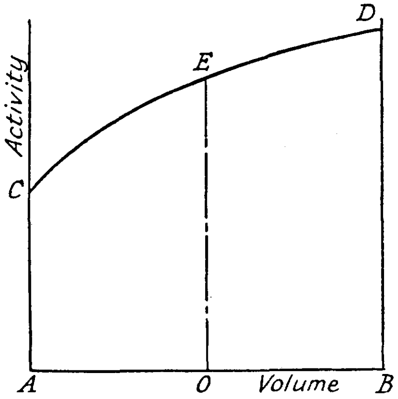
If the flow rate is large enough for the volume of the counter to be small compared with the volume passed through the counter during the time of measurement of the count, then provided the radioactivity does not vary sharply, the observed activity will be the mean activity for the volume passed during the time of measurement. Errors are, however, introduced because the counter has a finite volume if the flow rate becomes so slow that a volume comparable with the capacity of the counter is passed during the time of measurement, or if there are discontinuities in the volume distribution of the radioactivity.

Consider Fig. 11 where *AB* represents the capacity of the counter and the curve (*CED*) represents the activity (A) per unit volume at different volumes of solution, given by $A = \mathrm{f}(v)$. Now, at any instant, the activity measured is equal to

$$\int_{v-\frac{1}{2}v_c}^{v+\frac{1}{2}v_c} \mathrm{f}(v) \, . \, dv \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \qquad (7)$$

where v is the volume of liquid which has passed through the counter at the mid-point in time of the observation, and v_c is the capacity of the counter. If the flow rate is small enough to assume that the total radioactivity in the counter volume is constant during the time of measurement, expression (7) is the same as the observed count. The true activity at a volume v is given by the mid-point *E*, and is equal to $v_c \, . \, \mathrm{f}(v)$. The difference between the observed and the true counting rate is zero only when there is a linear variation in activity with volume. In other cases, the error obtained may be estimated if some reasonable assumption can be made as to the form of $\mathrm{f}(v)$, *e.g.*, that it is parabolic, or that it is to a first approximation the same as the observed activity–volume plot.

FIG. 11.

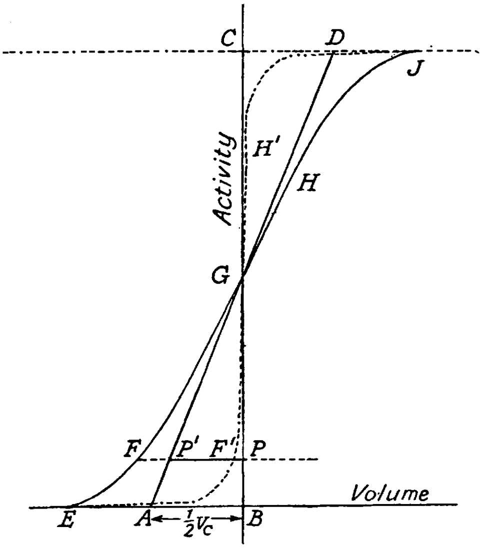


Distribution of activity in a flow counter.

If the flow rate is not negligible during the time of counting, it is necessary to integrate the count over the volume passed (v_1 to v_2) and compare it with the true count obtained at the mid-point of the volume measured, *viz.*, $v_c \, . \, \mathrm{f}(v)_{\frac{1}{2}(v_1+v_2)}$. The difference may again be estimated if the function $\mathrm{f}(v)$ is known or may be assumed.

Care should be taken in the use of flow counters when determining the shapes of sharp boundaries formed by replacement of an inactive by an active solution. Thus in Fig. 12 if the ideal boundary is represented by *ABCD*, then the observed boundary will be linear and given by *AD*. In practice, however, boundaries are seldom strictly linear, but distorted at the edges by diffusion of the ions across the interface, and have a form such as *EFGHJ*. If, however, the tangent at the point of inflexion of such an observed boundary is drawn, and this cuts the abscissa at a volume difference equal to $\frac{1}{2}v_c$ from the abscissa at the point of inflexion, then the shape of the true boundary may be estimated to a first approximation. For a given activity at a point *P* on a sharp boundary *ABCD*, the observed point would be *P'*. The difference *PP'* is therefore a measure of the distortion of a sharp boundary. Now the observed boundary *EFGHJ* cannot be obtained from a truly sharp boundary such as *ABCD*, and therefore the distortion of the boundary will not necessarily be the same. To a first approximation, however, it will be of the same order, and an approximate correction for the observed boundary may be obtained by subtracting the distance *PP'* from *FP* to give the point *F'*, which will be a point on a curve *EF'GH'J* approximating to the true boundary, which may be constructed.

