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ABSOLUTE BETA ASSAY WITH END-WINDOW
GEIGER-MUELLER COUNTERS

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ABSTRACT

Experimentally determined curves, which can be used to convert the activity of heavy samples (5 to 40 mg/cm²), mounted on aluminum cards and covered with a thin plastic film, to that of weightless samples on essentially zero mass backing, are presented. Some preliminary data is presented which shows the effect of sample thickness on the activity of thin samples (1 to 5 mg/cm²) on zero mass backing.

Half-thicknesses, which can be used to correct weightless sample activities for absorption by absorbers, air and counter window, are listed.

Most of the nuclides studied are beta emitters having energies of 1 Mev or greater.

INTRODUCTION

Samples prepared for radiometric assay with end-window counters are usually thick (5 to 40 mg/cm²), are mounted on a heavy backing (cardboard or aluminum), and are covered with a thin sheet of plastic material. When the activities of such samples are to be compared or when their disintegration rates are to be determined, there are several factors which enter into the measurements and which may lead to large errors. Two of these are scattering and absorption of beta particles. The reproducibility of the sample position and distribution is another factor, and the geometry is important when absolute disintegration rates are to be determined.

The backing, sample, sample cover, sample holder, air, and the shield are sources of scattered radiations. The sample, sample cover, counter window, and added absorbers act as absorbers. Scattered radiations cause an apparent increase in the activity of the sample, since radiations which start out in such a direction as not to enter the counter may be deflected into it. Scattered radiations are degraded in energy and, therefore, complicate the absorption characteristics of the sample.

*Most of the material presented has been extracted from the Ph.D. thesis of the author (Univ. of Chicago, 1952).

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Backscattering generally makes the largest contribution to the scattered radiation detected by the counter. It ranges in magnitude from zero to 80 percent and increases with the thickness and the atomic number of the backing and the energy of the beta particle.¹ Increasing the thickness of the backing causes the amount of scattered radiation to increase until the effect becomes saturated at a thickness of about one-sixth of the range of the particles. Backscattering has been extensively investigated.^{1,2,3,4}

Scattering by the sample itself makes an important contribution to the beta counting rate. The observed counting rate is affected by a combination of self-scattering and self-absorption factors. These factors cannot be readily separated. The magnitude (10 to 30 percent) of these effects has been studied as a part of this investigation and will be discussed later. Novey, *et al.*,⁵ Collie, *et al.*,⁶ and Nervik and Stevenson⁷ have made studies of this effect.

Air scatter has been shown to be negligible when the source is near the window.^{4,8} At a distance of several centimeters, however, the effect may amount to several percent. Wall and sample holder scattering are negligible if Lucite or aluminum is used as the structural material. However, scatter from the floor of the shield becomes important when the sample is placed on a low shelf.²

Absorption effects are usually small for hard betas (~ 1 Mev) when only a few milligrams of absorbing material are present between the counter and the sample. The magnitude of the absorption effect is usually determined by determining an absorption curve, a plot of activity versus thickness (mg/cm^2) of added aluminum absorber. This curve can be smoothly extrapolated back to zero absorber; hence, a condition denoting no air, window nor

¹"Radiochemical Studies: The Fission Products" (McGraw-Hill Book Co., Inc., New York, 1951), National Nuclear Energy Series, Div. IV, Vol. 9, paper No. 5 by D. W. Engelkemeir, J. A. Seiler, E. P. Steinberg, L. Winsberg, and T. B. Novey. This volume will be referred to as N.N.E.S., Vol. 9.

²B. P. Burt, *Nucleonics* 5, 28 (1949).

³L. Yaffe and K. M. Justus, *J. Chem. Soc.* 72, supplement No. 2, 342 (1949).

⁴L. R. Zumwalt, Mon-C-397 (1949).

⁵T. B. Novey, D. W. Engelkemeir, and W. Robinson, N.N.E.S., Vol. 9, paper No. 1.

⁶C. H. Collie, P. F. D. Shaw, and H. J. Gale, *Proc. Phys. Soc. (London)* A43, 282 (1950).

⁷W. E. Nervik and P. C. Stevenson, *Nucleonics* 10, 18 (1952).

⁸T. B. Novey and N. Elliott, N.N.E.S., Vol. 9, paper No. 3.

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added absorber. The initial shape of an absorption curve determines how accurately it can be extrapolated. This shape depends on the sample thickness, the backing, the position of the sample relative to the counter window, and the position of the absorbers.

