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A CALCULATION OF THE BETA ENERGY ABSORBED BY TRITIATED GASES IN SPHERICAL VESSELS

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A CALCULATION OF THE BETA ENERGY ABSORBED BY TRITIATED GASES IN SPHERICAL VESSELS

by

Marvin M. Mueller

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ABSTRACT

A calculation of the tritium beta energy absorption in spherical bulbs filled with mixtures of tritium and other gases is carried out under the empirically substantiated assumption that the energy absorption per spherical shell surrounding a point tritium source is representable by a pure exponential function. Primary and successive backscattering at the wall of the vessel is taken into account by means of a very approximate calculation. The computed energy absorption function for hydrogen gas in a silvered bulb agrees within ± 3% over a wide range of gas pressure with the one available set of spherical ionization chamber data.

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Chapter 1

INTRODUCTION

The need for information concerning the tritium beta energy absorption by spherical volumes of gas arose out of an experimental study of isotope exchange rates in mixtures of tritium and methane gas conducted in Group W-7. These experiments were carried out in spherical glass bulbs varying in volume from 100 to 1000 ml and at total gas pressures varying from 65 to 400 mm Hg. In the majority of cases, the gas mixture was predominantly tritium. Since under these conditions the range of the tritium beta particles is of the order of the bulb diameter, it is clear that not all of the emitted beta energy is absorbed by the gas.

The problem to be treated in this report is essentially this: given a specified homogeneous medium containing a dispersed beta emitter bounded by a spherical wall of known composition, find the fraction of the total emitted beta energy that is absorbed by the medium. While the formal content of this report would be applicable to any state of aggregation of a medium of fairly low atomic number, for the sake of concreteness we shall use a nomenclature appropriate to the gaseous state.

Because of the great complexity of electron scattering processes, any fundamental approach to the problem we entertain here would be far from simple. Indeed, no theoretical solution has yet been obtained for the electron flux distribution in a physically bounded medium.

Since no direct, purely theoretical scheme of calculation is presently available, we shall have recourse to a less fundamental, two-step approach to the problem. In the first step we neglect backscattering from the vessel wall. This is equivalent to assuming that the medium is homogeneous and unbounded in its absorbing properties, although the beta emitter is restricted to a definite sphere in the medium. Then in the second step we calculate the energy absorption due to the backscattered beta particles and add this to the energy absorption calculated in the first step.

If both the medium and the dispersion of the beta emitter therein were infinite (relative to the range of the beta particles), the calculation for the first step would be trivial, since the energy absorbed per unit volume must perforce equal the energy emitted per unit volume. In the actual case, a real problem arises since the rate of energy absorption is a function of the distance from the center of the sphere due to the escape of beta particles from the sphere of beta emission. In order to calculate this energy absorption function, we must know the spacial dependence of the energy absorption surrounding a point beta emitter in the absorbing medium. This aspect of the problem will be treated in the chapter which follows.

Chapter 2

THE ENERGY ABSORPTION FUNCTION FOR SPHERICAL GEOMETRY NEGLECTING BOUNDARY BACKSCATTERING

The physical input required for this calculation is the knowledge of the energy absorption per spherical shell of absorbing gas surrounding a point source of tritium. While a direct experimental study of this point-source function has not been carried out for the case of tritium, this has been done for beta sources of higher energy. From these data at higher energies, we shall make an attempt to adduce the point-source absorption function for tritium beta particles.

First, however, let us note that we are not discussing "absorption" in the sense it is customarily used in radiation physics, but rather in its radiological sense of "dose." For what is usually referred to as "absorption" in physics could probably be better termed "attenuation," since it is the reduction of particle beam intensity as a function of absorbing film thickness which is usually studied. Experimentally, it is found that the intensity of a beam of beta particles usually decreases in an approximately exponential manner with absorbing film thickness. This fact is, however, of no use to our calculation since there is no necessary relation between the energy absorption function we seek and the particle attenuation function usually measured.

Fortunately, however, an experimental study of the rates of ionization in hydrogen, air, and argon surrounding point beta sources of relatively low energy has been carried out.[1] The authors conclude that the energy absorption per spherical shell in these gases is rather accurately represented by a decreasing, pure exponential function of the distance from the source for beta sources of less than about 200 kev mean beta energy. It was also found that this exponential dependence is far from being valid in the case of high atomic number absorbers and/or higher beta energies. Indeed, for these cases, a semilogarithmic plot gives S-shaped or even peaked curves. Thus, the exponential point-source absorption function must be regarded as merely a fortuitous simplification, valid in the dual region of moderately low atomic numbers and low-energy beta particles.

Since, however, the lowest energy beta emitter studied is S^{35} with a mean beta energy 8.6 times as large as the

5.7 kev mean beta energy of tritium, some question arises as to whether the assumption of an exponential absorption function for tritium is justifiable. Happily, the assumption is rather well substantiated--although indirectly--by the work of Grün[2] who measured light emission caused by monoenergetic, collimated electron beams in air. For electron beams varying in energy from 5 to 54 kev, Grün obtained plane energy absorption functions (roughly of haystack shape) having practically the same shape for all energies. This would seem to constitute good evidence that the shape of the point-source energy absorption function for tritium is the same as for S^{35}--that is, exponential.

Hence, in what follows, we assume with a fair degree of confidence that the energy absorbed per spherical shell of gas of moderately low atomic number surrounding a point source of tritium may be adequately represented by the expression

$$D(r) = Ae^{-\mu r}$$

where D(r) is the dose rate for a spherical shell of gas of radius r concentrically surrounding the point tritium source, and μ is the linear energy absorption coefficient for the particular absorbing gas. To a later section we leave the problem of estimating the numerical values of μ for various gases. Our present concern will be to make a formal calculation of the dose rate for a spherical volume of gas neglecting boundary backscattering. The constant A in the above expression can be related to the source strength s of the point beta emitter by the relation

$$s = \int_0^\infty D(r)dr = \frac{A}{\mu}$$

and we thus obtain the normalized expression,

$$D(r) = \mu s e^{-\mu r}. \qquad (1)$$

In this calculation and in all that follows we neglect the fact that the range of the beta particles is finite, whereas the exponential function extends to infinity. However, since the data of Sommermeyer[1] show that the point-source absorption function is exponential out to a range of at least $4/\mu$, this neglect produces only slight error, while affording considerable mathematical simplification. If the finite range of the beta particles were to be taken into account, the calculated spherical absorption function would be increased by an amount varying from 0 to about 1.5%, depending in a complicated manner on the specific conditions.

We will now make use of eq. (1) to calculate the dose rate (energy absorbed per unit of time) as a function of position in a sphere of gas in which tritium is distributed uniformly. The geometry involved is illustrated by Figure 1.

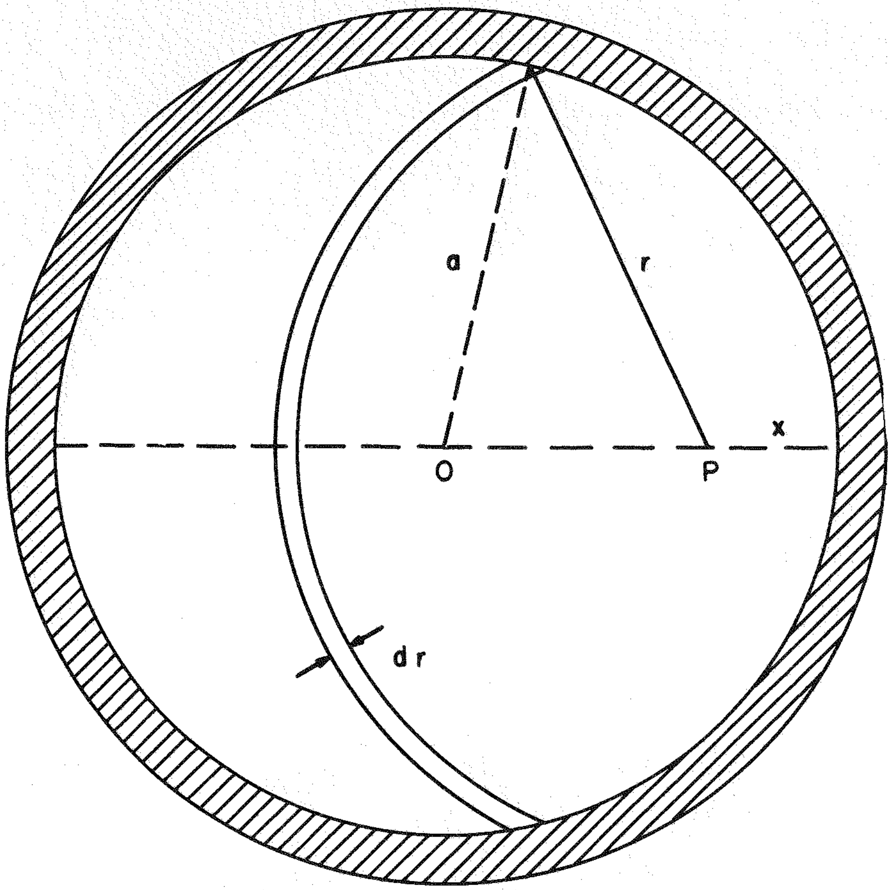

Fig. 1. The spherical absorption geometry.

