

The Calculation of Self Attenuation Factors for Simple Bodies in the Far Field Approximation.

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Abstract

Microsoft Excel spreadsheet functions have been developed for the calculation of the self attenuation of gamma-rays in simple bodies viewed from afar. The cases are a uniform rod viewed along its axis, a sphere viewed along a diameter and a cylinder (or disk) viewed along a mid-plane diameter. The results for the former two cases can be expressed in closed form while the algorithm used for the third is described. We also develop useful expressions for small and large lump cases. The self attenuation functions have been used along with other numerical methods to generate and validate test data for exercising a proposed new lump correction algorithm based on exploiting the differential attenuation of different energy γ -rays emitted by an item.

Introduction

The measurement of special nuclear materials such as ^{235}U and ^{239}Pu , in radioactive waste by the application of high resolution gamma-ray spectrometry is a widely used technique [1]. If the nuclides are present the form of “lumps” (such as shavings, chips, pellets, foundry spills, crevice accumulations and the like) rather than as dilutely distributed activity in and on the bulk waste matrix, then self attenuation may occur. Self attenuation is not accounted for by the usual transmission source and weight based matrix correction factor methods. This is because dense lumps of significant size, sufficient to affect the assay result, are still physically small in relation to the overall size of most waste container types [2]. Therefore, traditional gross matrix correction methods are not sensitive to the presence of lumps.

If the presence of lumps goes unrecognized then an assay system calibrated using dilute (minimally absorbing) standards will underreport when put into operation. This is because the number of γ -rays emerging from the lump per unit mass of nuclide present will be less than assumed.

In order to examine the importance of self attenuation by numerical simulation it is desirable to have a simple way to calculate the attenuation factor for lumps in a variety of shapes. In essence, a shape defines a particular distribution of emergent path lengths. By using a given lump shape or combination of shapes various measurement scenarios can be played out to examine the impact of self attenuation as a function of photon energy and nuclide mass given assumptions on the nature (chemical composition, density,

enrichment) of the lumps. This is important to do as part of the assessment of a reasonable and justifiable total measurement uncertainty and also when bounding or limiting assay results are to be reported.

Self Attenuation Factors

Expressions for the self attenuation factors (SAF's) for several common shapes have been given by Dixon [3]. The SAF is the ratio of the emergent photon rate to the corresponding rate if the specimen were perfectly transparent. In particular Dixon considered: 1) a rod (or linear) source viewed end on, 2) a right cylindrical (elliptical cylinder or disk) source viewed along a diameter, and 3) a spherical source viewed along a diameter. In all cases the radioactivity was assumed to be uniformly distributed within the uniform and homogenous material of the solid body. The source to detector separation was assumed to be large in relation to either the size of the detector or the size of the source so that only rays (for all practical purposes) parallel to the direction of viewing are detected. In addition to this far field assumption the photon spectrometer is assumed to confer the conditions of "good geometry" (perfect energy selection) onto the measurement so that only full energy photons are of interest.

Under the specified conditions the SAF, f , may be written as:

$$f(x=0)=1$$

$$f(x)=\Gamma(p+1)\cdot\left(\frac{2}{x}\right)^p\cdot[I_p(x)-L_p(x)]$$

where Γ is the gamma function [4], I_p is the modified Bessel function of order p [5] and L_p is the Modified Struve function of order p [5]. The value of p takes on the value $\frac{1}{2}$ for rod sources, 1 for cylindrical (elliptical) sources and $\frac{3}{2}$ for spherical sources. The parameter x is the "thickness" of the source. Denoting the linear attenuation coefficient by, μ , then x is given by $(\mu\cdot L)$ for a rod source of length L ; by $(2\cdot\mu\cdot a)$ for an (elliptical) cylinder of principal axis $2\cdot a$ (diameter in the case of a right circular cylinder); and by $(2\cdot\mu\cdot a)$ for a sphere of diameter $(2\cdot a)$.

