A Correction for Gamma-Ray Self-Attenuation in Regular Heterogeneous Materials

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ABSTRACT

A procedure for determining the total correction factor for gamma-ray self-attenuation in regular heterogeneous materials is derived and discussed. The result of a practical application of the procedure to the passive gamma-ray assay of the ^{235}U content of high-temperature gas reactor fuel is presented.

The problem of microsphere attenuation in the gamma-ray assay of high-temperature gas reactor (HTGR) fuel materials is discussed in Ref. 1. HTGR fuels are regular heterogeneous materials, defined here as mixtures of a low-Z, low-density matrix with a quantity of identical higher Z, higher density particles containing the gamma-ray emitting substance. The procedures described in Ref. 1 were essentially correct, although the recommended forms for the various correction factors were approximate. Experience and increased understanding of the general problem make it both possible and profitable to revise and extend the discussion.¹

The most difficult part of a gamma-ray assay problem is the correction for the gamma-ray absorption within the sample itself. For samples meeting the assumption that the mixture of assay material and matrix is reasonably uniform and that the particles of assay material are small enough to ignore self-absorption within the individual particles, the correction-factor problem is well understood for most common and reasonable sample configurations. The most generally useful correction method involves determining the linear attenuation coefficient μ of the sample by measuring the transmission through the sample of an external beam of gamma rays and then computing the correction for the sample from its known dimensions and measured μ .^{2,3} The method by itself, however, is not adequate for regular heterogeneous materials such as the HTGR

fuels, which stimulated this study and which, indeed, are a relatively uniform mixture of matrix ($\rho \approx 2$, $Z \approx 6$) and small, high-density particles ($\rho \approx 10$, $Z \approx 90$) of gamma-ray emitting materials, which individually have significant ($\approx 10\%$) self-absorption.

The discussion of this more difficult problem begins with a consideration of the definition of correction factors in general. A useful definition for the total correction factor for the self-absorption within a sample is given by

$$CF_S \equiv R_0/R_S \quad , \quad (1)$$

where

CF_S = sample correction factor,

R_0 = rate of gamma-ray events that would have been detected if
the sample had no self-absorption, and

R_S = actual detected rate from a given sample.

Clearly, CF_S is not computed from the defining relation because R_0 is the ultimately desired quantity that is obtained by using the relation in the form

$$R_0 = R_S \cdot CF_S \quad . \quad (2)$$

The observed rate R_S must also be corrected for losses from electronic pileup and deadtime. Corrections for these rate-related losses are not considered here, and in what follows it will be assumed that the necessary corrections have been made.

Normally, R_S is determined from a gamma-ray spectrum from the sample, and CF_S can be determined in several ways, usually by the transmission measurement method mentioned above.^{2,3} Another much discussed but little used method of determining CF_S compares the ratio of two gamma rays of significantly different energies from the sample with the ratio of the same two gamma

rays from a "no attenuation" source. Let i and j denote gamma rays of different energy. Then,

$$R_S(i) \cdot CF_S(i) = R_O(i) , \quad (3)$$

and

$$R_S(j) \cdot CF_S(j) = R_O(j) . \quad (4)$$

Then, by division and minor manipulation,

$$\frac{CF_S(i)}{CF_S(j)} = \frac{\left[\frac{R_O(i)}{R_O(j)} \right]}{\left[\frac{R_S(i)}{R_S(j)} \right]} , \quad (5)$$

which implies that by themselves ratios of gamma-ray pairs can give only the ratio of the actual correction factors for the two gamma rays. When combined with some known or assumed knowledge of the sample, the actual correction factors often can be determined, and various procedures have been worked out to do so for several types of samples.^{1,4-7}

In the case of regular heterogeneous materials, the total sample correction factors can be found by using both transmission and ratio measurements in addition to some modest prior knowledge of the material.¹ The reason that both techniques are required is that there are actually two corrections to be determined, one for the self-absorption within the emitting particles and one for absorption after the gamma rays leave the emitting particles. The two correction factors are independent, so

$$CF_s = CF_p \cdot CF_t , \quad (6)$$

where CF_p is the correction factor for absorption within the particles themselves and CF_t is the correction factor for all subsequent absorption, which is described by the transmission through the sample. Thus,

$$R_s(i) \cdot CF_p(i) \cdot CF_t(i) = R_o(i) , \quad (7)$$

and

$$R_s(j) \cdot CF_p(j) \cdot CF_t(j) = R_o(j) , \quad (8)$$

from which

$$\frac{CF_p(i)}{CF_p(j)} = \frac{\left[\frac{R_o(i)}{R_o(j)} \right]}{\left[\frac{R_s(i)}{R_s(j)} \right]} \cdot \frac{1}{\left[\frac{CF_t(i)}{CF_t(j)} \right]} . \quad (9)$$