Consider a point P located a distance a – x from the center O of a sphere of radius a. The beta energy emitted per second per unit volume of the gas is ϵ_o. If the medium were infinite, the dose rate in an infinitesimal volume element dV located at P due to all of the distributed sources located in the spherical shell of radius r surrounding P would be

$$D(r)\ dr\ dV = \mu\ \epsilon_o\ e^{-\mu r}\ dr\ dV.$$

However, since the medium is finite, only a fraction of the spherical shell for $r>x$ contains beta sources. From solid angle considerations one can show that the fraction of the shell for $x \leq r \leq 2a - x$ that is within the emitting medium is

$$\frac{1}{4(a - x)}\ (2a - x - r)(1 + \frac{x}{r}).$$

For $r \leq x$, the fraction is unity and for $r \gtrsim 2a - x$, the fraction is zero.

Hence, the actual dose rate at P due to the spherical shell of radius r for $x \leq r \leq 2a - x$ is

$$d\epsilon = \frac{\mu\ \epsilon_o}{4(a - x)}\ (2a - x - r)(1 + \frac{x}{r})e^{-\mu r}\ dr$$

and the total dose rate at P per unit volume due to all shells having portions in the emitting medium is therefore

$$\epsilon = \int_o^x \mu\ \epsilon_o e^{-\mu r}\ dr \qquad (+)$$

$$+ \int_{x}^{2a - x} \frac{\mu \ \epsilon_o}{4(a - x)} \ (2a - x - r)(1 + \frac{x}{r})e^{-\mu r} \ dr. \qquad (2)$$

The integration is straightforward and results in the expression

$$\frac{4 \epsilon}{\mu \epsilon_o} = \frac{4}{\mu} - \frac{2}{\mu} e^{-\mu x}$$
$$+ \left[\frac{1}{\mu^2 (a - x)} + \frac{x}{\mu (a - x)}\right] \cdot \left[e^{-\mu (2a - x)} - e^{-\mu x}\right]$$
$$+ \frac{x \ (2a - x)}{a - x} \left\{ \ln \frac{2a - x}{x} + \sum_{n=1}^{\infty} \frac{(-1)^n \ \mu^n}{n \ n!} \left[(2a - x)^n - x^n\right] \right\} \qquad (3)$$

which gives the dose rate ϵ per unit volume as a function of the distance $a - x$ from the center of the sphere.

Our main interest, however, lies in the total dose rate E for the whole sphere. Now, E is given by

$$E = \int_{o}^{a} 4 \pi \epsilon (a - x)^2 \ d(a - x).$$

This integration is analytically tractable but lengthy. The result is

$$E = 4 \pi \ a^3 \ \epsilon_o \sum_{n=o}^{\infty} \frac{(-1)^n (n + 3)}{(n + 4)!} \ (2 \mu a)^{n+1}.$$

Defining G to be the fraction of the emitted beta energy that is absorbed by the sphere of gas and writing α for $2 \mu a$, we have

$$G = 3 \sum_{n=o}^{\infty} \frac{(-1)^n (n + 3)}{(n + 4)!} \alpha^{n+1} . \qquad (4)$$

For computational purposes, this infinite series converges too slowly for large α. For α greater than unity an alternative expression (which is valid for all values of α) is more convenient:

$$G = 1 - \frac{3}{2\alpha} + \frac{3}{\alpha^3} \left[1 - (\alpha + 1) e^{-\alpha} \right] . \qquad (4')$$

A graph of $G(\alpha)$ appears in Figure 2. (A tabulation of $G(\alpha)$ is given in Table 2, Chapter 6.)

The calculation of the energy absorption neglecting backscattering is thus formally complete. It may be noted that α varies with the composition and density of the absorbing gas as well as with the diameter of the emitting volume. If ρ is the gas density, μ/ρ is a constant for a given gas; and therefore α varies directly as the product of the gas density and the diameter of the emitting volume.

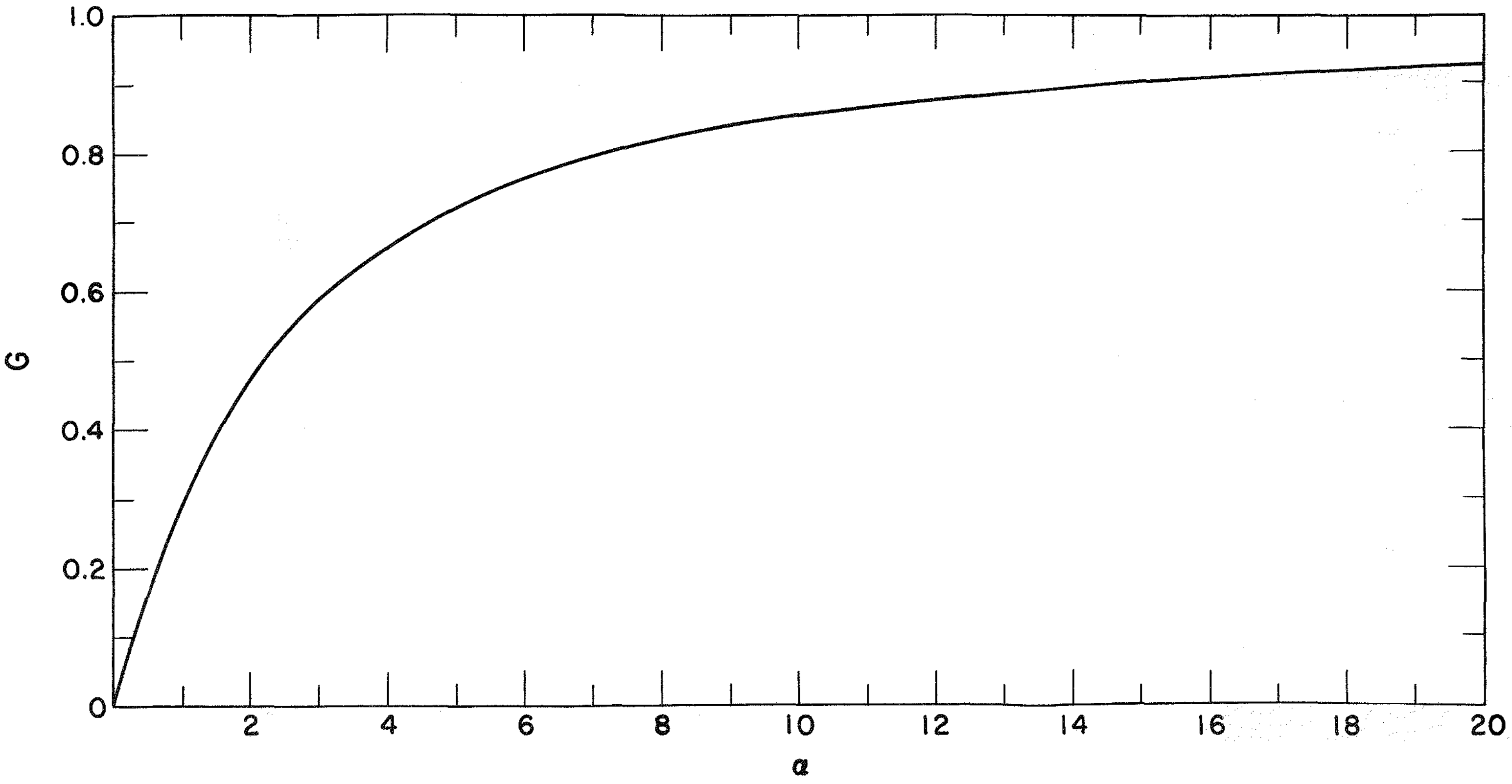

Fig. 2. The geometric absorption fraction G as a function of α.

Chapter 3

BACKSCATTERING CONSIDERATIONS

When an electron beam is incident on a relatively dense medium, an appreciable fraction of the beam is (more or less diffusely) scattered back into the half-space of its origin. This fraction is called the 2π (or half-space) backscattering coefficient. Typically, the backscattered electrons penetrate the medium for a distance varying from about one-twentieth to one-half of their rectilinear range in the medium before being turned back.

Generally considered, backscattering is a vastly complicated phenomenon and considerable experimental effort has been expended toward its elucidation. Much of this work is beyond the scope and interest of this report. No monograph or book seems to have been written on the subject and the literature is rather dispersed. However, the references cited in this report can serve as a guide to the main body of literature.

Accurate experimental work on backscattering is as rare as it is difficult. By far the largest amount of data is of a crude and exploratory nature. In the energy range of interest here only the work of Palluel,[3] Sternglass,[4] and Kanter[5] can be considered definitive.

The magnitude of the backscattering coefficient increases markedly with the atomic number of the backscattering medium[3-8] and also depends on the angle of incidence[5-7] of the electrons and on the angular aperture of the counter or collector.[7] Because of the angle effects, it makes considerable difference whether the incident electron flux is collimated or diffuse.[9] But, in any case, the directional distribution of the backscattered electrons is more characteristic of diffusive than of specular reflection.

