Energy of Beta-Radiation from S^{35} and C^{14}

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Aluminum-absorption curves have been obtained for C^{14} and S^{35}. The ranges in aluminum, measured as suggested by Feather, are 27.9 ± 0.3 mg/cm^2 for C^{14} and 31.4 ± 0.5 mg/cm^2 for S^{35}. The energies deduced from these ranges are 154 ± 4 kev for C^{14} and 167 ± 4 kev for S^{35}. Measurements have also been made of the self-absorption of $BaC^{14}O_3$, yielding an absorption coefficient $\alpha=0.29$.

1. INTRODUCTION

WITH the intense sources of radioactive S^{35} and C^{14} now available from the Atomic Energy Commission, it is possible to assay the maximum energy of the emitted beta-radiation by the absorption method with a high degree of accuracy. Feather[1] has developed the technique of absorption measurements to give precise values of the maximum energy of several beta-emitting isotopes. His method depends upon comparison of the unknown range with the known range (476 mg/cm^2 of aluminum) and spectrum of RaE, which he chose because it is free from gamma-emission. In particular, the radiation transmitted through aluminum foil from the source being studied is compared with that transmitted by RaE at several absorber thicknesses. In practice the net count per minute from the source is plotted against absorber weight expressed as mg/cm^2. From the RaE absorption curve, obtained with identical geometry, the percent of RaE radiation transmitted can be obtained at absorber thicknesses chosen as 0.1, 0.2, 0.3 up to 1.0 times the maximum thickness for RaE, i.e., 47.6, 95.2, etc., mg/cm^2 of aluminum. Then, by comparison with the unknown source, say $BaC^{14}O_3$, the thickness of absorber corresponding to these transmission percentages for C^{14} can be obtained. Finally these values, multiplied by $10/n$, i.e., 10, 5, 3.33, etc., are plotted against n, the number of tenths of absorber. These points lie along a smooth curve called a *Feather plot*, a curve which can easily be extrapolated to a good end point in case the sample is too weak or too gamma-active to permit a complete beta-absorption curve. If the unknown source should have a spectrum identical with that of RaE, the Feather plot would be a straight line, parallel to the abscissa.

[1] Feather, Proc. Camb. Phil. Soc. **35**, 599 (1938).

2. EXPERIMENTAL DETAILS

A low background thin-window counter sketched in Fig. 1 has been devised for use in these measurements and also for the routine measurement of $BaC^{14}O_3$ samples in biochemical research. Counters of this type give stable low backgrounds; indeed, throughout all the absorption experiments reported here, the average background varied only between 4.3 and 5.3 counts/min. Thin mica windows about 2.0 to 2.5 mg/cm^2 are sealed onto the counter with

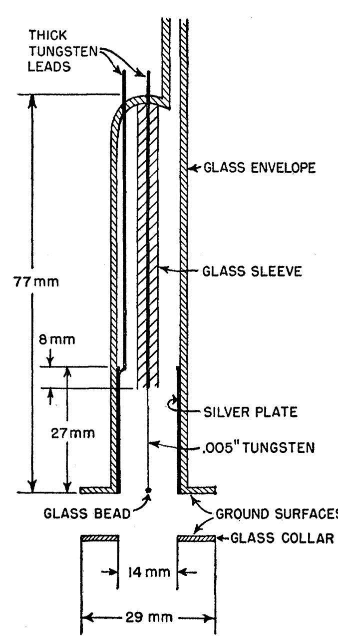

FIG. 1. End-window counter.