If a weightless sample mounted on backing of zero mass is counted near the window (within a few mm), scattered radiations, which are degraded in energy and which therefore give a false absorption curve, are minimized. Furthermore, under these counting conditions an absorption curve taken with absorbers against the window of the counter is usually initially linear on a semilogarithmic plot. In such a case the initial half-thickness, $T_{\frac{1}{2}}$ (mg/cm²), can be used to calculate the absorption correction for a total amount of absorber, t (mg/cm²), using the standard expression $\exp(-\lambda t)$, where $\lambda = 0.693/T_{\frac{1}{2}}$.

The effect of source to window distance on scattering and absorption has been indicated above. The reproducibility of the sample position and distribution are also of importance.^{9,10,11} A vertical displacement of from 4 to 4.5 mm may lead to a 4.6 percent change in counting rate. At large distances the counting rate becomes less sensitive to small vertical displacements.

The distribution of active material can lead to large changes in the measured activity of a sample.^{10,11} This situation is particularly critical when thin samples (1 to 5 mg/cm²) are counted, since the active material may deposit itself all at one point or at the periphery of the sample area. For a sample at about 4 mm from the counter window (window diameter ~2.6 cm) changes in counting rate of as much as 10 percent are possible when the active material is displaced from a point 0.7 cm from the counter tube axis to 1.0 cm.

The counter geometry is important when absolute disintegration rates are required. If samples are only to be intercompared then absorption and scattering corrections are all that are necessary. A standard method for determining counter geometries, using weightless RaDEF standards mounted on essentially weightless backing, has been discussed by Novey.¹² The RaE beta activity of the standard is measured in the given counting set-up and is corrected for absorption and counting losses. The geometry is the ratio of the corrected RaE beta activity to the RaE disintegration rate (RaF alpha counting rate corrected for geometry, counting loss and genetics).

⁹L. R. Zumwalt, MDDC 1346.

¹⁰G. B. Cook and J. F. Duncan, J. Chem. Soc., 79, supplement No. 2, 369 (1949).

¹¹G. W. Reed, Thesis, University of Chicago (1952).

¹²T. B. Novey, Rev. Sci. Instr. 21 280 (1950).

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The accuracy with which the geometry can be determined is about 2 to 3 percent. The uncertainty arises predominantly from the sample distribution, the extrapolation to zero total absorber (RaDEF must be counted through sufficient absorber to remove the RaF alphas) and from contaminating radiations (see reference 11). These radiations are soft γ and X rays from RaD; they make a 1.5 percent contribution to the Geiger activity when counters filled with 8 to 10 cm of argon are used. The error from this source is negligible for helium-filled counters.

SCATTERING AND SELF ABSORPTION FACTORS

The object of the experiments described below was to determine factors to be used to convert the observed activity of a heavy sample (5 to 40 mg) on filter paper mounted on an aluminum card (~ 1.5 mm thick) and covered with about 0.5 mg/cm^2 rubber hydrochloride (RHCl) film to the activity of a weightless sample on zero backing with no cover and having the same number of active atoms. The sample area was 1.5 cm^2 . The samples were counted from a shelf about 4 mm from the counter window. The mica window was between 1.7 and 3.0 mg/cm^2 and had a diameter of about 2.6 cm. The counter tube was mounted on a sample holder of Lucite which was inserted into an aluminum-lined lead shield.

The general technique used in determining the scattering curves was similar to that described by Plutonium Project workers.¹³ In that work 2-cm^2 samples on filter paper were mounted on cardboard and covered with about 3 mg/cm^2 cellophane sheet. The weightless samples were evaporated onto 2 mg/cm^2 mica and were covered with cellophane for the first shelf (~ 5 mm from the counter window) experiments.

The experimental procedure used in the work described here was as follows: Radiochemically pure solutions of high specific activity of the nuclide to be studied were prepared. Aliquots (0.1 to 0.2 ml) of the tracer solution were deposited on RHCl films stretched over aluminum rings and into centrifuge tubes containing inert carrier. The solution on the film was confined to an area equal to the sample area (1.5 cm^2) and was spread evenly over this area. Insulin solution was used to define the area. Several such weightless (0.1 to 0.2 mg/cm^2) samples were prepared as checks. The samples to which carrier had been added were precipitated, filtered using a filter chimney to get a better defined area, washed, and dried according to the standard techniques used in fission product analysis. These samples were mounted on aluminum cards and covered with RHCl. All the samples were counted from the first shelf position in a standard counting arrangement (Fig. 1). Each sample was counted to at least one percent standard error.