The spherical case is of particular interest because the SAF for a sphere is the lowest of any uniform solid. This is a result of the high volume to surface area quotient. Because of this spherical lumps represent a worst case assumption and can be used to derive bounding assay values.

The cylindrical case is of practical interest because many calibration samples are in the form of small cylindrical containers filled with powder (e.g. PuO₂, U₃O₈ etc...). These are often arranged perpendicular to the detector axis in a geometry which closely approximates far field conditions.

We note in passing that the evaluation of escape probabilities is closely related to the problem of calculating the collision probabilities for external irradiation. Results for certain other shapes are therefore also available [see 8 and references therein] for the case of isotropic incoming irradiation. In general the use of such results for SAF evaluation would therefore correspond to viewing an ensemble of randomly orientated bodies in the far field approximation in order for an integrated over angle (IOA) signal to be obtained.

Evaluation

The SAF for the rod and spherical sources can be expressed in terms of elementary functions for straightforward numerical evaluation. Evaluation of the SAF for the cylindrical source may be achieved by series summation. The functions SAF_rod(x), SAF_sph(x) and SAF_cyl(x) created to evaluate the SAF, for the rod, spherical and cylindrical sources respectively will now be defined in turn. These functions have been implemented in MS Excel™ as user defined functions for ease of use in applications.

SAF_rod(x)

$$x = \mu \cdot L > 0, \quad \text{where } L = \text{length of the rod}$$

$$f = (1 - e^{-x})/x \quad (2)$$

Evans [6] provides a direct derivation of Eqn. (2).

SAF_sph(x)

$$x = 2 \cdot \mu \cdot a > 0, \quad a = \text{radius of the sphere}$$

$$f = (3/x^3) \cdot [x^2/2 - 1 + (1 + x) \cdot e^{-x}] \quad (3)$$

See also Croft [7] for a direct derivation and examples of the evaluation and use of Eqn. (3).

SAF_cyl(x)

$$x = 2 \cdot \mu \cdot a \geq 0$$

where a = radius of the cylinder (half-length of the principle axis in question in the case of an elliptical cross-section viewed along that axis).

The evaluation varies depending on whether x is less than 18.5 or not. This choice was based on convergence properties of the series involved.

If $x \leq 18.5$: Considering the definition and properties of I_1 and L_1 [5] we chose to evaluate $\text{SAF}_{\text{cyl}}(x)$ by series summation. The choice of the summation is conditional on the value of x so that adequate convergence is achieved with a relatively small number of terms.

$$f = \sum_{n=0}^{59} J_n$$

where

$$\begin{aligned} J_0 &= +1 \\ J_1 &= -\left(\frac{4}{3\pi}\right) \cdot x \\ J_n &= \frac{x^2}{n(n+2)} \cdot J_{n-2} \end{aligned}$$

else, if $x > 18.5$

$$f = \frac{4}{\pi} \cdot \frac{1}{x} \cdot \left[1 - \sum_{n=0}^{25} T_n \right]$$

where

$$T_0 = \frac{1}{x^2}$$

and

$$T_n = \left[(4 \cdot n - 1) / x^2 \right] \cdot T_{n-1} \quad (4)$$

For the present purposes (non-destructive assay of waste and scrap for sentencing and safeguards) the proposed scheme for evaluating $\text{SAF}_{\text{cyl}}(x)$ can be considered exact. With suitable floating point precision the expansions themselves are accurate to better than the 5th significant figure. This far exceeds the accuracy of the linear attenuation coefficients of real materials and is therefore not limiting.

Specific Cases

It is of interest to note the behavior of the three functions in the extreme limits of $x \ll 1$ and also as x tends to ∞ . These are summarized in Table.1. Also shown for comparison are the corresponding forms cast in terms of the transmission factor, T , across the body where $x = \ln(1/T)$. These extreme forms can also be of practical interest in forming simple expressions to cover a wide dynamic range by forming a suitably weighted sum of the weak and strong behavior.