Now, $CF_t(i)$ and $CF_t(j)$ are found by determining the transmissions of the sample at energies $E(i)$ and $E(j)$ and then computing for the particular geometry. If the particle dimensions (they are usually spherical or can be considered such) are very small relative to the size of the sample, $CF_t(i)$ and $CF_t(j)$ may be computed with the same algorithms used in computing the CF_t for a uniform, homogeneous sample. However, if the particle diameter is a significant fraction of the sample dimension, the algorithms must be modified. This is the case with the HTGR fuels because of the size of their high-density particle kernels and because of the relatively thick coating of low-Z material surrounding the kernels.

The ratio $[R_S(i)/R_S(j)]$ is, of course, taken from the gamma spectrum of the sample. The ratio $[R_O(i)/R_O(j)]$ may be found in several ways. Often it is found by using a thin source with near-zero self-absorption counted in the same geometry as the sample. It may also be found by measurements of a convenient standard with negligible particle self-absorption $[CF_p(i) = CF_p(j) = 1]$ and use of the relation

$$\frac{R_O(i)}{R_O(j)} = \frac{R_S(i) \cdot CF_t(i)}{R_S(j) \cdot CF_t(j)} . \quad (10)$$

Finally, the ratio $[R_O(i)/R_O(j)]$ can be computed if the relative efficiency for the particular detector is well known and if the relative branching ratios of the two gamma rays also are well known. At any rate, the ratio of the particle correction factors is now known from Eq. (9), and it remains to combine that information with the minimum amount of prior knowledge of the sample to obtain the actual value of $CF_p(i)$ and $CF_p(j)$.

First, however, a few comments on the analytic forms of CF_p and CF_t are in order. In the discussion of Ref. 1, the form for CF_p was given as $CF_p = 0.74 \cdot \mu\rho D / (1 - e^{-0.74 \cdot \mu\rho D})$. This form is a simple, useful approximation accurate within 1% for $CF_p \leq 1.5$. However, an exact expression has been derived by several investigators, which though more complicated is usable even with simple programmable calculators.⁸ The exact expression is

$$CF_p = \left(\frac{3}{2\mu\rho D} \left\{ 1 - \frac{2}{(\mu\rho D)^2} + e^{-\mu\rho D} \left[\frac{2}{\mu\rho D} + \frac{2}{(\mu\rho D)^2} \right] \right\} \right)^{-1} , \quad (11)$$

where

μ = mass attenuation coefficient for the spherical emitter,

ρ = density of sphere, and

D = diameter of sphere.

Equation (11) is derived as $1/CF_p$, the fraction of gamma rays escaping from the spherical emitter. If the detector is assumed to be a portion of a concentric spherical shell (in most cases a very good approximation), the reciprocal of that fraction is exactly the correction factor required.

It should be noted that in Ref. 1, CF_t was given by

$$CF_t = \frac{-\frac{\pi}{4} \ln(t)}{1 - t^{\frac{\pi}{4}}}, \quad (12)$$

where t is the measured transmission through a cylindrical sample. This expression is also a simple, useful, approximate form, especially if one's computational equipment is confined to nonprogrammable calculators, but it can be in error by as much as 10%, depending on the value of t and the counting geometry. To explain briefly, the approximate form is a function of t only, whereas it is known that CF_t is also a mild function of D/R , where D is the distance from the sample center to the detector and R is the sample radius. Unfortunately, no exact analytic form is now known to exist for $CF_t(t, D/R)$. However, a reasonably convenient way to get a very good approximation consists of calculating $CF_t(t, D/R)$ by numeric integration for a number of values of t for a given geometry (D/R), then fitting to a quadratic of form $CF_t = A + B \log t + C (\log t)^2$. Inasmuch as most plots of $CF_t(t, D/R)$ vs $\log(t)$ are nearly linear, a given set of A , B , and C yields results accurate to $\leq 0.1\%$ over rather wide ranges of t . Where the most accurate results are desired (especially near-field counting geometries), the numerically computed values of CF_t also may include the effects of sample height and container attenuation.