¹³D. W. Engelkemeir, J. S. Seiler, E. P. Steinberg, and L. Winsberg, N.N.E.S., Vol. 9, paper No. 4

Fig. 1. Standard Counting Set-Up. (a) absorber, (b) spacer to raise absorber to counter flange, (c) source mounting ring with rubber hydrochloride film attached, (d) source in place on shelf.

Curves to be used in converting the heavy sample activity to that of a weightless one were constructed by plotting the ratio of the activity of the heavy sample to that of the weightless ones corrected to the same chemical yield against the mass of the heavy sample. This ratio was defined as the scattering and self absorption factor and is plotted as the ordinate in Fig. 2 a-g. The point at (0, 1.00) in Figs. 2 a-g refers to a weightless, backless source. The counting rate of a weightless sample is determined from that of a heavy one by dividing the counting rate of the latter by the scattering factor corresponding to the sample weight.

Curves are given for:

Nuclide	E_{β} max (Mev)
Ba ¹³⁹	2.3
Zr ⁹⁷ -Nb ⁹⁷	1.9, 1.3
Sr ⁸⁹	1.5
Mo ⁹⁹	1.2, 0.45
Ba ¹⁴⁰ -La ¹⁴⁰	1.0, 0.48, 2.3, 1.7, 1.3
Zr ⁹⁵	0.4

Fig. 2a. Sr^{89} Scattering and Self-Absorption Curve. Source area - 1.5 cm^2 . Source to window distance - 0.4 cm . Total absorber - 2.4 mg/cm^2 .

Fig. 2b. Zr^{97} - Nb^{97} Scattering and Self-Absorption Curve. Source area - 1.5 cm^2 . Source to window distance - 0.4 cm . Total absorber - 2.4 mg/cm^2 .

Fig. 2c. Mo^{99} Scattering and Self-Absorption Curve. Source area - 1.5 cm^2 . Source to window distance - 0.4 cm . Total absorber - 23.4 mg/cm^2 .

Fig. 2d. Mo⁹⁹ Scattering and Self-Absorption Curve.
Source area - 1.5 cm². Source to window
distance - 0.4 cm. Total absorber -
12.3 mg/cm².

Fig. 2e. Ba¹³⁹ Scattering and Self-Absorption Curve.
Source area - 1.5 cm². Source to window
distance - 0.4 cm. Total absorber -
2.4 mg/cm².

Fig. 2f. Ba¹⁴⁰-La¹⁴⁰ Scattering and Self-Absorption
Curve. Source area - 1.5 cm². Source to
window distance - 0.4 cm. Total absorber -
2.4 mg/cm².

Fig. 2g. Zr^{95} Scattering and Self-Absorption Curve.
 Source area - 1.5 cm^2 . Source to window distance - 0.4 cm . Total absorber - 2.4 mg/cm^2 .
 Solid Curve - 65-day Zr^{95} into which 35-day Nb^{95} has grown for 12-13 days.
 Dashed Curve - Pure Zr^{95} obtained by correcting experimental curve for Nb^{95} activity.

Some auxiliary data were gathered that may be of use in some experiments. Sr^{89} samples were counted on two different tubes and the scattering factors were found not to change significantly. The effect of absorbers placed against the counter window was studied. The scattering factor was found to change slowly with increasing absorber weight. Sr^{89} samples gave a scattering factor of 1.37 ± 0.01 with no added absorber and 1.33 ± 0.02 with 31.2 mg/cm^2 aluminum against the window. Mo^{99} scattering curves with 21.0 and 9.85 mg/cm^2 added aluminum absorber are given in Fig. 2c and d. (Mo^{99} was always counted through absorber to cut out the Tc^{99m} conversion electrons.) Data were also taken with 13.3 mg/cm^2 of added absorber. In the sample mass range from about 9 to 25 mg the average scattering factors from the 21.0, 13.3, and 9.85 mg/cm^2 curves were 1.40 ± 0.02 , 1.34 ± 0.03 , and 1.33 ± 0.02 , respectively.

The data has been summarized in Fig. 3 in a manner analogous to that used by Engelkemeir, *et al.*¹³ The reciprocal of the scattering factor is plotted versus the E_{max} of the beta emitter for samples having a mass of 20 mg. For the purpose of this plot, some of the factors were corrected for: contaminating activities (Nb^{95} in Zr^{95}), conversion electrons (Ba^{139}), and strongly absorbed weak components (the 0.48-Mev beta in the Ba^{140} - La^{140} mixture).