Table.1. Limiting approximation for the SAF expressions

SAF Function	Limit $x \ll 1$	Limit $x \gg 1$
Rod γ 's emerging from end	$f \sim 1 - \frac{x}{2} + \frac{x^2}{6} - \dots$ $\sim 1 - \frac{1}{2} \cdot (1-T) - \frac{1}{12} \cdot (1-T)^2$	$f \sim \frac{1}{x}$ $\sim \frac{1}{\ln(1/T)}$
Sphere all directions equivalent	$f \sim 1 - \frac{3}{8} \cdot x + \frac{1}{10} \cdot x^2 - \dots$ $\sim 1 - \frac{3}{8} (1-T) - \frac{7}{80} \cdot (1-T)^2$	$f \sim \frac{3}{2} \cdot \frac{1}{x}$ $\sim \frac{3}{2} \cdot \frac{1}{\ln(1/T)}$
Cylinder perpendicular emergence	$f \sim 1 - \frac{4}{3 \cdot \pi} \cdot x + \frac{1}{8} \cdot x^2 - \dots$ $\sim 1 - \frac{4}{3 \cdot \pi} (1-T) - \frac{(16-3\pi)}{24\pi} \cdot (1-T)^2$	$f \sim \frac{4}{\pi} \cdot \frac{1}{x}$ $\sim \frac{4}{\pi} \cdot \frac{1}{\ln(1/T)}$

For light attenuation, the expansions given in Table.1 shows that simple low order polynomial approximations in x or $(1-T)$ may be used. When the attenuation is heavy the SAF tends to be directly proportional to $(1/x)$ in all three cases. This reciprocal logarithmic dependence on T at this extreme, illustrates that the SAF becomes sublinear with T at low transmission factors.

Self attenuation factors for rods, cylinders, and spheres evaluated as a function of x according to the full expressions developed above are given in Table.2. These results, which agree with the numerical results listed by Dixon, can be used to check independent implementations of the expressions 2, 3, and 4, and also alternative ways of evaluating SAF such as Monte Carlo simulation.

Table.2. SAF as a function of “thickness” x for rod, cylindrical, and spherical bodies.

x	SAF_Rod	SAF_Cyl	SAF_Sph
0	1.0000	1.0000	1.0000
0.2	0.9063	0.9199	0.9288
0.4	0.8242	0.8485	0.8648
0.6	0.7520	0.7849	0.8069
0.8	0.6883	0.7279	0.7546
1	0.6321	0.6768	0.7073
1.2	0.5823	0.6309	0.6643
1.4	0.5381	0.5895	0.6252
1.6	0.4988	0.5522	0.5895
1.8	0.4637	0.5185	0.5570
2	0.4323	0.4879	0.5273
2.4	0.3789	0.4348	0.4749
2.8	0.3354	0.3905	0.4306
3.2	0.2998	0.3534	0.3929
3.6	0.2702	0.3219	0.3604
3.8	0.2573	0.3080	0.3459
4	0.2454	0.2951	0.3324
5	0.1987	0.2430	0.2770
6	0.1663	0.2056	0.2364
8	0.1250	0.1565	0.1817
10	0.1000	0.1260	0.1470
12.5	0.0800	0.1012	0.1185
15	0.0667	0.0845	0.0991
17.5	0.0571	0.0725	0.0852
20	0.0500	0.0635	0.0746
25	0.0400	0.0508	0.0598
30	0.0333	0.0424	0.0499
50	0.0200	0.0255	0.0300

Let us now consider behavior for a particular chemical form. For a given mass of fissile material we may write $m = \rho \cdot V$, where V is the volume and ρ is the density. Suppose the rod has a square cross section of width equal to the thickness, i.e. is a cube.

