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Study of the Scattering Correction for Thick Uranium-Oxide and Other α-Particle Sources—I: Theoretical

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Theoretical values of the 2π α-particle counting rate, $C_{2\pi}$, including scattering, divided by the disintegration rate, N_0, are calculated for α-particle sources mounted on flat backing materials. The theoretical values (Part I) and experimental values (Part II) are in good agreement, and show that

(a) $C_{2\pi}/N_0$ values as a function of α-particle energy and source thickness can be calculated on the basis of a simple physical model.
(b) The scattering correction, expressed as a fraction of the disintegration rate, decreases with increasing source thickness.
(c) The α-particle scattering in uranium-oxide is much less than the scattering in platinum, in disagreement with previous estimates.

The calculations are readily extended to other source compositions and α-particle energies.

INTRODUCTION

THE NATIONAL BUREAU of Standards has, for many years, produced and calibrated α-particle radioactivity standards. For reasons of stability and convenience, such α-particle sources are commonly mounted on flat metal backings, such that the possible solid angle for emission is 2π steradians. When such a source is measured in a counter having 2π geometry, the 2π counting rate, $C_{2\pi}$, is not one-half of the disintegration rate, N_0, however, because some of the α-particles initially emitted downward are backscattered into the sensitive volume of the counter, while some of the α-particles initially emitted upward are scattered and/or absorbed in the source. As a result, the measured $C_{2\pi}/N_0$ ratio can vary significantly from 0·50, depending upon the composition and thickness of the backing and the source material.

In order to accurately determine N_0 from a measurement of $C_{2\pi}$, the corrections for scattering and self-absorption must be well known. A few measurements and calculations of $C_{2\pi}/N_0$ have appeared in the literature,[1–3] but, in general, the uncertainty in the reported values is unacceptably large for our purposes. In Part I of this paper we present a simple method for accurately calculating $C_{2\pi}/N_0$, based upon the following assumptions:

(1) A large number of α-particles emitted from the same initial position with identical velocity vectors will not all have the same final position. Because of small-angle scattering along the trajectory (the probability of large angle scattering is negligible), the distribution of final positions is Gaussian about the mean final position.

(2) The Gaussian distribution about the mean final position is spherically symmetric, so that the probability of a final position different from the mean by an amount Δy is independent of the initial angle of emission. Hence, the calculation can be carried out in terms of a one-dimensional Gaussian distribution.

(3) For α-particles that pass through more than one material, the standard deviation, σ, which characterizes the Gaussian distribution at the end of the trajectory, can be calculated in terms of the σ values for each of the materials along the trajectory.

In Part II of this paper we present additional

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experimental evidence that confirms the accuracy of the theoretical calculations.

THEORETICAL

The ratio $C_{2\pi}/N_0$ may be calculated using the model shown in Fig. 1. The model consists of an active source layer of thickness d, a front absorber of thickness t, a backing material of thickness b, a very thick rear mounting material, and

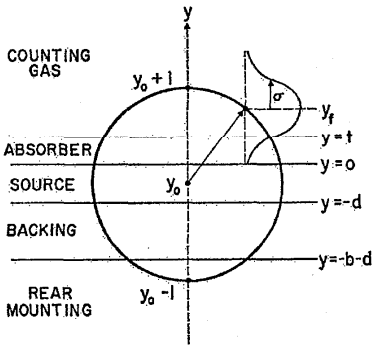

FIG. 1. The model used to calculate $C_{2\pi}/N_0$. All of the parameters (source thickness, backing thickness, etc.) are in dimensionless reduced form, obtained by expressing them as a fraction of the α-particle range in the same material. Thus the total range always has a value of 1. The scattering is shown greatly exaggerated.

a very thick layer of counting gas. The layers are assumed to be homogeneous, parallel, and laterally semi-infinite. As a matter of mathematical convenience, the thickness of these layers, as well as all of the other distances in this calculation, are expressed in dimensionless reduced form. This dimensionless reduced form is obtained by dividing the distance in physical units (e.g. cm) by the α-particle range in the *same* material. With all of the distances expressed in this reduced form, the total range always has a value of 1. Because of small-angle scattering along the trajectories, α-particles emitted from the same initial position with identical velocity vectors exhibit a distribution in position at the end of their ranges. The endpoint distribution is approximately Gaussian and approximately spherically symmetrical about the mean endpoint.* This Gaussian endpoint distribution is characterized by its standard deviation, σ, which is a function of the projectile charge, velocity, and mass and of the material(s) traversed.