On the other hand, the backscattering coefficient is relatively insensitive to changes in electron energy above about 3 kev.[4] In particular, for atomic numbers of less than about thirty, no energy dependence is evident above about 3 kev. For high atomic numbers, the backscattering coefficient is by no means small. For example, it is about 0.7 for diffuse incidence on lead[9] in the Mev energy region.

So far we have only been discussing particle backscattering without any concern about the energy distribution of the backscattered electrons. However, our interest in the backscattering process arises from our need to know the energy "reflected" back into the sphere of absorbing gas. The energy distribution depends on the atomic number of the backscattering medium and on the angle of incidence.[4-8] The shape of the distribution is practically independent of the energy of the incident electrons for low energies[4] ($\sim$ 1 kev), but exhibits some dependence at high energies[8] ($\sim$ 500 kev). Moreover, the dependence of the distribution on atomic number is much larger at high energies than at low.[4]

The ratio of the mean energy of the backscattered electrons to the mean energy of the incident electrons is, in the region of low energy, essentially independent of the incident energy and only slightly dependent on the atomic number. This ratio varies from about 0.5 for low atomic numbers to about 0.6 for high atomic numbers in the energy region from about 1 to 10 kev.[4,5] Use will be made of this fact in our computation of the backscattering contribution.

As might be expected, the presently available data on backscattering processes--even though fairly extensive--are by no means sufficient to make possible a precise calculation of the energy deposition in the sphere of gas due to the backscattered electrons. A precise calculation would demand detailed data on the particle flux, directional distribution, and energy distribution of the beta particles incident on the wall, and would also require detailed data on the backscattering coefficient per element of solid angle as a function of atomic number, (incident and reflected) direction, and energy, and, in addition, would require detailed data on the energy per backscattered electron as a function of the above parameters. Moreover, especially at low values of the absorption parameter α (discussed in the last section), second-order backscattering (i.e., backscattering of the backscattered electrons after traversing the gas) and even third-order backscattering must be taken into account.

It is therefore apparent that, even if accurate data were available in profusion, the precise calculation of the backscattering contribution to the energy absorption in the gas would be a most formidable task. Indeed, because of the geometrical complexities involved, the task might well be practically impossible.

For these reasons we here proffer a rather crude calculation as a substitute for exact analysis. In a later section we shall see that, in spite of the severe approximations, some empirical evidence suggests that the calculation is not grossly in error even for wall materials of atomic number as high as silver. Because the relative backscattering contribution diminishes with the atomic number, this calculation should be sufficiently accurate for wall materials of moderately low atomic number.

Our approach will involve the following basic assumptions:

a. Each surface element of the spherical wall emits backscattered beta particles isotropically into the half-space of the absorbing gas.

b. The average energy of the once-backscattered beta particles is degraded by an amount such that the average absorption coefficient is $\gamma_1\mu$, where $\gamma_1 > 1$ and μ is the average absorption coefficient of the unbackscattered beta particles in the gas.

c. The total energy of the once-backscattered beta particles is $4/3\pi a^3 \epsilon_o b(1 - G)$ so that the relative (to the total beta energy emitted in the sphere of gas) energy is $b(1 - G)$, where $b < 1$ is assumed to be a constant depending only on the atomic number of the wall material. $(1 - G)$ is the fraction of the total emitted beta energy that is "incident" on the spherical wall.

d. Successive backscattering can be taken into account by successive application of the considerations given in (a), (b), and (c).

In accordance with (a) and (b) and using the same geometry as in Fig. 1, save that $x = 0$, we find the energy absorbed per second in a spherical shell of the absorbing gas due to an element of surface area on the wall of emission strength ϵ_o' to be

$$d\epsilon' = \gamma_1 \mu \epsilon_o' \frac{2a - r}{2a} e^{-\gamma_1 \mu r} dr.$$

Now, the total energy absorbed per second by all of the gas is just

$$\epsilon' = \int_0^{2a} d\epsilon' = \epsilon_o' \left[1 - \frac{1}{\gamma_1 \alpha} (1 - e^{-\gamma_1 \alpha}) \right]$$

where, as before, $\alpha \equiv 2\mu a$.

Since the same relation holds for every element of wall area, the total dose rate in the absorbing gas due to once-backscattered electrons is

$$\frac{4}{3}\pi a^3 \epsilon_o b(1 - G) \left[1 - \frac{1}{\gamma_1 \alpha} (1 - e^{-\gamma_1 \alpha}) \right]$$

and the dose rate relative to the total beta energy emitted in the medium is

$$B_1 = b(1 - G)f_1$$

where $f_1 \equiv 1 - \dfrac{1}{\gamma_1 \alpha} (1 - e^{-\gamma_1 \alpha})$.

It is evident that not all of the backscattered beta particles will be absorbed by the gas: a fraction $1 - f_1$ of the original $b(1 - G)$ energy will be left over to "reimpinge" on the wall. The resultant, twice-backscattered beta particles will thus carry a total relative energy amounting to $b^2(1 - G)(1 - f_1)$ and will have an average absorption coefficient $\gamma_2\mu$ where $\gamma_2 > \gamma_1$. Of this energy, a fraction $f_2 \equiv 1 - (1/\gamma_2\alpha)\ (1 - e^{-\gamma_2\alpha})$ will be absorbed by the gas. Thus, $B_2 = b^2(1 - G)(1 - f_1)\ f_2$.

Similarly, the dose rate in the gas due to the thrice-backscattered beta particles will be $B_3 = b^3(1 - G)(1 - f_1)(1 - f_2)\ f_3$, where $\gamma_3 > \gamma_2$, and $f_3 \equiv 1 - (1/\gamma_3\alpha)(1 - e^{-\gamma_3\alpha})$.

In general, the total relative dose rate due to backscattering of all orders is $B = \sum_{i=1}^{\infty} B_i$. In practice, where α is not smaller than about 0.1 in the region of interest, B may be taken as $B_1 + B_2 + B_3$. For α greater than about 20, only B_1 need be computed. A graph of $f(\gamma\alpha)$ vs $\gamma\alpha$ appears in Fig. 3.

At this point the formal part of our calculation is complete. The fraction of the total beta emission energy that is absorbed by the spherical volume of gas is $F \equiv G + B$. While G depends only on α, B depends (through b) on the atomic number of the wall material as well as on α. Our next task will be to assign numerical values to b, γ_i, and μ. This will be undertaken in the following chapter.

First, however, comments on the nature of b may be worthwhile. b is the fraction of the total beta energy

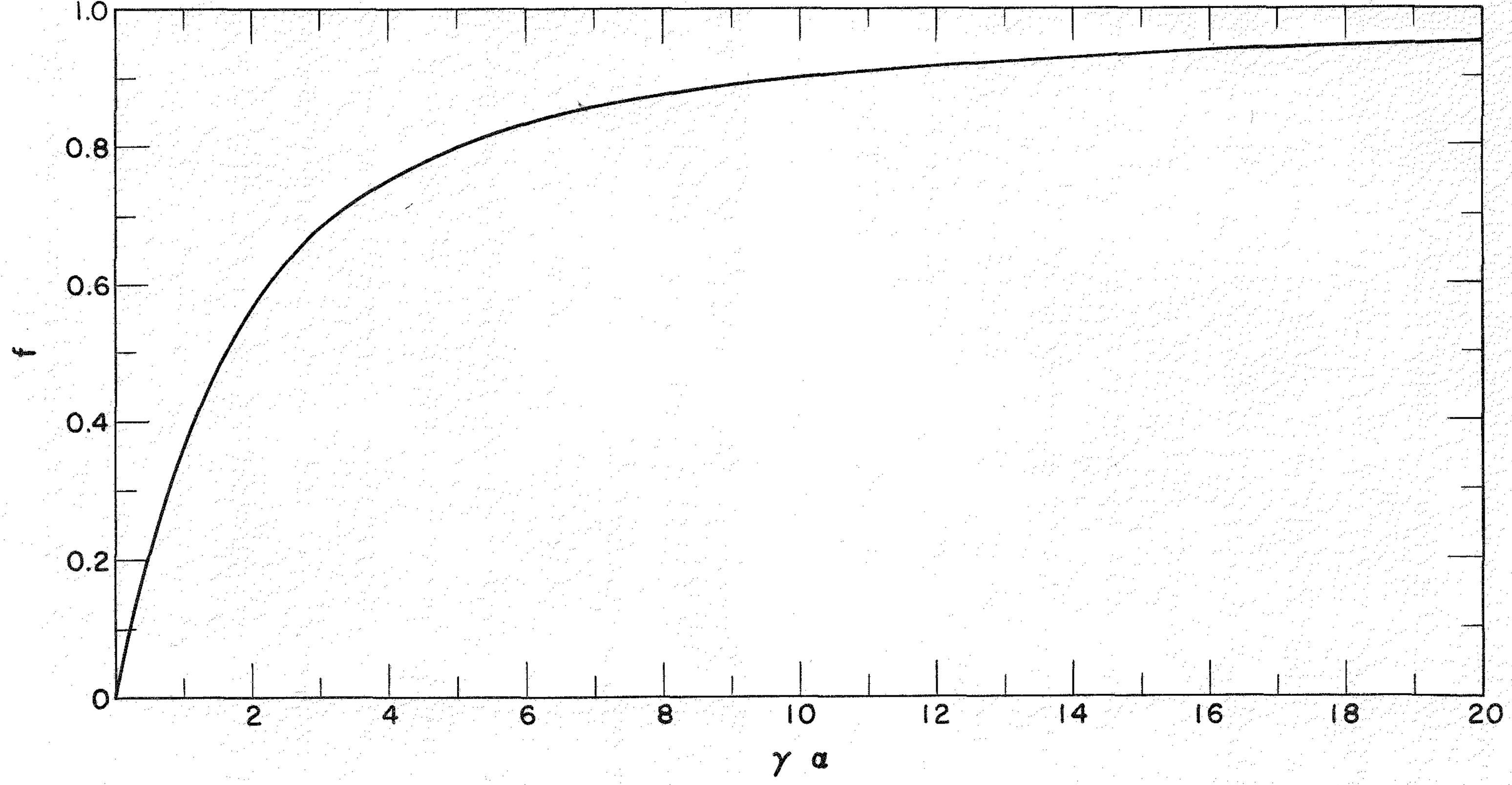

Fig. 3. The backscattering absorption fraction f as a function of $\gamma\alpha$.