$$L = \frac{x}{\mu}; m = \rho \cdot L^3$$

$$x = \mu \cdot \left(\frac{m}{\rho} \right)^{\frac{1}{3}} \quad \text{cube}$$

Suppose the cylinder is squat, that is $D = 2a = H$

$$a = \frac{x}{2\mu}; m = \rho \cdot \pi a^2 \cdot 2a$$

$$x = 2\mu \cdot \left(\frac{1}{2\pi}\right)^{1/3} \cdot \left(\frac{m}{\rho}\right)^{1/3} \quad \text{squat cylinder}$$

For the sphere we have:

$$a = \frac{x}{2\mu}; m = \rho \cdot \frac{4}{3}\pi a^3$$

$$x = 2\mu \cdot \left(\frac{3}{4\pi}\right)^{1/3} \cdot \left(\frac{m}{\rho}\right)^{1/3} \quad \text{sphere}$$

Introducing the variable $x_o = \mu \cdot \left(\frac{m}{\rho}\right)^{1/3}$

we can then write

$$x = 1 \cdot x_o \quad \text{cube}$$

$$x = 2 \cdot \left(\frac{1}{2\pi}\right)^{1/3} \cdot x_o \quad \text{squat cylinder, viewed perpendicular to the axis}$$

$$x = 2 \cdot \left(\frac{3}{4\pi}\right)^{1/3} \cdot x_o \quad \text{sphere.}$$

These forms may now be substituted into the limiting expressions presented in Table 1. The result is limiting expressions which may be compared for these particular geometries (the escape path distributions defined by the shape and orientation of the body) for lumps of equal fissile mass. This is instructive since it illustrates the behavior from a different perspective. Often one is interested in the potential for underreporting as a function of mass (rather than physical size) and it is important to recognize and quantify the differences due to the assumptions underlying the model.

Table.1.a. Limiting approximations for the SAF expression cast in terms of $x_0 = (m/\rho)^{1/3}$ so that lumps of the same mass and form can be compared directly.

SAF Function	Weak attenuation limit	Strong attenuation limit
Cube viewed perpendicular to a face	$f \sim 1 - \frac{x_0}{2} + \frac{x_0^2}{6} - \dots$ $\sim 1 - 0.5000x_0 + 0.1666x_0^2 - \dots$	$f \sim \frac{1}{x_0}$
Sphere	$f \sim 1 - \frac{3}{4} \left(\frac{3}{4\pi} \right)^{1/3} \cdot x_0 + \frac{2}{5} \left(\frac{3}{4\pi} \right)^{2/3} \cdot x_0 - \dots$ $\sim 1 - 0.4653x_0 + 0.1539x_0^2 - \dots$	$f \sim \frac{3}{4} \cdot \left(\frac{4\pi}{3} \right)^{1/3} \cdot \frac{1}{x_0}$ $f \sim \frac{1 \cdot 209}{x_0}$
Squat Cylinder viewed perpendicular to axis	$f \sim 1 - \frac{8}{3\pi} \left(\frac{1}{2\pi} \right)^{1/3} \cdot x_0 + \frac{1}{2} \left(\frac{1}{2\pi} \right)^{2/3} \cdot x_0 - \dots$ $\sim 1 - 0.4600x_0 + 0.1468x_0^2 - \dots$	$f \sim \frac{2}{\pi} \cdot \left(\frac{2\pi}{1} \right)^{1/3} \cdot \frac{1}{x_0}$ $f \sim \frac{1 \cdot 175}{x_0}$

From the limiting results summarized in Table.1a, we note that the behavior of the sphere and squat cylinder are quite similar. We note that this is a consequence of the choice of aspect ratio adopted for the cylinder. It follows from the assumption that individual lumps are generally similar in all of their dimensions. The attenuation for the cube of the same mass is slightly (in the context of the overall uncertainty budget for the NDA of drummed waste) stronger than for the other two cases listed. We normally think of the sphere as being the worst case due to it possessing the highest volume to surface ratio of any uniform solid non reentrant body. But this statement should also be qualified by the assumption that the lump is viewed isotropically from all orientations. In the models considered in the table this is NOT the case.