To evaluate $C_{2\pi}/N_0$ we integrate over all initial positions from $y_0=-d$ to $y_0=0$. For each initial position, y_0, we integrate over all mean final positions from $y_f=y_0-1$ to $y_f=y_0+1$.† For each initial and mean final position, y_0 and y_f, the Gaussian endpoint distribution is determined. Particles with an endpoint above the line $y=t$ are assumed to be counted; particles with an endpoint below this line are not counted. The value of $C_{2\pi}/N_0$ is then given by

$$\frac{C_{2\pi}}{N_0}=\frac{1}{2d}\int_{y_0=-d}^{y_0=0}\int_{y_f=y_0-1}^{y_f=y_0+1}\int_{\Delta y=t-y_f}^{\Delta y=+\infty}\frac{1}{\sigma\sqrt{2\pi}}\times\exp\left(\frac{-\Delta y^2}{2\sigma^2}\right)\mathrm{d}\Delta y\,\mathrm{d}y_f\,\mathrm{d}y_0. \quad (1)$$

If values of the probability integral are available only for positive Δy, the innermost integral in equation (1) may be rewritten as

$$1-\int_{\Delta y=|t-y_f|}^{\Delta y=+\infty}\frac{1}{\sigma\sqrt{2\pi}}\exp\left(\frac{-\Delta y^2}{2\sigma^2}\right)\mathrm{d}\Delta y, \quad (2)$$

* The approximately Gaussian nature of the endpoint distribution in both the longitudinal and transverse directions is well documented. See, for example,[1,4,5,6,8]. Although no single experimenter has measured both the longitudinal and transverse end-point distributions in the same material, comparison of the results of different experimenters[4,8] suggests that the two distributions are the same to within the experimental error.

† Consider a unit sphere about the point y_0. The surface area on this sphere that lies between the planes $y=y'$ and $y=y'+\mathrm{d}y$ is independent of y' for $y_0-1\leq y'\leq y_0+1$. Hence it is more convenient to integrate over the mean final y value, y_f, than to integrate over the angle of emission, θ. The equivalence of the two integration variables may be seen by noting that

$$\frac{dA}{dy_f}=\frac{-2\pi r^2\sin\theta\,\mathrm{d}\theta}{-r\sin\theta\,\mathrm{d}\theta}=2\pi r=\text{constant},$$

where θ is the angle between the y axis and the mean particle trajectory.

or $(t - y_f) < 0$. Equation (1) can also be written in the form‡

$$C_{2\pi}/N_0(b, d, t, \sigma) = C_{2\pi}/N_0(d, t, \sigma = 0) + B(b, d, t, \sigma), \quad (3)$$

where B is defined as the scattering correction.‖ The first term on the righthand side of equation (3) is simply $C_{2\pi}/N_0$ for the case where there is no scattering.

The problem that remains is to evaluate σ as a function of y_0 and y_f. We approximate the variance, σ^2, at the end of each trajectory as the weighted summation of the variances in each of the materials along the trajectory.§ The weighting factor is the fraction of the reduced range spent in each material. The value of σ as a function of y_0 and y_f is then given by the following equations, where σ_a, σ_b, σ_g, σ_r, and σ_s are the standard deviations of the scattering in the absorber, the backing, the counting gas, the rear mounting, and the source, respectively.

$$\begin{aligned}
&t \le y_f \le (y_0 + 1), \quad \sigma^2 = \sigma_s^2(-y_0)/C + \sigma_a^2 t/C + \sigma_g^2(1 + (y_0 - t)/C);\\
&0 \le y_f \le t, \quad \sigma^2 = \sigma_s^2(-y_0)/C + \sigma_a^2(1 + y_0/C);\\
&-d \le y_f \le 0, \quad \sigma = \sigma_s \qquad (4)\\
&-(b + d) \le y_f \le -d, \quad \sigma^2 = \sigma_s^2(-d - y_0)/C + \sigma_b^2(1 + (d + y_0)/C);\\
&(y_0 - 1) \le y_f \le -(b + d),\\
&\sigma^2 = \sigma_s^2(-d - y_0)/C + \sigma_b^2(-b)/C + \sigma_r^2(1 + (b + d + y_0)/C);
\end{aligned}$$

where

$$C = y_f - y_0.$$

The value of σ for solid materials can be determined by noting that if $d = t = 0$, $b \ge 1$, σ_g is negligibly small,‖ and σ_b is small compared to 1 ($\sigma_b \lesssim 0{\cdot}3$), then equation (1) becomes simply¶

$$C_{2\pi}/N_0 = 0{\cdot}5\left(1 + \sigma_b\sqrt{\frac{1}{2\pi}}\right) = 0{\cdot}5(1 + 0{\cdot}399\sigma_b). \quad (5)$$