FIG. 12.



Shape of observed boundary for sharp front.

Such a correction is only approximate and should be applied only when the tangent at the observed boundary cuts the volume axis at a distance approximating to $\frac{1}{2}v_c$ from the volume at the point of inflexion. If this is not obtained, or if the boundaries approximate to forms which may be expressed mathematically, corrections may be applied by means discussed elsewhere.

(iv) *Errors arising in the Measurement of* β*-Particles from Gases.*—If a gas emits an energetic β-particle, a vessel containing the gas may be directly presented to the counter. In this case the errors are very similar to those obtained in solid counting, *viz.*, external absorption, finite source size, etc.

In a more usual method the gas is introduced into the sensitive space of a counter, and in the case of the β-emitters of low energy this is the most reliable technique. When the gas is introduced into the counter, it is essential to be sure of the constancy of composition of the gaseous mixture used to operate the counter (usually the standard argon–alcohol filling). Similarly, the pressure should not be varied in comparative measurements since this alters the amount of activity introduced into the counter. Increase of pressure also increases the threshold voltage of the Geiger plateau, although this error is relatively small unless the pressure is altered greatly. The counter should not contain water vapour as otherwise the dead time will be increased, owing to the formation of positive ions (Korff, *op. cit.*), and considerable leakage of charge may occur (Kamen, *op. cit.*). If the determination of the absolute β-disintegration rate of a gas is necessary, the amount of radioactive gas actually measured must be found by estimation of the cathode volume of the counter, either geometrically or by calibration with a gas of known activity.

In comparative measurements, it is necessary to make correction for the paralysis time of the assembly. The paralysis time depends on the composition of the gas phase, and should be determined experimentally, unless the conditions of measurement are maintained rigidly constant throughout the given series of experiments or the paralysis time of the electronic circuit is set longer than any time during which the counter is likely to be inoperative. The error from this cause is usually not large for low counting rates (<2000 counts/min.).

In gas counting, care should be taken to ensure that the active gas remains in the gas phase and does not deposit active solids or become adsorbed on the walls of the counter. In most cases deposition is not serious but is most likely when using radio-isotopes with radioactive daughters; to lessen adsorption on the walls, counters should be of smooth finish.

Gas counting is most often employed for the study of β-emitters of low energy such as ^{14}C (0·14 Mev.) and ^{3}H (16 Kev.), which may be measured as gaseous carbon dioxide and hydrogen, respectively. A mixture of alcohol and hydrogen may be used for the determination of tritium, but gives poor Geiger plateaux, and it is usually better to convert the tritium into the oxide and measure it as $^{3}H_2O$ vapour, using a 9 : 1 argon–alcohol mixture for preference, and keeping the water vapour pressure below 2 mm. For ^{14}C a mixture of carbon dioxide and disulphide forms a satisfactory counting mixture (Brown and Miller, *Rev. Sci. Instr.*, 1947, **18**, 496).

After each measurement of an active gas, the apparatus must be carefully flushed out several times with an inactive gas to remove all traces of activity. Owing to adsorption on the walls of the counter, gas counters are subject to " memory " effects, which are more serious for some gases than for others. Tritium in particular is held badly on the walls of counters. After introduction of the various components into the gas counter, an interval is usually necessary before a constant counting rate is obtained. These two difficulties make gas counting tedious.

Flow gas counters have been used, on occasions, but since the gas is never in equilibrium in the counter the meaning of the results is always difficult to interpret. Errors due to the gas flow—*e.g.*, turbulent mixing, etc.—also make the results doubtful.

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