"incident" on the wall that is backscattered. Hence, b is given by the product of the particle backscattering coefficient (averaged over all angles and, if necessary, over all incident energies) and the average energy of the backscattered beta particles relative to the average energy of the emitted beta particles. Now, the average energy of the backscattered beta particles is, in turn, given by the product of the average energy of the beta particles <u>incident</u> on the wall and the average fraction of the incident energy retained by the particles during backscattering. In our range of energy, this latter fraction is independent of the incident beta particle energy and only slightly dependent on the atomic number of the backscattering material.[4,5]

However, it is not difficult to see that the average energy of the beta particles incident on the wall depends somewhat on the α of the sphere of gas. For extremely small α, the average energy of the incident beta particles should be close to the average energy of the beta emission. For very large α, the average energy of the incident betas should be in the neighborhood of one-half of the average emission energy. The actual dependence on α would be rather difficult to calculate and will not be attempted here. Even an approximate calculation of the dependence of the energy distribution on depth of penetration in a medium is a rather formidable task.[10] Furthermore, the uncertainties in the other assumptions are probably as large as the error introduced by assuming b to be constant with α over the range of α we are concerned with (typically, from 1 to 20). In this range, we estimate that the average energy of the incident beta particles is roughly 0.7 of the average energy (5.7 kev) of the beta emission from tritium.

Chapter 4

NUMERICAL ASSIGNMENTS

For the sake of continuity with the discussion of the last section, we shall consider the b values first. For the values of the backscattering coefficients, we shall take values from the curve given by Kanter[11] for the energy interval from 3 to 10 kev. The agreement between these values and earlier values reported in the literature is fairly good. From Sternglass,[4] we choose the value 0.6 for the average fraction of the energy retained by the electrons during the backscattering process. And, for reasons mentioned in the previous chapter, we choose 0.7 as the average energy of the incident beta particles relative to the average emission energy.

Therefore, b is given by b = 0.6 (0.7) (backscattering coefficient). For example, for silver, b = 0.6 (0.7) (0.38) = 0.16. Now, it has been shown[12] that insulators backscatter like metal if the average atomic number is used. Thus, for borosilicate glass we calculate $\overline{Z} = 9.5$ and use this value to get a backscattering coefficient of about 0.107 from the curve. Therefore, for borosilicate glass, $b = 0.6\ (0.7)\ (0.107) = 0.045$.

Values of b calculated by this procedure for seven atomic numbers are listed below.

Substance	Z	b
Be	4	0.02
C	6	0.03
glass	9.5	0.045
Al	13	0.055
Ni	28	0.11
Ag	47	0.16
Au	79	0.18

Next, we seek numerical values for γ_1, γ_2, and γ_3. Fortunately, because of compensating effects, the accuracy of these numerical assignments need not be great. This is especially true for γ_2 and γ_3.

There is some evidence[1] that the mass absorption coefficient $k \equiv \mu/\rho$, where ρ is the material density, is given (at least to a fair approximation) by $k = \eta/\epsilon$, where η is the mean energy loss rate (mean stopping power) of the beta particles and ϵ is the mean energy of the beta particles. This relation has only been checked for mean energies equal to or greater than 49 kev, the mean energy of S^{35} beta particles. We shall make the assumption that it is also valid down to about 1 kev.

As noted in the previous chapter, the mean energy of once-backscattered tritium beta particles is about 0.6 (0.7)(5.7 kev) = 2.4 kev; that is, about 0.4 of the mean beta emission energy.

To compute γ_1, we use the well-verified Bethe-Bloch theory with the mean excitation potential values taken from Nelms[13] to calculate η at an electron energy of 2.4 kev. Now, considered strictly, the mean energy loss rate is not equal to the energy loss rate calculated at the mean beta energy. Moreover, the mean backscattered beta particle energy <u>in the gas</u> is not equal to the mean energy of the backscattered beta particles. However, since we shall only be dealing with ratios, one can show that the error introduced by these simplifying procedures is small compared with the uncertainty probably inherent in the basic assumptions.

Now, by definition, $\gamma_1 = k_1/k$ and, in turn, $k_1/k = \eta_1/\eta \cdot \epsilon/\epsilon_1$. Therefore, $\gamma_1 = 2.38\ \eta_1/\eta$. Fortunately for simplicity, the calculated ratio η_1/η varies only slightly with the nature of the gas. We take 1.9 as a typical value. This gives $\gamma_1 = 4.5$, approximately, for all gases.

Two items of experimental information lend credence to this method of calculation. In aluminum, range-energy measurements[14,15] show that the Bethe-Bloch theory is valid down to at least 1 kev. In aluminum, again, a study[16] of the rate of secondary electron formation indicates indirectly that $k_1/k = 4.9$, approximately, for once-backscattered electrons.

Applying this method of calculation to γ_2, we find $\gamma_2 = k_2/k = \eta_2/\eta \cdot \epsilon/\epsilon_2 = 6.3\ \eta_2/\eta$, and since η_2/η equals about 4.0 for all gases, $\gamma_2 = 25$.

For γ_3 we obtain, similarly, $\gamma_3 = 17\ \eta_3/\eta = 140$ for all gases.

Using these values of γ_i and taking $b = 0.16$, the energy absorption B due to backscattering of all orders from a silver wall may be calculated. The result appears in Fig. 4. Note the surprisingly sharp peak at about $\alpha = 0.9$. Parenthetically, we may remark that this value of α corresponds to about 60 mm Hg of H_2 gas at 25°C in a bulb of 100 ml volume. The shape of the curve $B(\alpha)$ is approximately the same for all wall materials and the ordinates are roughly proportional to the respective b-values.

The most important numerical assignment yet remains--that of the μ values. From Chapter 2 we recall that if the linear absorption coefficient μ is known for the absorbing