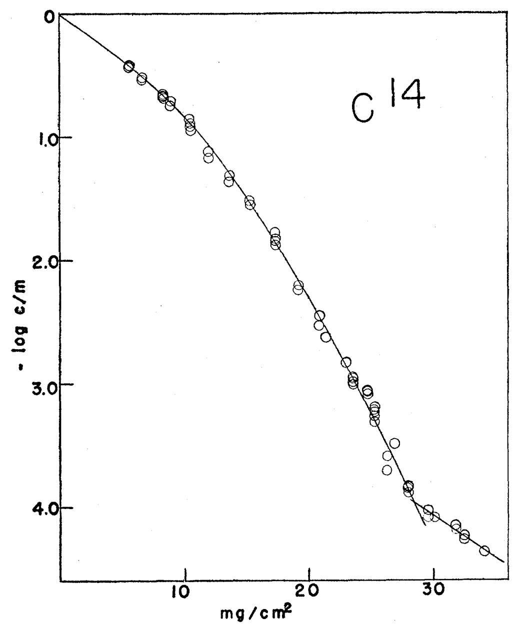

FIG. 2. Absorption of C^{14} by aluminum.

Vinyl-seal AYAB resin in acetone (70 percent).* Using so small an area there would appear to be no difficulty in sealing on windows as thin as 1.0 to 1.5 mg/cm^2; in practice we have had more trouble with faulty cementing than with window thickness.

For counting, the samples were placed in thin (0.017 inch) aluminum-stamped cups, which fit into an accurately machined dural slide. The slides in turn were placed on shelves in an accurately machined dural housing. For these experiments a shelf was chosen in which the base of the cup was 2.18 cm below the mica window. This large air gap was required to accommodate the aluminum foils used for the RaE absorption experiments. The whole counting assembly was housed inside a dural-lined lead chamber, one inch in thickness. All three sources, C^{14}, S^{35}, and RaE, were very thin; by visual estimate total weights of 0.1 to 0.5 mg in an area of 3.1 cm^2. The initial strength of the sources was C^{14}: 13,100 counts/min., S^{35}: 11,800 counts/min., RaE: 8800 counts/min. The contribution due to γ-rays and other unabsorbed contaminants was: C^{14}: 2.5 counts/min., S^{35}: 15 counts/min., RaE: 2.0 counts/min.

For RaE, the visual range in aluminum was found to be 476 mg/cm^2 in agreement with the value accepted by Feather.[1] The visual ranges for C^{14} and S^{35} are, respectively, 28.5$\pm$0.3 mg/cm^2 and 31.4$\pm$0.6 mg/cm^2; the data from which these conclusions are drawn are plotted in Figs. 2 and 3. The Feather ranges are 27.9 $\pm$0.3 mg/cm^2 for C^{14} and 31.4$\pm$0.5 mg/cm^2 for S^{35}; the Feather plots are given in Fig. 4.

3. SELF-ABSORPTION

In the course of biological experiments it is of particular interest to measure self-absorption so that samples of varying weight may be compared exactly. With elements having radiation as soft as those of C^{14} and S^{35}, the self-absorption correction becomes very large even with thin samples. For the biological work and, incidentally, for the determination of self-absorption correction, samples were deposited in stainless-steel cups accurately machined to provide wells measuring 1.4 cm in diameter by 1 mm deep. Initially, considerable difficulty was experienced in forming a $BaC^{14}O_3$ precipitate flat and uniform

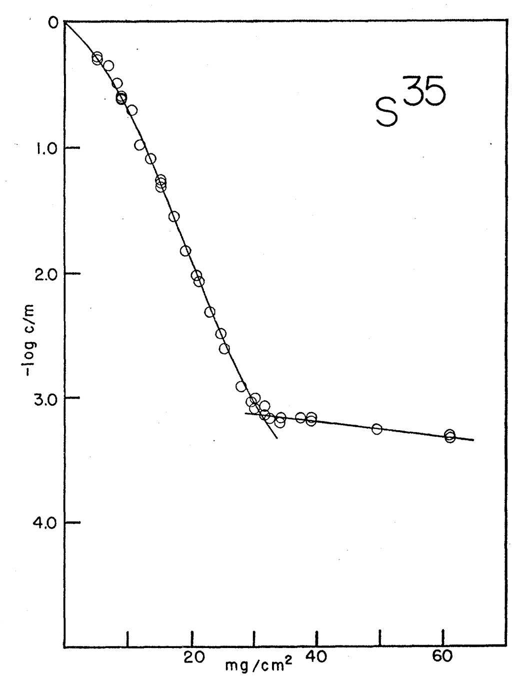

FIG. 3. Absorption of S^{35} by aluminum.