In the discussion of Ref. 1 it was shown that having adequately accurate forms for CF_p and CF_t , knowing the mass attenuation coefficients of the particles at the two energies concerned [$\mu(i)$ and $\mu(j)$], and knowing the density of the particles was sufficient to enable determination of the absolute values of $CF_p(i)$ and $CF_p(j)$. Note that knowing the mass attenuation coefficients is equivalent to knowing the chemical composition of the particles. It will be shown below that it is sufficient to have the forms for CF_p and CF_t and to know

only the ratio of $\mu(i)$ and $\mu(j)$. This requirement represents a considerable relaxation, particularly for HTGR-type materials, which have compositions such as UC_2 , ThO_2 , and UN . The low-Z component of the HTGR-type materials contributes $\leq 1\%$ to the μ values for energies ≤ 200 keV. It is also true that for high-Z elements (for example, $Z > 80$) all their plots of $\log \mu$ vs $\log E$ are essentially parallel, especially for energies ≤ 500 keV, which are of the most interest. This fact means that for HTGR-type compounds the ratio $\mu(i)/\mu(j)$ is nearly independent of composition and is essentially a function of the two energies alone. Thus, the details of chemical composition are unimportant. It does not matter, for example, if the composition is $\text{UC}_{1.5}$ rather than UC_2 , or if the particles are mixtures of uranium, thorium, and plutonium compounds.

The fact that only the ratio of $\mu(i)$ and $\mu(j)$ is required in addition to the forms of CF_p and CF_t in order to obtain values for $\text{CF}_p(i)$ and $\text{CF}_p(j)$ from the ratio $\text{CF}_p(i)/\text{CF}_p(j)$ follows essentially from the fact that CF_p is a function of the product $\mu\rho D$ only. Obviously, $\mu(i) \neq \mu(j)$ for different energies, and thus $\text{CF}_p(i)$ and $\text{CF}_p(j)$ are different functions of ρD . Figure 1 shows an example for $E(i) = 143.8$ keV and $E(j) = 185.7$ keV. The values of $\mu(i)$ and $\mu(j)$ are computed for the UC_2 particles of the current round-robin assay exercise. Clearly, a previously determined value of $\text{CF}_p(i)/\text{CF}_p(j)$ determines a unique ρD , which, combined with the known $\mu(i)$ and $\mu(j)$, yields the desired values of $\text{CF}_p(i)$ and $\text{CF}_p(j)$. Now assume that $\mu(i)$ and $\mu(j)$ have been chosen incorrectly, both being in error by the factor k , so that $\mu'(i) = k\mu(i)$ and $\mu'(j) = k\mu(j)$. Obviously, $\mu(i)/\mu(j) = \mu'(i)/\mu'(j)$. Clearly, $\text{CF}_p[\mu'(i)\rho D]$ and $\text{CF}_p[\mu'(j)\rho D]$ will have the same values at argument $\rho D/k$ as do $\text{CF}_p[\mu(i)\rho D]$ and $\text{CF}_p[\mu(j)\rho D]$ at argument ρD . If the values are the same, the ratio is the same, and therefore a given ratio $\text{CF}_p(i)/\text{CF}_p(j)$, when used with μ values "incorrect" by the factor k , determines a ρD value "incorrect" by a factor $1/k$. The total arguments determined are the same (trivially $\mu\rho D = k\mu\rho D/k$), and therefore the absolute values of $\text{CF}_p(i)$ and $\text{CF}_p(j)$ are the same as if the correct values of $\mu(i)$ and $\mu(j)$ had been used. The error only makes it impossible to get an accurate estimate of ρD , which otherwise is also obtained along with the values of the correction factors $\text{CF}_p(i)$ and $\text{CF}_p(j)$.

The procedures discussed here have been successfully applied to the determination of CF_p for HTGR fuel rods even though the 143.8-keV and 185.7-keV gamma rays that must be used for the ratio measurements are too close together for the best results. (The 129.3-keV and 413.7-keV pair of ^{239}Pu , for example,

Fig. 1.

Self-attenuation correction factors for spherical UC_2 particles at 185.7 and 143.7 keV as a function of ρD (ρ = density, D = particle diameter). The ratio of the two correction factors is given also (right-hand ordinate).

would be considerably more advantageous.) Because the composition, density, and diameter of the HTGR particles are well known, a computed value in this case could be compared with the measured value. The computed value was 1.10 and the measured value was 1.11 with an uncertainty of ~1%.

Although the number of regular heterogeneous materials found in practice is probably not large, they do exist, and the procedure described is useful in obtaining less biased NDA results when measurements of heterogeneous materials

are made relative to uniform, homogeneous standards. The procedure also minimizes the number of standards required.

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