In the present discussion these three cases provide a range of estimates for squat shaped lumps. The range of values reminds us that precise estimates are not possible for real waste items. The shape, size and orientation of the lumps matter. This is true whether the assay system for real waste items is being used to measure unknown drums or whether it is being calibrated. A cylindrical reference source will yield different apparent masses depending on its orientation – for instance whether it is aligned with its axis parallel to or perpendicular to the axis of the drum rotation.

Extensions

Suppose instead of the cube we consider a rod viewed end on where the rod has a circular cross section x we assume length = diameter (as for the squat cylinder).

$$L = \frac{x}{\mu} = 2a; m = \rho \cdot \pi a^2 \cdot 2a$$

$$x = \mu \cdot 2 \cdot \left(\frac{1}{2\pi}\right)^{1/3} \cdot \left(\frac{m}{\rho}\right)^{1/3}$$

$$x = 2 \cdot \left(\frac{1}{2\pi}\right)^{1/3} \cdot x_0$$

The weak approximation for a squat circular cylinder rod viewed end on becomes

$$f \sim 1 - \left(\frac{1}{2\pi}\right)^{1/3} \cdot x_0 + \frac{2}{3} \left(\frac{1}{2\pi}\right)^{2/3} \cdot x_0^2 - \dots$$

$$\sim 1 - 0.5419 \cdot x_0 + 0.1958 \cdot x_0^2 - \dots$$

The corresponding strong attenuation approximation becomes

$$f \sim \frac{(2\pi)^{1/3}}{2} \cdot \frac{1}{x_0} \sim \frac{0.9226}{x_0}$$

The concept of squat rods can be extended to any rod cross section: e.g. an equilateral triangle cross-section.

$$L = \frac{x}{\mu} = a; m = \rho \cdot \frac{a}{2} \cdot \frac{\sqrt{3}}{2} \cdot a \cdot a = \rho \cdot \frac{\sqrt{3}}{4} \cdot a^3$$

$$x = \mu a = \mu \cdot \left(\frac{4}{\sqrt{3}}\right)^{1/3} \cdot \left(\frac{m}{\rho}\right)^{1/3}$$

$$= \left(\frac{4}{\sqrt{3}}\right)^{1/3} \cdot x_0$$

From which we develop the weak attenuation approximation:

$$f \sim 1 - \frac{1}{2} \left(\frac{4}{\sqrt{3}}\right)^{1/3} \cdot x_0 + \frac{1}{6} \left(\frac{4}{\sqrt{3}}\right)^{2/3} \cdot x_0^2 - \dots$$

$$\sim 1 - 0.6609 \cdot x_0 + 0.2912 \cdot x_0^2 - \dots$$

and the strong attenuation approximation:

$$f \sim \left(\frac{\sqrt{3}}{4} \right)^{1/3} \cdot \frac{1}{x_0}$$

$$\sim \frac{0.7565}{x_0}$$

Again, these expressions for the circular and equilateral triangle rods can be compared with the other three shapes directly for a lump of the same mass.

In drawing this section to a conclusion we wish to underscore the fundamental point that the behavior of the SAF as a function of lump mass depends on the assumed geometry of the lump. That is to say, the path length distribution (shape and orientation of the lump) governs the behavior. If we imagine that an ensemble of lumps can be represented by an arrangement of elemental needles then the slab or rod geometry involves only a single needle length. Other bodies and shapes which are viewed from a variety of perspectives (i.e. randomly orientated lumps or lumps assayed in a rotating drum which is also scanned by the HRGS detector systems can be thought of as a distribution of needle lengths. This introduces non-linearity (i.e. non-additivity) into the description and inversion.

Illustration of use

Consider the measurement of ^{235}U by γ – spectrometry in the case of uranium (HEU) metal waste with a ^{235}U weight fraction above 90 weight%. The live-time, background, and matrix corrected net full energy peak, count-rate, R, at a given photon energy will be directly proportional to the nuclide mass, m_0 , present as a modified by the SAF. That is:

$$R = k \cdot m_0 \cdot \text{SAF} \quad (5)$$

where k is the specific count-rate calibration coefficient ($\text{cnt} \cdot \text{s}^{-1} \cdot \text{g}^{235}\text{U}^{-1}$) determined for dilute ^{235}U uniformly dispersed throughout an empty measurement item.