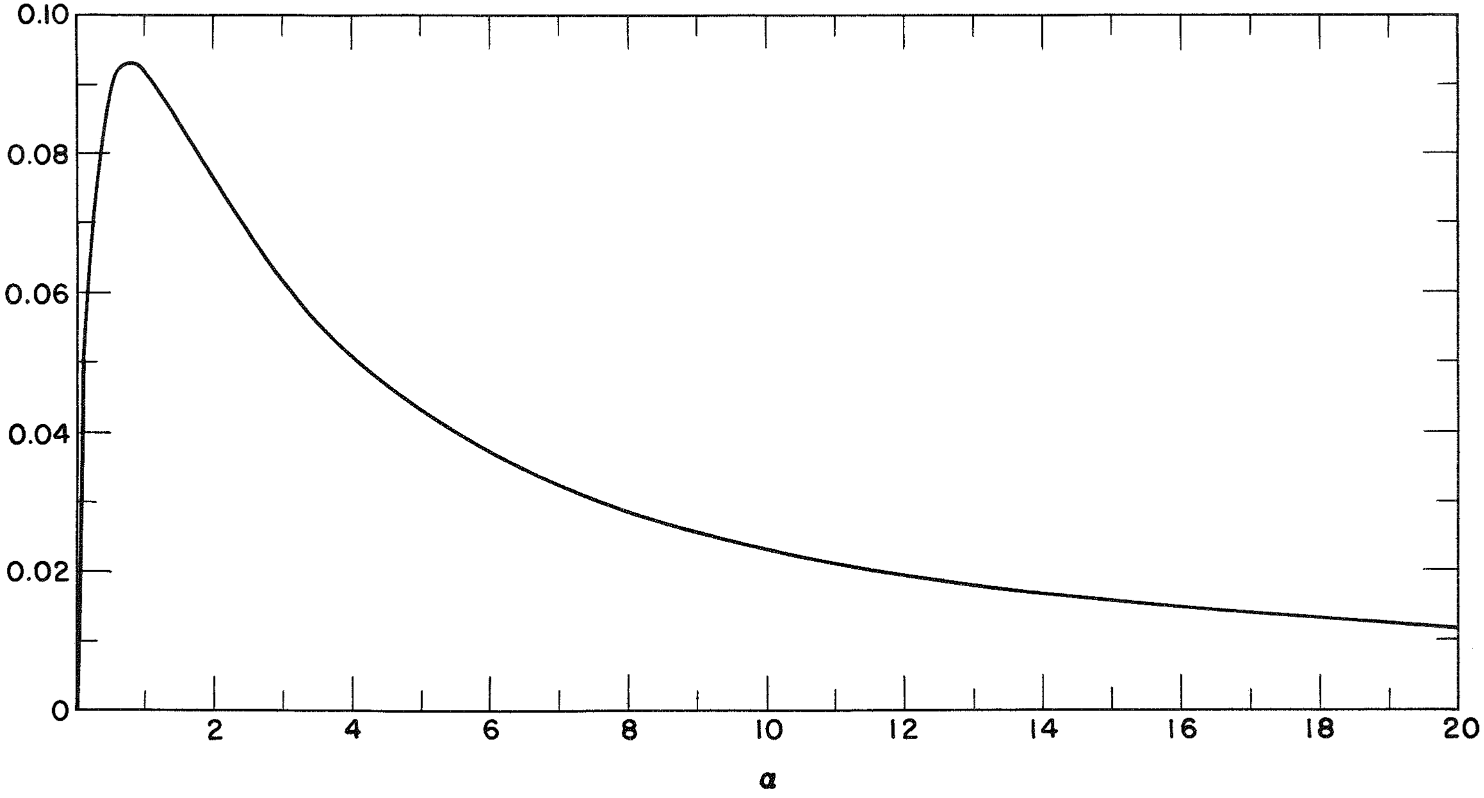

Fig. 4. The backscattering contribution B for a silver wall as a function of α.

gas, α and thence $G(\alpha)$ can be computed. We have just shown how $B(\alpha)$ can be computed for a specified wall material if α is known. Hence, if μ is known, the problem of calculating $F = G + B$ (the fraction of the total tritium beta emission energy absorbed by the spherical volume of gas) is solved.

Our approach to the calculation of μ will be to use the relation of Sommermeyer[1] referred to above, $k \equiv \mu/\rho = \eta/\epsilon$, and compute k for tritium beta particles from Sommermeyer's measurements[1] of k for S^{35} beta particles.

From the most recent and most precise measurement of the heat output of tritium[17] and the end point (18.6 kev) of the tritium beta spectrum,[18] the mean beta emission energy of tritium is calculated to be 5.7 kev. The mean beta emission energy of S^{35} is 49 kev.[19] Hence,

$$\frac{k_{5.7}}{k_{49}} = \frac{49}{5.7} \cdot \frac{\eta_{5.7}}{\eta_{49}} = 8.60\,\frac{\eta_{5.7}}{\eta_{49}}.$$

For hydrogen gas, the η ratio computed by means of the Bethe-Bloch theory is 5.50. Therefore, for H_2, $k_{5.7}/k_{49} = 47.3$. Now, Sommermeyer[1] finds for H_2 a k-value of 0.465 cm^2/mg for S^{35} beta particles. Thus, we calculate for tritium beta particles in hydrogen gas that $k = 22.0\ cm^2/mg$.

Since Sommermeyer does not give an estimate of the uncertainty in his measurement of k for S^{35} beta particles, and since it is difficult to estimate the uncertainty inherent in the assumptions by which the k for tritium beta particles is calculated from Sommermeyer's measurement, no concrete assessment of the expected error can be given. However, for what it is worth, we believe that the actual value of k for tritium beta particles in hydrogen gas is likely to lie between 21 and 23 cm^2/mg. In the next chapter we shall see that some empirical evidence tends to support this contention. In any case, we shall take $k = 22.0$ cm^2/mg for H_2 as our point of departure from which the k values of other substances will be calculated.

Sommermeyer[1] also made measurements on air and argon traversed by S^{35} beta particles. These k values are 0.204 and 0.193 cm^2/mg, respectively. By the procedure just outlined, we calculate the corresponding k values for tritium beta particles to be 8.8 and 7.8 cm^2/mg.

Using the Bethe-Bloch theory with the values of the mean excitation potentials taken from Nelms[13] to calculate η for various substances at an electron energy of 5.7 kev, we may then calculate k_1 for various substances by using the relation, $k_1/k_H = \eta/\eta_{H_2}$. Under the assumption that $k_H =$ 22.0 cm^2/mg, k_1 values from the Bethe-Bloch theory have been calculated for fourteen substances and are listed in the second column of Table 1.

TABLE 1

ABSORPTION COEFFICIENTS FOR TRITIUM BETA PARTICLES

Substance (Z)	k_1 $\left(\frac{cm^2}{mg}\right)$	k_2 $\left(\frac{cm^2}{mg}\right)$	k $\left(\frac{cm^2}{mg}\right)$	ρ at 25°C 760 mm $\left(\frac{mg}{ml}\right)$	μ at 25°C 760 mm (cm^{-1})
H_2 (1)	22.0	22.0	22.0	0.0824	1.81
He (2)	9.47	11.1	10.3	0.1635	1.68
C (6)	8.41	11.1	9.75	--	--
N_2 (7)	8.11	11.1	9.60	1.146	11.0
O_2 (8)	7.86	11.1	9.48	1.309	12.4
Ne (10)	7.38	11.0	9.19	0.8249	7.59
S (16)	6.60	11.1	8.85	--	--
A (18)	5.75	10.0	7.87	1.634	12.9
Kr (36)	4.22	9.52	6.87	3.397	23.4
Xe (54)	3.37	9.11	6.24	5.360	33.4
CH_4	11.8	13.8	12.8	0.6567	8.40
CO_2	8.00	11.1	9.55	1.811	17.3
Air	8.00	11.1	9.55	1.185	11.3
H_2S	7.52	11.7	9.61	1.410	13.5

Comparison of these k_1 values with those calculated from the measurements of Sommermeyer on air and argon shows that the corresponding k_1 values are about 10% low for air and about 25% low for argon.

The explanation for this discrepancy may possibly be found in some recent experimental work[11,20] in which the so-called practical range of electrons was studied in the kev energy region. It was established that, for electrons of energy less than about 7 kev, the practical range expressed in mass per unit area depends only on A/Z (the ratio of atomic mass to atomic number) and thus varies only slightly with atomic number for elements above hydrogen. (Six elements varying in atomic number from carbon to gold were studied.)

Now, in this region of energy the range is approximately inversely proportional to the energy loss rate so that we are led to posit the relation, $k_2/k_H = (Z/A)/(Z/A)_H$. Assuming, as before, that $k_H = 22.0$ cm^2/mg, we have then $k_2 = (22.0/0.992)\ (Z/A)$ for the various substances. Values of k_2 thus computed appear in the third column of Table 1. Comparison with the k values for air and argon computed from the measurements of Sommermeyer shows that the k_2 values are too high by about 26% and 28%, respectively.

The reason for this discrepancy is not difficult to fathom: the Z/A relation is valid only up to about 7 kev, but an appreciable fraction of the tritium beta emission lies above 7 kev. Because of the uncertainty in the experimental values of Sommermeyer and because of the uncertainty in the method of calculation of the k values therefrom, it does not seem worthwhile to attempt to "blend" the k_1 and k_2 calculations in a physically meaningful manner to arrive at agreement with the values calculated from Sommermeyer's data. We are therefore led to the expedient compromise of taking as k the mean of the k_1 and k_2 values for a given substance. These expedient k values, which we shall use in all future calculations, are displayed in the fourth column of Table 1.

In passing, it may be of interest to note that the k values of compounds and mixtures are calculated by means of atomic mass ratios applied to the constituent atoms. Using methane as an illustration, we see that

$$k_{CH_4} = \frac{A_c}{A_{CH_4}} k_c + \frac{4A_H}{A_{CH_4}} k_H .$$

This procedure is based on the assumption that the energy loss rate of a compound is additively made up of the energy loss rates of its constituent elements. Measurements with protons[21] show that this assumption is valid down to proton energies of roughly 200 kev. The corresponding electron energy is about 0.1 kev. Therefore the assumption should be sufficiently valid for tritium beta particles.

Comparison of these expedient k values with those calculated for air and argon from Sommermeyer's data shows agreement in the case of argon, but about 8% difference in the case of air. Considering the many uncertainties inherent in our calculations (not to mention possible experimental errors), we regard the agreement as satisfactory.

With this, the task of assigning numerical values to the parameters of the formal development is essentially complete. All that remains is to multiply the k values in column four by the gas density in mg/ml at 25°C, 760 mm Hg as given in column five in order to find the μ values at 25°C, 760 mm Hg for the twelve gases listed. These μ values are displayed in the sixth column of Table 1.

For a mixture of gases, it is easy to show that the effective linear absorption coefficient is given by $\Sigma_i \mu_i \upsilon_i$, where υ_i is the mole fraction of constituent i.