* In order to obtain good seals, it is necessary to let the solvent evaporate almost completely before attaching the mica.

enough to give reproducible counting rates. Finally, we adopted a technique of transferring precipitates by micro-pipette. After precipitation in a 15-ml centrifuge tube, the $BaC^{14}O_3$ is centrifuged. The excess $Ba(OH)_2$ is removed and the precipitate washed by suspension in a mixture of 1 ml of CO_2-free water and 2–3-ml alcohol (to prevent creeping). After centrifugation, the wash solution is poured off and the precipitate drained. The precipitate is then suspended in a few drops of water (less than 0.3 ml) and transferred to the stainless-steel cups with a micro-pipette (an eye dropper with the tip drawn to a capillary). During evaporation to dryness over a hot plate maintained at 45–60°C in a hood, the suspension is stirred with a thin rod to make it more uniform. A set of ten identical measurements showed that this technique gave results reproducible to better than 1.5 percent, including errors in counting. Some effort was made to extend the absorption experiments to densities lower than 1 mg/cm², but it was found that recovery is apparently incomplete when the total amount of solid is less than 1 mg corresponding to a density of 0.65 mg/cm².**

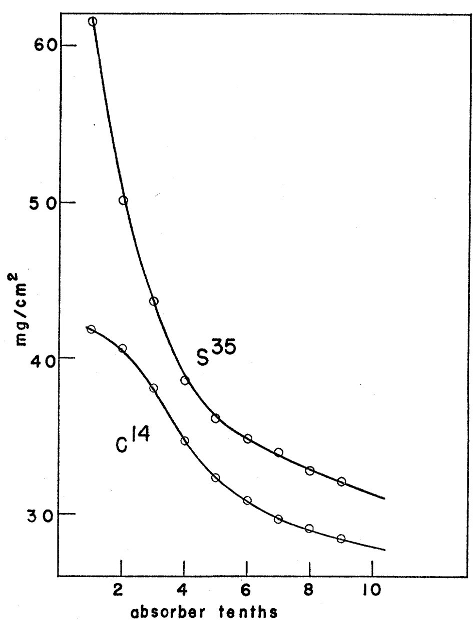

FIG. 4. Feather plot for C^{14} and S^{35}.

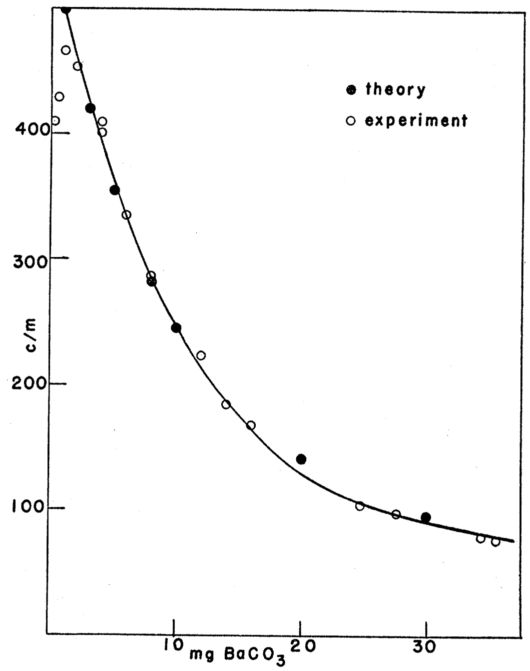

FIG. 5. Self-absorption of $BaC^{14}O_3$.

The simple equation governing self-absorption is given by Henriques[2] *et al.*, and also by Libby.[3] Derived from a simple absorption law, $I = I_0 e^{-\alpha d}$, the self-absorption equation is:

$$I = I_0 \frac{I - e^{-\alpha d}}{\alpha d}$$

where I = intensity transmitted, I_0 = intensity before absorption, α = absorption coefficient, and d = thickness in mg/cm².