‡ For $(d + t) \le 1$, the first term on the righthand side of equation (3) can be written in non-reduced form as[7]

$$C_{2\pi}/N_0(d, t, \sigma = 0) = 0{\cdot}5(1 - d/2R_s - t/R_a),$$

where R_a and R_s are the α-particle ranges in the absorber and the source, respectively, and all of the parameters are expressed in physical units.

‖ The scattering correction, B, is often referred to as the "backscattering." In general, however, the scattering correction is due to:

(a) Particles emitted in the forward direction that would reach the sensitive volume if there were no scattering, but which are scattered back into the source (or backing or absorber) before reaching the counting gas.

(b) Particles emitted in the forward direction that would not reach the sensitive volume if there were no scattering, but which are scattered out of the source (or backing or absorber) into the counting gas.

(c) Particles emitted in the backward direction that are scattered back into the forward direction and reach the counting gas.

In a strict sense, only contribution (c) is really backscattering.

§ Early workers found that the mean scattering angle, and hence the σ value, of α-particles traversing metal foils was approximately proportional to the thickness of the metal.[4] More recent work with both solids and gases as scattering materials[5,8] gives results that are in better agreement with a linear increase in variance along the trajectory. A similar study of α-particle energy straggling in UO_2 and ThO_2[6] gives results that are somewhat intermediate, but closer to a linear increase in variance. The difference between $C_{2\pi}/N_0$ calculated using a weighted summation of standard deviations, σ, and $C_{2\pi}/N_0$ calculated using a weighted summation of variances, σ^2, is a maximum of 0·0010 and an average of less than 0·0005 ($< 0{\cdot}1\%$ of $C_{2\pi}/N_0$) for the present

‖ The scattering in gases is actually quite small (see Table 1), but not completely negligible. However, σ_g is assumed to be effectively zero, based upon the argument that α-particles scattered out of the gas probably still lose enough energy in the gas to be counted.

¶ Equation (5) is equivalent to the result of CRAWFORD[1] *for this case only*. CRAWFORD defines his backscattering B' such that

$$2B = B' = 0{\cdot}402\Phi,$$

where 0·402 is CRAWFORD's value corresponding to $\sqrt{\frac{1}{2\pi}}$ and Φ is the RMS scattering angle. Hence, there is a direct correspondence between σ and CRAWFORD's value of Φ at the end of the particle range under these conditions.

TABLE 1. Values of σ for α-particles in various scattering materials.

Scattering material	$C_{2\pi}/N_0$ (5·31 MeV)	Ref.	σ (5·31 MeV)	n	σ (4·64 MeV)	σ (4·19 MeV)
Be	0·4997[a]	9	<0·001[b]	1·57	<0·001[c]	<0·001
C	0·5002	9	~0·001	1·60	~0·001	~0·001
Al	0·5020	9	0·014	1·45	0·015	0·017
Ti	0·5043	9	0·026	1·53	0·028	0·031
Stainless Steel	0·5052	9	0·030	1·46	0·033	0·036
Fe	0·5053	9	0·030	1·45	0·033	0·036
Ni	0·5058	9	0·033	1·39	0·036	0·039
Monel	0·5058	9	0·033	1·39	0·036	0·039
Brass	0·5061	9	0·035	1·36	0·038	0·041
Cu	0·5061	9	0·035	1·37	0·038	0·041
Ag	0·5107	9	0·058	1·41	0·063	0·068
Ta	0·5174	9	0·091	1·28	0·099	0·106
Pt	0·5187	9	0·098	1·32	0·107	0·114
Au	0·5189	9	0·099	1·32	0·108	0·115
U_3O_8	0·5049	This work	0·026	1·30	0·028	0·030
UO_2	—	6	0·030	1·29	0·033	0·036
Air	—	5	0·010	1·58	0·011	0·012

(a) The $C_{2\pi}/N_0$ values shown for metals were calculated using the least squares equation given in reference 9 for ^{210}Po in thin ($d \approx 10^{-3}$) collodion films.