Isotopes of gases can also be treated simply. Since the energy loss rate expressed in energy per unit mass per unit area is proportional to f(Z)/A, it is clear that its variation among isotopes of a given element depends inversely on the variation of A. Hence, for a given element, k varies inversely as the isotopic mass. However, $\mu = k\rho$ and ρ varies directly as A. Therefore, we conclude that the linear absorption coefficients of all isotopes of a given element are identical. Taking tritium as an example, we see that its k value is (22.0/3) (cm^2/mg), but its μ value is the same as that of protium.

Finally, some cautionary comments: the μ values given in Table 1 have been calculated from data of unknown accuracy by an involved series of assumptions required to effect the extrapolation into an energy region of a lower order of magnitude. It would indeed be surprising if they were not in error by a fair amount.

Furthermore, even if the μ values were accurate, considerable error might be found in the case of gases of high atomic number. The reason is that our basic premise--that the point-source energy absorption function is a pure exponential--might not be valid at high atomic numbers. Sommermeyer finds that S^{35} beta particles in argon exhibit only a slight departure from exponential, but in gold give a peaked absorption function. (Air and hydrogen give rather exact exponential functions.) From this we conclude that S^{35} beta particles in krypton and xenon may give significantly nonexponential absorption functions. However, this conclusion does not necessarily follow for tritium beta particles, which are an order of magnitude less energetic than S^{35} beta particles. Furthermore, consider the fact that in the case of P^{32} beta particles (which are an order of magnitude more energetic than S^{35} beta particles), no substance studied gives an exponential absorption function. In view of these relations, we tentatively surmise that the point-source absorption functions of tritium beta particles in krypton and xenon probably do not depart significantly from pure exponentials.

Chapter 5

COMPARISON WITH AN EXPERIMENT

We now have at hand all of the information--tentative though some of it may be--required to compute the fraction of the tritium emission energy absorbed by any kind of gas enclosed in a spherical vessel of any wall material. However, in view of the many assumptions inherent in the calculation, an experimental test is strongly indicated.

Apparently, only one piece of experimental work along these lines has been carried out. This is the work of Dorfman[22] who studied the absorption of tritium beta particles in hydrogen, helium, and oxygen by measuring the ionization current generated in spherical glass bulb ionization chambers with silvered walls. The reported measurements were carried out in an ionization chamber of 5.70 cm internal diameter ($\sim$100 ml volume) at H_2 gas pressures varying from about 10 to 630 mm Hg. Data for He were taken with fewer points over a comparable range. The O_2 data will not be considered here because, as the author pointed out, the precision is poor.

The principle of the method for determining the spherical-vessel absorption function is to measure the ionization currents (at saturation voltage) as a function of gas pressure and divide these currents by the current measured at "saturation" pressure; that is, at a practically infinite pressure so that essentially all of the beta energy is absorbed. This procedure thus gives the fraction absorbed as a function of pressure for a particular gas at a given temperature in a given vessel.

From the previously developed theory, let us compute the pressure of hydrogen gas at 25°C in a 5.7 cm diameter silvered bulb required for 99% energy absorption. At large values of α, the expressions may be considerably simplified so that we may, to a good approximation, write:

$$F = G + B = 1 - \frac{3}{2\alpha} + 0.16 \left(\frac{3}{2\alpha}\right)$$

$$= 1 - \frac{1.26}{\alpha}$$

Putting $F = 0.99$ and solving for α, we obtain $\alpha = 126$. This implies that, in a 5.7 cm diameter bulb, the pressure is 12.2 atm. Similarly, putting $F = 0.97$, we find that the pressure is 4.1 atm. It is therefore clear that, in a 100 ml bulb, relatively large pressures of hydrogen gas are required for essentially complete energy absorption.

The saturation pressure chosen for the aforementioned experiment appears, in the light of our calculations, to have been too low. Unfortunately, neither the raw data nor the saturation pressure used in computing the published curves is available at the present time.[23] However, the author indicates[23] that the saturation pressure was taken at about the end of the curve in graph #KH-9A3057.[24] This would indicate a saturation pressure of roughly 900 mm Hg of hydrogen gas at 25°C. Using this value, we calculate from the theory that $F = 0.90$ for hydrogen in a 100 ml silvered bulb. This suggests that the values published in reference 22 of the fractional energy absorption should be multiplied by about 0.90.

Because of the use of too low a saturation pressure, the plot of $\log (1 - F')$, where F' is the incorrectly large absorption fraction, versus gas pressure led to a straight line over the major portion of the pressure range. This led to the incorrect assumption that (neglecting backscattering) the spherical-vessel absorption function could be represented by an exponential function. If any other saturation pressure (except in the neighborhood of the one chosen) had been used, no portion of the curve would have approximated a straight line on a semilogarithmic plot.

Moreover, the starting equation and attendant nomenclature in reference 22 refer to particle attenuation in absorbers and not to energy absorption. It is a coincidence that both may be expressed as an exponential function though, albeit with numerically different absorption coefficients.

Dorfman attributes the marked departure of the data from linearity on the semilogarithmic plot for pressures less than about 250 mm Hg to backscattering. Our later analysis shows that only about one-half of this deviation is due to backscattering; the remainder is a geometrical effect characteristic of a spherical absorption geometry. Thus, the experimenter's procedure of reading the pressure-for-half-absorption from his straight line appears incorrect on two counts: F' is about 11% too large, and using the straight line considerably overcorrects for backscattering. But, oddly enough, the errors approximately compensate so that our procedure gives approximately the same result.

The experimenter's procedure for calculating the mass absorption coefficients from the pressure-for-half-absorption values appears to be incorrect since it presupposes that the spherical-vessel absorption function is a pure exponential. There seems to be no valid way to calculate absorption coefficients from observed data without carrying through an analysis of the type presented in this report. However, surprisingly Dorfman's method of calculating k values from his data gives, because of compensating errors, approximately the same numerical results as does our analysis.

The solid line in Fig. 5 represents the theoretical energy absorption function for hydrogen gas at 25°C in a silvered spherical bulb of 5.7 cm inside diameter. Since $\mu = 1.81 \text{ cm}^{-1}$, α is obtained by multiplying the pressure in mm Hg by 0.0136. Referring to the backscattering curve for silver given in Fig. 4, we see that the contribution to the total ionization due to backscattering is 51% of the total at 10 mm Hg of hydrogen, 27% at 60 mm Hg, and 2.5% at 760 mm Hg.

Dorfman's data for hydrogen and helium are also plotted in Fig. 5. For reasons explained above, the published values for the energy absorption function have all been multiplied by 0.90. The data points have been read, as accurately as possible, from the published graphs. Nevertheless, some error is undoubtedly introduced by this procedure.

Except for pressures under 30 mm Hg, the agreement between experiment and theory for hydrogen is within ± 3%. We interpret this as a verification of the theory for the case of hydrogen. Moreover, the agreement shows that our numerical assignments for b, γ_i, and μ for hydrogen gas enclosed in a silvered vessel cannot be grossly in error.