Fig. 3. Scattering and Self-Absorption Factors vs. $E_{\beta \text{ max}}$.
 Source area - 1.5 cm^2 . Source to window distance -
 0.4 cm.
 Circles - Original points from experimental data.
 Squares - Experimental points corrected for decay
 complexities.

DATA RELEVANT TO FIGURE 3

Nuclide	$E_{\beta \text{ max}}$	f_s (exp)	f_s^a (corrected)	f_s^{-1} (corrected)
Ba ¹³⁹ (a)	~2.3	1.30	1.33	0.746
Zr ⁹⁷ -Nb ⁹⁷	1.9, 1.3	1.46	1.46	0.685
Sr ⁸⁹	1.5	1.37	1.37	0.730
Mo ⁹⁹ (b)	1.2, 0.45	1.41	1.41	0.711
Ba ¹⁴⁰ -La ¹⁴⁰ (a)	1.0, 0.40, 1.3, 1.7, 2.3	1.26	1.39	0.719
Zr ⁹⁵ (a)	0.4	0.77	0.81	1.23

Average ($E_{\beta \text{ max}} > 1 \text{ Mev}$)

1.39

0.718

^aSee text, page 5.

^bMo⁹⁹ counted through 23.4 mg/cm^2 total absorber.

The average increase in activity for 1-Mev beta emitters mounted on aluminum was about 37 percent as against about 12 percent for samples on cardboard.¹³ The Plutonium Project results are affected by the fact that about 3.5 mg/cm² of cellophane covered the weightless samples. Scattering from this plus that from the mica backing (~2 percent) might easily increase the measured activity of the weightless sample by 10 percent with a corresponding decrease in the observed scattering factor.

The curve in Fig. 3 is flat for beta energies greater than 1 Mev, indicating that in this region the amount of scattering is relatively independent of the energy of the beta. The flat portion of the curve was extended down to 0.6 Mev on the basis of the results of Engelkemeir, *et al.*

From the figures it can be seen that backscattering makes the major contribution to the scattering factor. Merely placing the weightless samples deposited on RHC1 in contact with an aluminum card as a backing raised the counting rate 33 percent for Sr⁸⁹, 27 to 30 percent for Mo⁹⁹ and 24 percent for Ba¹⁴⁰-La¹⁴⁰; this represents, on the average, 95 percent of the scattering factor. The other 5 percent arises from self scattering and self absorption. Experiments measuring this effect are described below.

SELF-SCATTERING AND SELF-ABSORPTION BY RADIOACTIVE SOURCES

Whereas the scattering from the backing was most important in the effect described above, scattering by the sample itself may also be large. Self-scattering and self-absorption curves were determined for Sr⁸⁹, Mo⁹⁹, Ba¹⁴⁰-La¹⁴⁰, Ba¹⁴⁰, and UX₂. Usually the samples were prepared by adding a given amount of tracer activity to several RHC1 source discs (sample area of 3.14 cm² except for the UX₂ experiment in which it was 1 cm²) and then adding varying amounts of carrier directly to the tracer on the film. The solutions were evaporated and the residue was spread evenly using a glass rod drawn to a point and rounded off. In the case of UX₂, the oxalate was precipitated, centrifuged and transferred as a slurry by means of a pipette to the sample disc. The total amount of activity was accounted for. Strontium and barium were evaporated as the chlorides and molybdenum as PbMoO₄.

The factor determined was the ratio of the activity of the samples with carrier to that of the weightless sample. This ratio was plotted against the sample mass in mg/cm² to obtain the scattering curves (Fig. 4 a-d). The UX₂ and Mo⁹⁹ samples were counted through sufficient absorber to remove the UX₁ and Tc^{99m} radiations, respectively.

Fig. 4a. Sr^{89} Self-Scattering and Self-Absorption Curve. Source area - 3.14 cm^2 . Source to window distance - 0.4 cm. Total absorber - 3.5 mg/cm^2 .

Fig. 4b. Ba^{140} , $\text{Ba}^{140}\text{-La}^{140}$ Self-Scattering and Self-Absorption Curves. Source area - 3.14 cm^2 . Source to window distance - 0.4 cm. Total absorber - 3.5 mg/cm^2 .

Fig. 4c. Mo⁹⁹ Self-Scattering and Self-Absorption Curve. Source area - 3.14 cm². Source to window distance - 0.4 cm. Total absorber - 13.4 mg/cm².

Fig. 4d. UX₂ Self-Scattering and Self-Absorption Curve. Upper curve: Source to window distance - 0.4 cm. Lower curve: Source to window distance - 0.9 cm. Source area - 1 cm². Total absorber - 24.5 mg/cm².