The self-absorption results obtained in the course of this research are shown in Fig. 5. With a value of α of 0.29 cm²/mg, a good fit can be obtained from 0.65 mg/cm² to 13.0 mg/cm², a somewhat surprising result in view of the over-simplifications of the theory, and the fact that, as shown in Fig. 2, the absorption of C^{14} electrons in aluminum does not follow a simple exponential

** Note added in proof: I am indebted to L. E. Glendenin for pointing out that this effect is due to a hitherto unpublished phenomenon called "self-scattering." Apparently, sizable precipitates of the order of 5% of maximum range expressed as mg/cm²—exert a focussing effect on the beta particles, and scatter more particles up and down at the expense of those that emerge in a horizontal direction, thereby increasing the counting rate of the source. A. K. S.

[2] Henriques, Kistiakowsky, Margnetti, and Schneider, Ind. and Eng. Chem. (Anal. Ed.) 18, 349 (1946).

[3] Libby, Ind. Eng. Chem. (Anal. Ed.) 19, 2 (1947).

law. Yankwich[4] obtains a value of 0.285 ± 0.008 for α in good agreement with the present results.

4. DISCUSSION

To convert the range in aluminum of C^{14} and S^{35} to absolute energy values is difficult. The data of Marshall and Ward[5] on monoenergetic electron absorption cover this range, though the equation of Feather[1] does not extend to energies so low. However, Glendenin[6] has constructed a smooth empirical curve covering the whole range of beta-absorption, from 0.01 Mev to 3 Mev using the values from monoenergetic electrons in the low range and beta-particles determined in a beta-ray spectrograph in the high range. This curve flows smoothly from the data of Marshall and Ward for monoenergetic electrons to data obtained with beta-ray spectrometers, and does not leave the Marshall-Ward curve until above 0.5 Mev. Consequently, it seems valid to assign from the Marshall-Ward curve a value of 154 ± 4 kev for C^{14} and 167 ± 4 kev for S^{35}.

The value for C^{14} is in good agreement with the original absorption limit of 145 ± 15 kev given by Ruben and Kamen.[7] The recent report of Stephens and Lewis[8] gives a value of 150 ± 20 kev determined by the beta-ray spectrometer, also in good agreement as is the value of 154 kev obtained by Levy.[9] The S^{35} value differs by more than the experimental error from the value of 107 ± 20 kev given by Libby and Lee,[10] but is in better agreement with Kamen's[11] determination of 120 ± 15 kev. Recent unpublished work by Osborne[12] gives a beta-ray spectrometer value of 169 ± 3 kev in good agreement with the result given here.

Since the experimentally determined self-absorption curve follows a simple exponential closely, it is tempting to try to correlate the self-absorption coefficient obtained for $BaC^{14}O_3$ with the aluminum-absorption coefficient for C^{14}. Such a comparison should be valid since, as is well-known, the composition of the absorber is of small importance in these low energy regions, where density of absorber is the major factor. However, examination of Fig. 2 shows that in the case of C^{14} no single unequivocal absorption coefficient can be picked since the aluminum absorption is so far from exponential. In the case of S^{35}, the aluminum absorption is more nearly exponential, and an absorption coefficient, $\alpha = 0.29$ can be obtained in good agreement with the value of 0.27 given by Henriques[13] for S^{35}.

The radical difference in shape between the absorption curves for S^{35} and C^{14} is immediately apparent from Figs. 2, 3 and 4. This difference may well be attributed to the difference in the forbidden characters of the transitions, as evidenced by the great difference between the 87.1-day half-life[14] of S^{35} and the 4700-year half-life[15] of C^{14}.

In spite of this difference, the values for α are remarkably uniform, and it is probable that the equation proposed by Libby[3] that $\alpha l_0 = k$, where l_0 is the maximum range, and k is a constant, can serve as a guide to the estimation of α. From this research, the value of k would appear to be 8.6, higher than the value of $k = 5$ chosen by Libby from earlier data on the S^{35} range.

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