^{235}U decays with the emission of a strong ($0.572 \pm 0.005 \quad \gamma \cdot \text{disintegration}^{-1}$) emission of 185.72 keV [9] and this line is commonly used in quantitative assay work. However, the application of eqn.(5) is hampered by the fact that the SAF is generally not known for waste items. In order to circumvent this limitation one is naturally led to ask whether additional experimental data is available to shed light onto the problem. In the case of ^{235}U there are few viable additional γ -rays. There is a line at 163.33 keV ($0.0508 \pm 0.0004 \quad \gamma \cdot \text{disintegration}^{-1}$) and another of potential interest at 143.76 keV ($0.1096 \pm 0.0008 \quad \gamma \cdot \text{disintegration}^{-1}$) [9]. For the present purposes we shall not consider the x-ray region because this region of the spectrum is often cluttered and the lines do not penetrate waste materials very well. Also we wish to avoid dealing with the discontinuity in the mass

attenuation coefficients in the 98-116 keV region. For reasons of intensity and energy difference (which in turn confers lump size discriminating power) we consider using the 143.76 keV line to derive a self attenuation correction (SAC) factor.

Rearranging Eqn(5) we obtain the following expression for the apparent assay mass, m , in terms of the correct mass (or true) m_0 :

$$m = (R/k) = m_0 \cdot \text{SAF} \quad (6)$$

If we make the simplistic assertion that, at least to first order, the SAC behaves conceptually like exponential attenuation through a slab of appropriately chosen effective dimensions we have:

$$\text{SAF} = \exp(-\mu_m \cdot b) \quad (7)$$

where μ_m is the mass attenuation coefficient and b is a free parameter for the lump distribution under study.

Substituting Eqn.(7) into Eqn.(6) for each of the two energies $E_1 = 185.72$ keV and $E_2 = 143.76$ keV under consideration for ^{235}U and solving for m_0 results in Eqn.(8).

$$m_0 = m_1 \cdot (m_1/m_2)^p = m_1 \cdot \text{SAC} \quad (8)$$

where m_1 is the apparent mass observed at 185.72 keV
 m_2 is the apparent mass observed at 143.76 keV

and, in the framework of the simple model presented we find that:

$$p = 1/(\mu_2/\mu_1 - 1)$$

(μ_2/μ_1) being the ratio of mass attenuation coefficients of the lump material at 143.76 keV at 185.72 keV respectively.

The multiplier SAC has been introduced to represent the Self Attenuation Correction factor. For practical purposes p may be treated as an empirical model parameter the value of which should be chosen by direct experimental work against representative standards. For a wide range of U compounds the U component effectively determines the value of p and consequently if the form of the material is unknown p can be bounded by considering basic interaction coefficients. For example, if we consider to following list of materials to be representative of the range of materials types that might be encountered {U, UO₂, UC₂, UC, U₂C₃, U₃O₈, UO₃, UO₂F₂, UO₂Cl₂, UO₂(CH₃COO)₂, UO₂SO₄.3H₂O, UO₂(NO₃)₂.6H₂O} the value of p increases across the set from a low of 1.141, for elemental U, to a value of 1.219 for uranyl nitrate. Taking the mid range as a reasonable estimate for general purpose use and the half-spread as the corresponding bounding uncertainty we arrive at an estimate for p of (1.18±0.04).