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It can be seen that the self-scattering effect becomes saturated at fairly low sample masses, about 5 mg/cm² for strontium, molybdenum and UX₂. The barium data is sketchy, but the maximum at 1 mg/cm² might be associated with the 0.48-Mev beta. Scattering by the sample can, on the basis of these experiments, cause at least a 10 percent increase in the sample activity.

As has been pointed out, backscattering from weightless sources placed over aluminum cards accounted for 95 percent of the scattering factor observed for heavy samples mounted on aluminum. The other 5 percent, therefore, arises from self scattering and absorption by the sample and the cover. Since scattered radiation is degraded in energy and in heavy samples backscattered radiation must pass through the entire sample, the net contribution of scattered radiation is less than the amount scattered toward the counter. Thus the net scattered radiation from heavy samples on aluminum backing can be accounted for as equal to:

Backscattered +

Self-scattered -

The fraction of the backscattered absorbed by the sample.

ABSORPTION HALF-THICKNESSES

The initial half-thicknesses measured for weightless samples of 1.5 cm² area and mounted on 0.5 mg/cm² RHC1 are given below. The half-thickness obtained for heavy samples mounted on aluminum are also given to indicate the possible effect of scattered radiation. The last column lists data taken from Vol. 9.

Nuclide	E _{max} (Mev)	Approx. Range of added absorbers (mg/cm ²)	T _{1/2} (Weightless)	T _{1/2} (Heavy)	T _{1/2} (Plutonium Project)
Sr ⁸⁹	1.5	0 - 32	97.7 82.8 (3.142 cm ² sample area)	78	88
Zr ⁹⁷ -Nb ⁹⁷ (equil.)	1.9, 1.27	0 - 47	93.2	74	95
Zr ⁹⁵	0.4	0 - 10 10 - 40	9.4 15 - 16	9.8	
Mo ⁹⁹	0.45, 1.22	7.5 - 21.0 21.0 - 70	42.0 57	54	55
Ba ¹³⁹	2.3	0 - 70	134		

APPENDIX

A summary of the steps necessary to determine sample disintegration rates is outlined below. The ideal counting arrangement consists of a weightless, coverless, and backless sample measured in a counter where there is no absorption and, if disintegration rates are desired, at 100 percent geometry. Samples routinely prepared weigh about 20 mg, are mounted on filter paper and an aluminum card, and are covered with from 0.5 to 3.0 mg/cm² rubber hydrochloride or cellophane. The samples are counted at a low geometry (20 to 30 percent) and the radiations must pass through absorber, air and counter window. To attain the ideal situation the following steps are necessary.

1. After correction for coincidence losses, the observed activity (A) of the sample is divided by the scattering factor (ordinate of Figs. 2a-g). This gives the activity of a weightless sample on zero backing.
2. Correction for absorption is obtained by multiplying by $\exp(0.693 \times t/T_{1/2})$, where t is mg/cm² of added absorber, air and window and $T_{1/2}$ is the absorption half-thickness for a weightless and backless sample. These two corrections give the activity, $D \times G$.
3. If necessary, correct for contaminating radiations, gammas, betas, etc. This correction may be made on either the heavy or weightless sample activity depending on which one it is known for.
4. The activity ($D \times G$) can be converted to a disintegration rate by multiplying by a geometry factor ($1/G$). The geometry of the counter may be determined by counting a weightless RaDEF standard on the same shelf the sample is counted on.
 - a. Add ~ 10 mg/cm² total absorber to cut out RaF alphas when counting the standard.
 - b. Correct count c to zero total absorber

$$A' = c \times \exp(0.693 \times t/T_{1/2}) = c \times f(t)$$

where t = mg/cm² of window, air and added absorber.

$T_{1/2} = 33.3$ mg/cm² for a standard counted close to the window (within about 4 mm).

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- c. Correct count for any contribution from extraneous radiations, e.g., a 2 percent contribution from RaD electromagnetic radiations when argon-filled (8 to 10 cm) counters are used. No correction for helium-filled counters.

$$A = A' \times g; \quad g = 0.98 \text{ for argon-filled counters.}$$

Therefore,

$$\text{geometry} = A/D = c \times g \times f(t)/D,$$

where D is the actual disintegration rate of the standard at the time of the geometry measurement and is determined from the RaF alpha disintegration rate and the factor (1.018) expressing the genetic relationship for an equilibrium mixture of RaDEF.