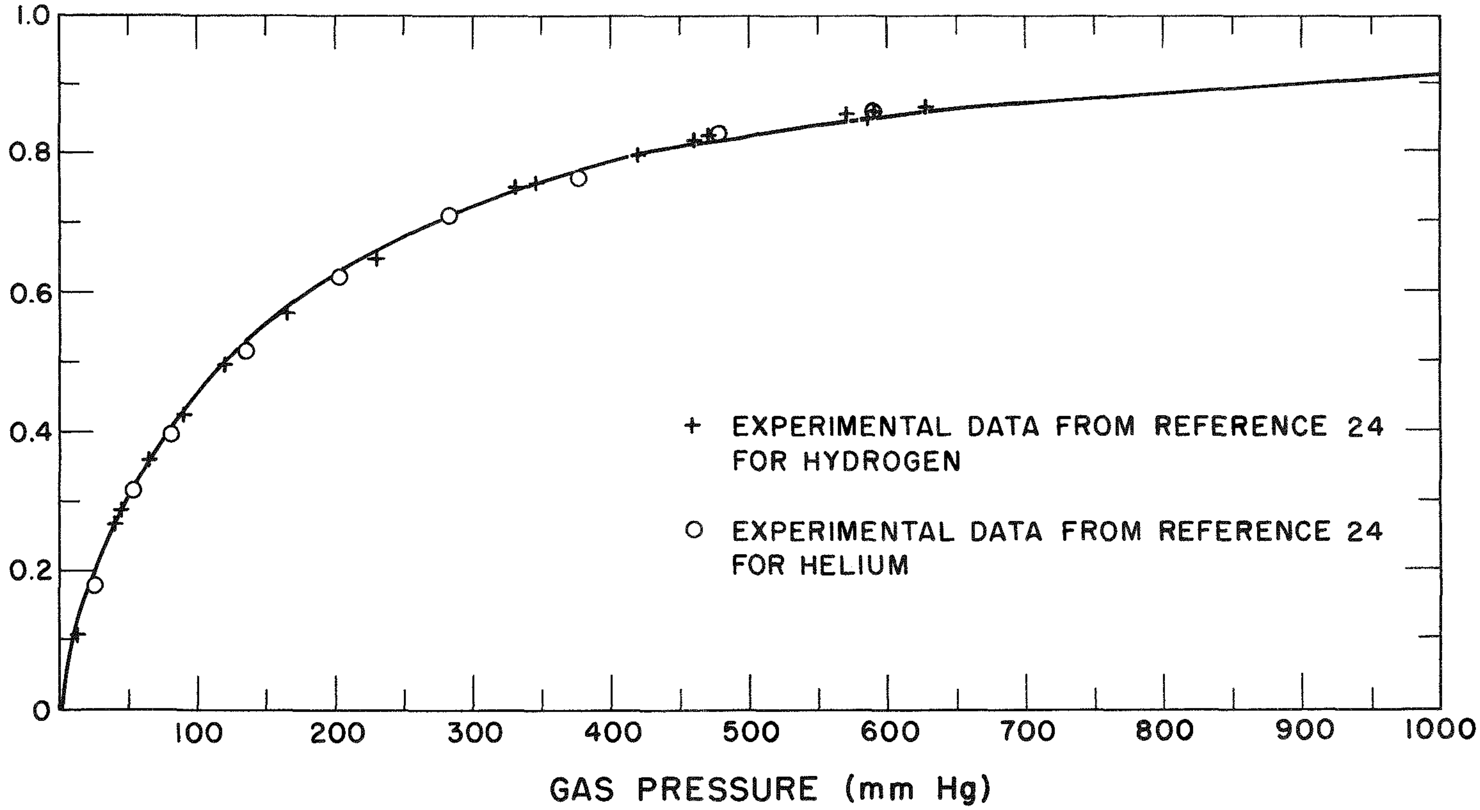

Fig. 5. The total absorption fraction F for hydrogen gas at 25°C in a 100 ml silvered bulb as a function of pressure.

For the case of helium gas the agreement is not as satisfactory. Dorfman's data seem to indicate that μ for helium is identical to μ for hydrogen, whereas our scheme of numerical assignment puts μ for He about 7% lower than μ for H_2. But, because of the uncertainty inherent in our semitheoretical estimate of μ, precise agreement could hardly be expected. However, it should also be borne in mind that, because of the effect of trace amounts of impurities, helium is well-known[25] to give erratic results in ionization experiments.

Chapter 6

GLASS BULBS

The case of the glass wall is of great practical importance since many studies of radiolysis, isotope exchange rates, and labeling are carried out in spherical glass reaction chambers.

Figure 6 exhibits the backscattering function $B(\alpha)$ for a spherical vessel of borosilicate glass, taking $b = 0.045$. Comparison with $B(\alpha)$ for silver (Fig. 4) shows that the shape is essentially the same, but the ordinates in the case of glass are only about one-fourth as large as the corresponding ordinates in the case of silver.

The calculated absorption function $F = G + B$ for spherical glass bulbs is displayed in Fig. 7. Numerical values are given in Table 2.

Since actual glass reaction chambers are usually made from round-bottom standard flasks, the geometry is not truly spherical. In most cases, however, the nonspherical portion

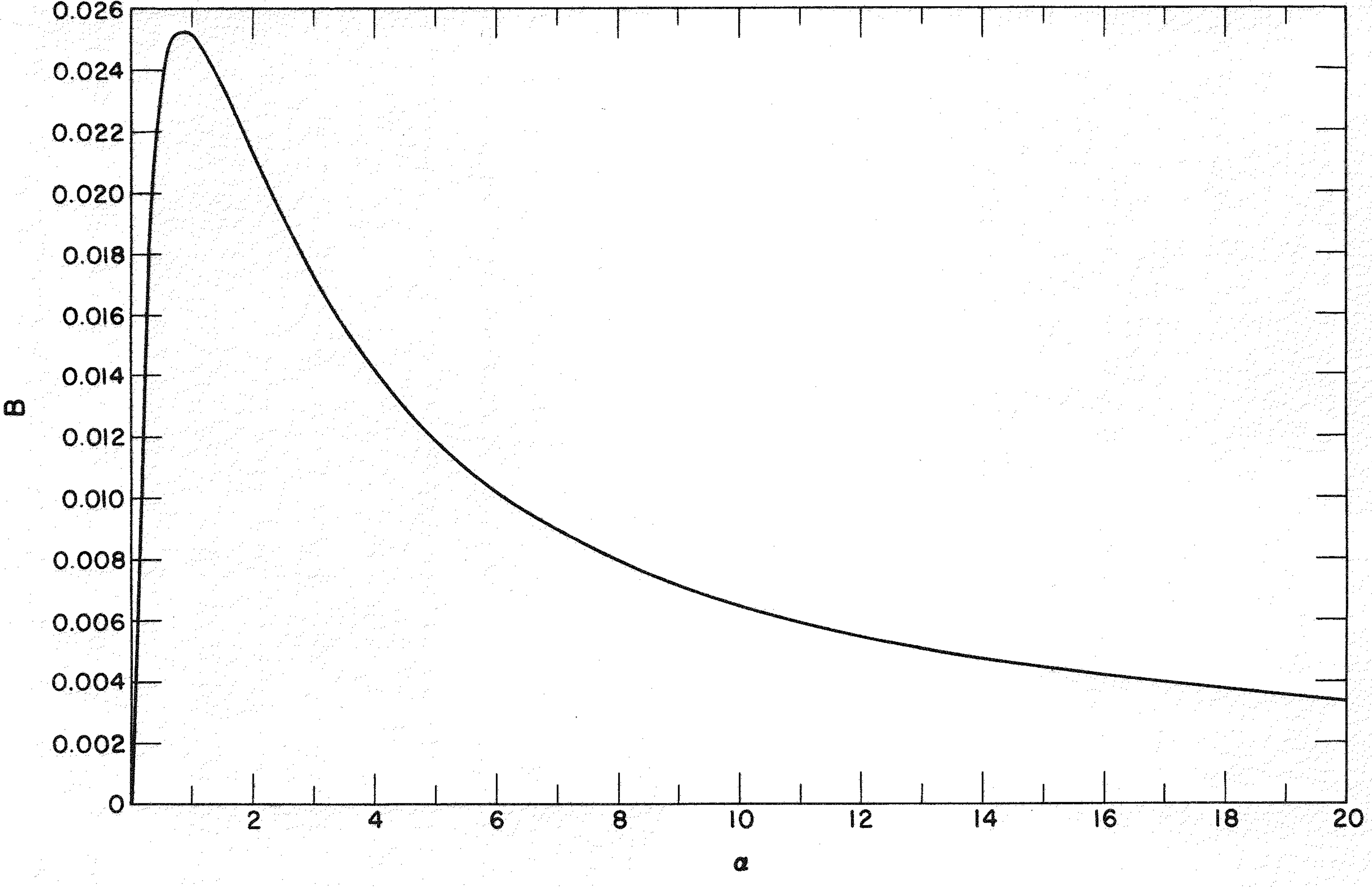

Fig. 6. The backscattering contribution B for a borosilicate glass wall as a function of α.