(b) The σ values for 5·31 MeV α-particles in the metals and in U_3O_8 have been adjusted to $d = 0$ (and $b \geq 1$ for U_3O_8) by means of a self-consistent calculation using equations (1) and (4). In this calculation a σ value of 0·010 was assumed for 5·31 MeV α-particles in collodion.

(c) The σ values for 4·64 and 4·19 MeV α-particles (with the exception of UO_2) were calculated from the σ values for 5·31 MeV α-particles using equation (11) and the n values shown. The σ values shown for UO_2 are all experimental.

Thus the value of σ for a solid material can be evaluated from a measurement of $C_{2\pi}/N_0$ for a weightless source deposited upon that material as a backing. Such measurements have been previously reported for a number of metal backings using ^{210}Po deposited in a very thin ($d \approx 10^{-3}$) collodion film.[9] These results and the corresponding values of σ are shown in Table 1. Also shown are the results of an identical measurement using ^{210}Po in collodion on a U_3O_8 backing and estimates of the value of σ for ^{210}Po α-particles in air[5] and UO_2.[6] Note that the values of σ for U_3O_8 and UO_2 are significantly less than the value of σ for platinum, in disagreement with previous estimates that they should be approximately equal.[1,3] It appears that σ is approximately linearly proportional to Z for metals and perhaps to the mean Z of the molecule for compounds such as UO_2 and U_3O_8. However, the data presently available do not permit a meaningful analysis of the σ vs Z relationship for compounds.

For the $^{235}UO_2$ sources measured in our laboratory, the mean α-particle energy is 4·64 MeV. (Approximately 65% of the α-particles are emitted from the ^{234}U in the source and approximately 35% from the ^{235}U[10].) For the $^{238}UO_2$ sources the mean α-particle energy is 4·19 MeV. In order to evaluate equation (1) for these sources, it is first necessary to adjust the σ (5·31 MeV) values given in Table 1 for the difference in α-particle energies. The variation of σ with α-particle energy is actually a complicated function of the properties of the projectile and the scattering material, but, for energy differences of less than about a factor of two, we approximate the variation by assuming that:

(a) The number of scattering collisions, N, is approximately proportional to the range in the

material. Thus

$$\frac{N(E_1)}{N(E_2)} = \frac{R^*(E_1)}{R^*(E_2)}, \tag{6}$$

where * signifies that the parameter is expressed in physical units, rather than in dimensionless reduced form.

(b) The scattering distribution is Gaussian, so that

$$\frac{\sigma^*(E_1)}{\sigma^*(E_2)} = \sqrt{\left(\frac{N(E_1)}{N(E_2)}\right)} = \sqrt{\left(\frac{R^*(E_1)}{R^*(E_2)}\right)}. \tag{7}$$

However, since by definition

$$\sigma(E_1) = \frac{\sigma^*(E_1)}{R^*(E_1)}, \tag{8}$$

it follows that**

$$\frac{\sigma(E_1)}{\sigma(E_2)} = \sqrt{\left(\frac{R^*(E_2)}{R^*(E_1)}\right)}. \tag{9}$$

The calculation of σ as a function of energy can be further simplified if the range ratio can be expressed as an analytic function of α-particle energy. Over the range of α-particle energies from 3 to 6 MeV, the range *ratio* is given quite accurately (±5%) by the equation

$$\frac{R^*(E_2)}{R^*(E_1)} = \left(\frac{E_2}{E_1}\right)^n, \tag{10}$$

where n has a value of approximately 1·5. The combination of equations (9) and (10) gives the variation of σ with energy as

$$\frac{\sigma(E_1)}{\sigma(E_2)} = \left(\frac{E_2}{E_1}\right)^{n/2}. \tag{11}$$

Values of n appropriate for 3–6 MeV α-particles in the various scattering materials are given in Table 1. The σ values predicted by equation (11) for 4·64 and 4·19 MeV α-particles are also shown. (The σ values for UO_2 are experimental.)