In the present work we have elected to use the theoretical expressions developed for SAF's of slab-like prisms, spheres and cylinders to investigate the utility Eqn. (8) for the case of both individual lumps and distribution of lumps. In doing so the (m_1/m_2) ratio over which useful and reliable correction factors can be determined and applied can be assessed. The SAF expressions developed have also been used to check other numerical methods including Canberra's ISOCS code and an independent in-house Monte-Carlo ray tracing code. These alternative codes offer the flexibility to study a wider variety of lump shapes but given that the shape and size and texture of real lumps is considerably uncertain a more detailed approach is not considered necessary here and the benefit of simple spreadsheet implementation outweighs any gain.

Given measured values of m_1 , m_2 and p along with their associated standard deviation σ_1 , σ_2 and σ_p respectively, the standard deviation σ_0 in m_0 can be evaluated accordingly to Eqn(19) under the assumption that the three terms are independent.

$$(\sigma_o/m_o) = \left[(1+p)^2 \cdot (\sigma_1/m_1)^2 + p^2 \cdot (\sigma_2/m_2)^2 + \left[\ln\left(\frac{m_1}{m_2}\right) \right]^2 \cdot \sigma_p^2 \right]^{0.5} \quad (9)$$

For completeness we note that when more than two gamma-ray rays of known relative intensity are available that the extraction for the SAF from the full data set, rather than from just a single pair, can be based on non-linear least squares fitting making use of Eqn.(7) with an implicit or explicit energy dependence for the mass attenuation coefficient. Also if sufficient lines are available the model can be extended to two components allowing for a fraction of the material to be free from self attenuation effects while the remainder must be corrected. The two gamma-line approach and the variants just described for partial self shielding and multi-line analysis are available in the Canberra NDA2000 software package although by default the μ -values are assumed to scale in inverse proportion to the photon energy.

The simple form of the correction factor, involving only a single parameter, p , is based on the assumption of the exponential attenuation model Eqn.(7). The attraction of this model is that it is independent of the exact shape of the lump and is therefore quite general. Obviously however the exponential attenuation approximation will fail for all lump shapes once they become large. The meaning of large varies on a case by case basis. It is important to realize therefore that the SAC can only be derived reliably over a limit range of (m_1/m_2). It is also important to realize therefore that the uncertainty propagation formula, which assumes that the SAC formula is an exact representation of reality, will underestimate the overall uncertainty in the SAC.

To illustrate this further let us again consider U-metal and uranyl nitrate as the extreme case. For dilute material the (m_1/m_2) ratio is expected to be unity within experimental uncertainty. For a large lump only the external skin contributes to the emergent gamma-ray signal and so the (m_1/m_2) ratio tends to (μ_2/μ_1) which takes the value of 1.88 for U and 1.82 for uranyl nitrate. If the form of the material is unknown then this magnitude of

difference cannot be exploited in any meaningful way even if experimentally the (m_1/m_2) could be determined with adequate accuracy. The restriction on the upper limit of (m_1/m_2) which may be reliably exploited is more stringent however and is related to the breakdown of the simple exponential model to an actual lump. It also requires a subjective choice on what level of correction to bias is 'useful'. This can be immediately appreciated by noting that in the case of U-metal the upper physically meaningful limit of the SAC expression that can be obtained is 2.05 $(\sim(1.88)^{1.14})$. The corresponding value for uranyl nitrate is 2.08 $(\sim(1.82)^{1.22})$. Clearly however the SAC required for a real lump is unbounded. That is to say, under this scheme, after the (m_1/m_2) ratio has saturated there is no way of knowing how much fissile material remains hidden from view in the interior of the lump. To explore this further let us compare the SAC result with the true correction factor for the case of the slab. In this case the SAC results in a systematic under correction with the fractional degree of underreporting increasing monotonically with lump thickness. If we set the criteria for applying the SAC such that the ratio of true correction factor to the that predicted by the power law SAC formula be less than 1.2, then, for elemental U the areal density of the lump must be kept below about 1.116 g.cm^{-2} which corresponds to the (m_1/m_2) ratio being maintained at less than about 1.583. The percentage underreporting of the final assay result will be 16.7% and the true correction factor is approximately 2.03. The corresponding analysis for uranyl nitrate which has a U weight fraction of 0.5861, leads to the conclusion that the areal density of the lump must be kept below about 1.625 g.cm^{-2} which corresponds to the (m_1/m_2) ratio being less than about 1.508. The percentage underreporting of the final assay result will again be about 16.7% and the true correction factor is approximately 1.92. The tools described in the paper together with knowledge of mass attenuation coefficients for the materials concerned (in this case taken from the BNL XCOM database) are all that is required to replicate these results and it is straightforward to extend the calculations to the other lump shapes discussed and to other criteria. Based on considerations such as these it is concluded that a reasonable criterion for the application of the simple SAC algorithm described that the observed (m_1/m_2) ratio be well determined (including allowances for statistical, matrix and other effects!) and lie in the interval [1.00, 1.6]. Because the analysis was based on the use of the slab approximation it is somewhat conservative. As already noted the uncertainty propagation formula developed earlier does not account for the model mismatch and so an additional (empirical) fractional uncertainty contribution amounting to less than 0.2 (as a result of the applicability criterion) should be added in quadrature. [A conservative expression for this additional fractional uncertainty is $((m_1/m_2)^{0.4} - 1)$ although refinements are possible depending on the level of prior knowledge of the items being measured].