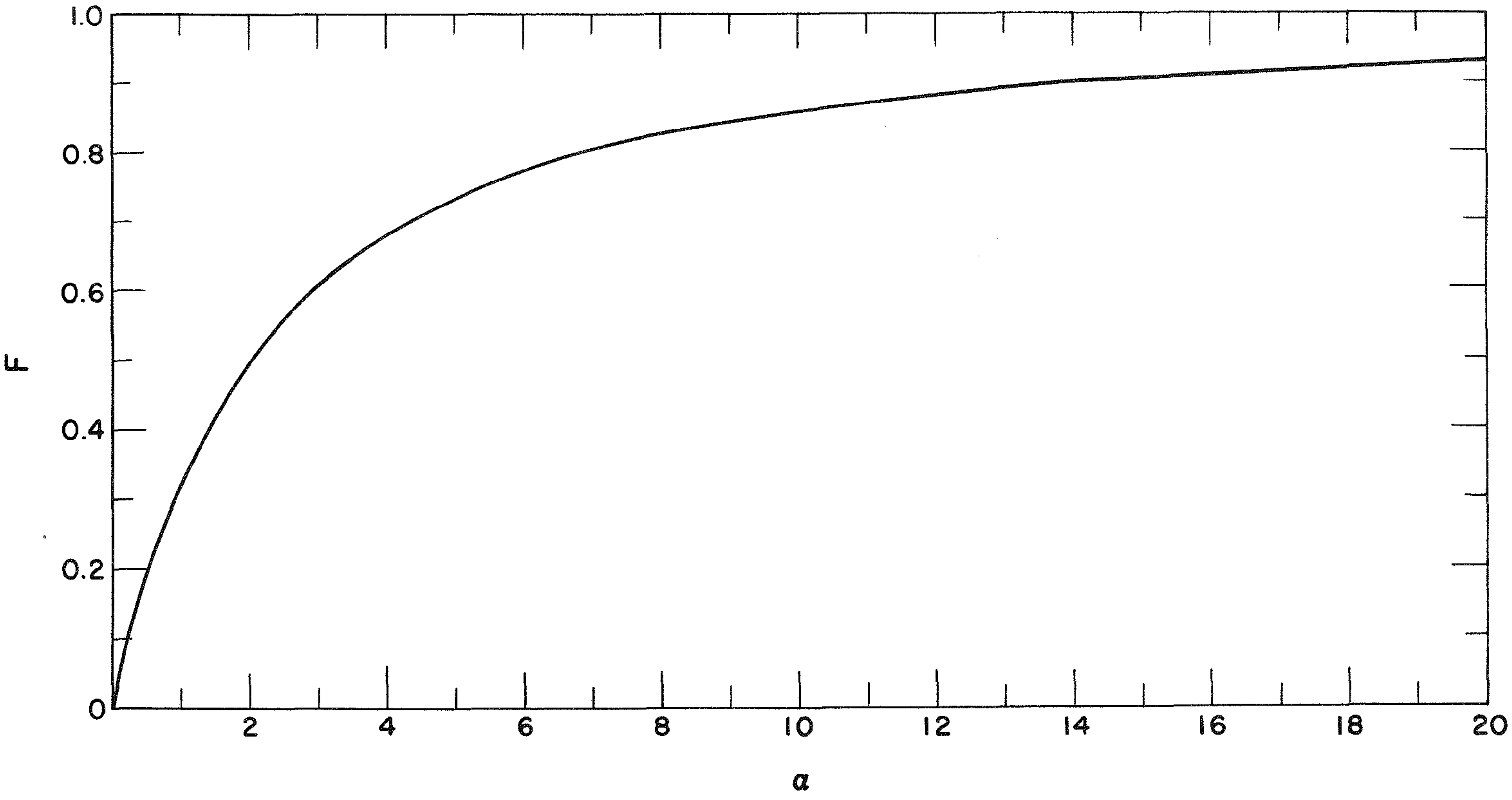

Fig. 7. The total absorption fraction F for borosilicate glass bulbs as a function of α.

TABLE 2

ABSORPTION FUNCTIONS FOR BOROSILICATE GLASS BULBS

α	G	B	F
0.1	0.0364	0.0095	0.0459
0.2	0.0712	0.0152	0.0864
0.3	0.1040	0.0191	0.1231
0.4	0.1352	0.0216	0.1568
0.5	0.1649	0.0233	0.1882
0.6	0.1931	0.0243	0.2174
0.7	0.2199	0.0249	0.2448
0.8	0.2454	0.0252	0.2705
0.9	0.2696	0.0252	0.2949
1.0	0.2927	0.0251	0.3179
1.1	0.3147	0.0249	0.3397
1.2	0.3357	0.0246	0.3603
1.3	0.3557	0.0243	0.3800
1.4	0.3748	0.0239	0.3987
1.5	0.3930	0.0234	0.4165
1.6	0.4105	0.0230	0.4335
1.7	0.4271	0.0226	0.4496
1.8	0.4430	0.0221	0.4651
1.9	0.4582	0.0217	0.4799
2.0	0.4727	0.0212	0.4940
2.1	0.4867	0.0208	0.5074
2.2	0.5000	0.0203	0.5203
2.3	0.5128	0.0199	0.5327
2.4	0.5251	0.0195	0.5446
2.5	0.5368	0.0191	0.5559
2.6	0.5481	0.0187	0.5668
2.7	0.5589	0.0183	0.5772
2.8	0.5694	0.0179	0.5873
2.9	0.5794	0.0175	0.5969
3.0	0.5890	0.0172	0.6062

TABLE 2 (Continued)

α	G	B	F
3.1	0.5982	0.0168	0.6151
3.2	0.6071	0.0165	0.6236
3.3	0.6157	0.0162	0.6319
3.4	0.6239	0.0159	0.6398
3.5	0.6319	0.0156	0.6475
3.6	0.6396	0.0153	0.6548
3.7	0.6469	0.0150	0.6619
3.8	0.6541	0.0147	0.6688
3.9	0.6609	0.0144	0.6754
4.0	0.6676	0.0142	0.6817
4.1	0.6740	0.0139	0.6879
4.2	0.6802	0.0137	0.6939
4.3	0.6862	0.0134	0.6996
4.4	0.6920	0.0132	0.7052
4.5	0.6976	0.0130	0.7105
4.6	0.7030	0.0127	0.7157
4.7	0.7082	0.0125	0.7208
4.8	0.7133	0.0123	0.7257
4.9	0.7183	0.0121	0.7304
5.0	0.7230	0.0119	0.7350
5.2	0.7321	0.0116	0.7437
5.4	0.7407	0.0112	0.7519
5.6	0.7488	0.0109	0.7597
5.8	0.7564	0.0106	0.7670
6.0	0.7636	0.0103	0.7739
6.2	0.7705	0.00998	0.7804
6.4	0.7769	0.00971	0.7866
6.6	0.7831	0.00945	0.7925
6.8	0.7889	0.00920	0.7981
7.0	0.7944	0.00897	0.8034
7.2	0.7997	0.00875	0.8084
7.4	0.8047	0.00854	0.8132
7.6	0.8094	0.00834	0.8178
7.8	0.8140	0.00814	0.8221
8.0	0.8183	0.00796	0.8263

TABLE 2 (Continued)

α	G	B	F
8.2	0.8225	0.00778	0.8303
8.4	0.8265	0.00761	0.8341
8.6	0.8303	0.00745	0.8377
8.8	0.8339	0.00729	0.8412
9.0	0.8374	0.00714	0.8446
9.2	0.8408	0.00700	0.8478
9.4	0.8440	0.00686	0.8509
9.6	0.8471	0.00673	0.8539
9.8	0.8501	0.00660	0.8567
10.0	0.8530	0.00647	0.8595
10.25	0.8564	0.00633	0.8628
10.50	0.8597	0.00618	0.8659
10.75	0.8629	0.00605	0.8689
11.00	0.8659	0.00592	0.8718
11.25	0.8688	0.00579	0.8746
11.50	0.8715	0.00567	0.8772
11.75	0.8742	0.00556	0.8797
12.00	0.8767	0.00545	0.8822
12.25	0.8792	0.00534	0.8845
12.50	0.8815	0.00524	0.8868
12.75	0.8838	0.00514	0.8889
13.00	0.8860	0.00505	0.8910
13.25	0.8881	0.00496	0.8930
13.50	0.8901	0.00487	0.8950
13.75	0.8921	0.00478	0.8968
14.00	0.8940	0.00470	0.8987
14.25	0.8958	0.00462	0.9004
14.50	0.8975	0.00454	0.9021
14.75	0.8992	0.00447	0.9037
15.00	0.9009	0.00440	0.9053
15.25	0.9025	0.00433	0.9068
15.50	0.9040	0.00426	0.9083
15.75	0.9055	0.00419	0.9097
16.00	0.9070	0.00413	0.9111
16.25	0.9084	0.00407	0.9125

TABLE 2 (Continued)

α	G	B	F
16.50	0.9098	0.00401	0.9138
16.75	0.9111	0.00395	0.9150
17.00	0.9124	0.00389	0.9163
17.25	0.9136	0.00384	0.9175
17.50	0.9148	0.00379	0.9186
17.75	0.9160	0.00373	0.9198
18.00	0.9172	0.00368	0.9209
18.25	0.9183	0.00363	0.9219
18.50	0.9194	0.00359	0.9230
18.75	0.9205	0.00354	0.9240
19.00	0.9215	0.00349	0.9250
19.25	0.9225	0.00345	0.9259
19.50	0.9235	0.00341	0.9269
19.75	0.9244	0.00336	0.9278
20.00	0.9254	0.00332	0.9287

(composed of the neck and possible tubulation ports) comprises less than 10% of the total reaction volume. A question therefore arises as to what effective spherical diameter should be used in calculating α.

Since an exact mathematical analysis for nonspherical geometries seems to be practically impossible, we can only proffer a rough rule of thumb: if the nonspherical portion of the total reaction volume amounts to about 10% or less, use the actual inside diameter of the spherical portion in calculating α. This procedure introduces only slight error (in the direction of too large a value for F) in the estimation of the fraction of the total emitted beta energy that is absorbed by the gas.

In order to obtain a rough estimate of the error inherent in this artifice, consider the mathematically tractable case of two spheres connected by a fill tube of negligible volume and filled with the same tritium and gas mixture. If the volume of the smaller sphere is 10% of the total volume, a simple calculation shows that the F for the system is about 95% of the F for the larger sphere if $\alpha < < 1$. The energy absorption fractions are practically identical if $\alpha > > 1$. Thus, depending on the value of α, the error introduced by taking F for the system to be identical with F for the larger sphere varies from zero to 5%. In the range of most interest, the error introduced is in the neighborhood of 1 to 2%. If the volume of the smaller sphere were only 1% of the reaction volume, the error introduced for $\alpha << 1$ would be less than 1%.

In all of the analysis presented in this report, homogeneous distribution of the tritium has been taken for granted. Under certain experimental conditions, however, it is possible that an appreciable portion of the tritium could be sorbed onto the wall. This effect would change the energy absorption function F through a change in both G and B. However, given sufficient information concerning the amount and depth of the sorbed tritium, it should be possible to calculate a correction for this effect.

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