As a test of equations (9) and (11), the ratio σ(Pt, 3·18 MeV)/σ(Pt, 5·31 MeV) was computed and is compared to the experimental data in Table 2. The predicted ratios are in good agreement with the experimental ratio, suggesting that equation (9) or (11) may be used with some confidence over this energy range, at least for α-particles scattered by platinum.††

** Crawford[1] predicts a similar dependence for his RMS scattering angle Φ.

COMPARISON OF RESULTS

Now that σ has been determined for UO_2, equation (1) can be evaluated for both 4·64 and 4·19 MeV α-particles emitted from a UO_2 source of thickness d on a thick ($b \geq 1$) platinum backing.§§ The results are shown in Fig. 1, Part II, along with the experimental points for our $^{235}UO_2$ sources. Also shown are the results of Gold and Armani[2] and White[3] for U_3O_8 sources evaporated on platinum backings. The diagonal straight line corresponds to self-absorption but no backscattering ($\sigma = 0$). The lower, heavy curve shown was calculated using equations (1) and (4) with $b > 1$, $\sigma_b = 0{\cdot}107$, $\sigma_g = 0$, $\sigma_s = 0{\cdot}033$, and $t = 0$. This corresponds to 4·64 MeV α-particles in $^{235}UO_2$ on a thick platinum backing (see Table 1). It also corresponds very closely to the conditions for the U_3O_8 foils of Gold and Armani[2] (4·76 MeV) and White[3] (4·7 MeV). The upper, dashed curve was calculated for 4·19 MeV α-particles in $^{238}UO_2$ ($\sigma_b = 0{\cdot}114$, $\sigma_s = 0{\cdot}036$). The difference between the two calculated curves is small, typically 0·0005–0·0010 (0·1–0·2% of $C_{2\pi}/N_0$). Thus, to within an uncertainty of 0·1–0·2%, a single curve can be used for all UO_2 and U_3O_8 sources on thick platinum backings.

An important difference between the present calculated results and the previous calculations of Crawford[1] is that equation (1) predicts that the scattering correction decreases rapidly with increasing source thickness, rather than

†† Note that the σ value for 5·31 MeV α-particles is lower when the ^{210}Po is deposited directly on the polished platinum backing (Table 2) than when the ^{210}Po is suspended in a thin collodion film (Table 1). This is possibly due to absorption caused by minute surface irregularities.[9] The effect should be comparable, or perhaps slightly greater, for the 3·18 MeV α-particles. The experimental σ ratio for ^{210}Po and ^{148}Gd sources in collodion film on platinum backings would therefore be expected to be comparable to, or slightly greater than, the experimental ratio shown in Table 2.

§§ A FORTRAN program for evaluating $C_{2\pi}/N_0$ using equations (1) and (4) with any combination of source parameters is available upon request to the authors.

TABLE 2. Calculated and experimental σ ratios for 3·18 and 5·31 MeV α-particles in platinum

Source	α-Energy (MeV)	σ(Pt)[a]	σ(Pt, 3·18 MeV)/σ(Pt, 5·31 MeV) Experimental	Equation (9)	Equation (1)
Gd148–1	3·18	0·121 ± 0·007	1·38 ± 0·14	1·40	1·40
Po210–1	5·31	0·088 ± 0·008			

(a) From Table 1, Part II, of this work.

remaining constant. The primary reason for this difference, as pointed out by WHITE[3], is that CRAWFORD's calculation ignores the scattering in the backward direction. This rapid decrease in B with increasing source thickness is predicted to occur even when σ_b and σ_s are identical (Fig. 2).

The largest uncertainty involved in comparing the calculated and experimental $C_{2\pi}/N_0$ values in Fig. 1, Part II, is due to the uncertainty in the value of the α-particle range in UO_2 and U_3O_8. The value of d for the experimental points is equal to d^*/R_s^*, and hence the uncertainty in the value of R^* results in an equal uncertainty in d. A comparison of calculated and experimental α-particle ranges in UO_2 and U_3O_8 is shown in Table 3. None of the available range tables[11,12,13] gives the range in UO_2 or U_3O_8 explicitly, so the range in these materials was estimated using the equation[14]

$$\frac{1}{R^*_{U_xO_y}(E)} = \frac{w_O}{R^*_O(E)} + \frac{w_U}{R^*_U(E)}. \qquad (12)$$

Here $R^*_{U_xO_y}(E)$, $R^*_O(E)$ and $R^*_U(E)$ are the ranges of an α-particle of energy E in the uranium oxide, in pure oxygen, and in pure uranium, respectively, and w_O and w_U are the weight fractions of oxygen and uranium in the uranium oxide. It should be emphasized that equation (12), despite common use, is only a crude approximation to the range in chemical compounds. As shown in Table 3, the error involved can exceed 30 %. The ranges calculated using equation (12) were used, however, to determine the range *ratio* for α-particles of the same energy in UO_2 and U_3O_8. This ratio and the experimental range data for α-particles in UO_2 were then used to calculate the α-particle ranges in U_3O_8. Approximate range values for collodion are also given in Table 3.