Discussion of Further Work

This paper is the first in a planned series in which we intend to build on the concepts introduced here to better quantify the potential for self attenuation corrections in realistic situations and also to present alternative approaches which we have developed that may

prove more robust. In this section we shall therefore only outline our approach to simulating to real cases.

Many physical distributions exhibit inverse dependence on the 'size' of the event. It seems reasonable therefore for a class of waste generation processes to create an assembly of lump activities (masses) each lying in the interval $[a_1, a_2]$ subject to a probability distribution proportional to $1/a$ by sampling $a_1 \cdot (a_2/a_1)^\xi$, where ξ is a pseudo-random number on the interval $[0, 1]$. Sampling schemes can also be created to place the lumps generated at random into the matrix of a waste drum so that matrix perturbed ratios of the apparent masses, following gross matrix corrections, can be extracted. In this way realistic uncertainty estimates of lump corrections can be evaluated.

In the present report we have concentrated on results applicable to single lumps. In practice without additional knowledge of the waste generation process the validity of applying corrections based on these ideas is difficult to address. There is an implicit assumption that all the lumps are similar in size. Large lumps in the interior of a dense item may be masked by matrix attenuation such that the apparent mass ratio is dominated by the 'visible portion' of the item (drum) in the outer regions and so may not appear as saturated as it is. Also the form of the material (shape, structure, chemical composition and enrichment) may result in bias additional. Therefore at present allowance for U-lump corrections is best applied only with extreme caution and only under the jurisdiction of a suitably qualified and experienced subject matter expert.

Conclusion

Expressions for the self-attenuation factors for the rod, sphere, and cylinder are given and evaluated. In particular a series summation method for the calculation of SAF_sph is presented. Limiting forms for the expressions corresponding to weakly and strongly attenuating lumps have also been derived.

The application of the SAF expression has been illustrated in the case of assaying HEU by the use of the 145 keV and 186 keV lines. A simple self absorption correction formula was proposed and exercised. A useful range of applicability was established. The approach can be generalized to other scenarios as required. Extension to other nuclides with suitable pairs of γ -ray lines (e.g. the 129keV and 414 keV pair from ²³⁹Pu) is obvious.

The analytical forms given may be used to test other generalized calculational tools. The extremal expressions can also be used to construct simple approximations to cover a wide dynamic range by forming suitably weighted sums.

In real waste lumps of different shape, different sizes and different chemical form may all be present together. They will be distributed non-uniformly within a possibly

heterogeneous matrix. Therefore any SAC method must also be judged in relation to how robustly it can be applied under these real life situations allowing for variability in the lumps, the spatial distribution and the uncertainty in the knowledge of the gross matrix correction factors. The tools described in this paper are part of a larger program currently in train to evaluate these interrelated factors.

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