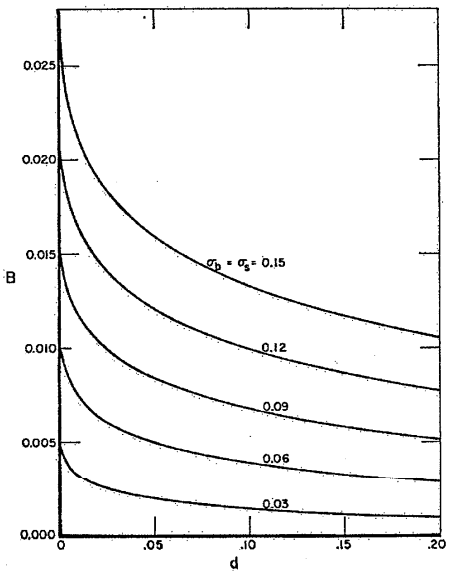

FIG. 2. The scattering correction, B (as defined in equation (3)), vs the source thickness, d, for sources with σ_b equal to σ_s. The values of the other variables are: $b > 1$; $t = 0$; $\sigma_s = 0$.

CONCLUSIONS

(1) The α-particle scattering in UO_2 and U_3O_8 is much less than the scattering in platinum, in disagreement with previous estimates.

(2) The scattering correction, *expressed as a fraction of the disintegration rate*, decreases with increasing source thickness.

(3) An equation is presented and tested that relates the relative α-particle scattering to the relative α-particle energy.

TABLE 3. α-particle ranges in UO_2 and U_3O_8

Scattering material	α-Energy (MeV)	Range (mg/cm²) From range tables Ref. 11*	Ref. 12	Ref. 13	Other Ref. 3	Experimental Ref. 6
UO_2	5·31	12·9	15·7	15·2		12·5 ± 0·2
UO_2	4·76	11·1	13·6	13·1		10·7 ± 0·2
UO_2	4·70	10·9	13·4	12·9		10·5 ± 0·2
UO_2	4·64	10·6	13·1	12·6		10·3 ± 0·2
UO_2	4·19	9·2	11·6	11·1		9·0 ± 0·2
U_3O_8	5·31	12·1	14·5	14·1		11·6 †
U_3O_8	4·76	10·3	12·5	12·1		10·0 †
U_3O_8	4·70	10·1	12·3	11·9	11·2	9·8 †
U_3O_8	4·64	9·9	12·1	11·7		9·6 †
U_3O_8	4·19	8·6	10·7	10·2		8·3 †
Collodion	5·31	3·8	4·0	—		—
U_3O_8/UO_2	5·31	0·94	0·92	0·93		
U_3O_8/UO_2	4·76	0·93	0·92	0·93		
U_3O_8/UO_2	4·70	0·93	0·92	0·93		
U_3O_8/UO_2	4·64	0·93	0·92	0·93		
U_3O_8/UO_2	4·19	0·93	0·92	0·93		

* Ranges for protons of equivalent E/M.
† Calculated as 0·93 times the range in UO_2 for α-particles of the same energy.

(4) A simple Gaussian-scattering model is used to calculate $C_{2\pi}/N_0$ for $^{235}UO_2$ and $^{238}UO_2$ sources as a function of source thickness. The calculated results are in good agreement with the experimental data. The calculation is readily extended to a wide variety of other source compositions.

(5) The standard deviation of the scattering, σ, expressed as a fraction of the α-particle range, is given for a number of scattering materials. For metals, σ increases approximately linearly with increasing Z. For chemical compounds, such as UO_2 and U_3O_8, σ appears to be proportional to the mean Z of the molecule. However, the limited data presently available do not permit a meaningful analysis of the σ vs Z relationship for compounds in general.

(6) Expressions such as equation (12), which attempt to determine particle ranges in chemical compounds simply on the basis of the particle ranges in the constituent elements, are often seriously in error. Such expressions should be used with caution. Experimental range data for chemical compounds are usually much more accurate and should be used if available.

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