

A NUMERICAL APPROACH TO PU GAMMA-RAY SELF-ABSORPTION CORRECTION

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ABSTRACT

Self absorption is a significant source of error in the assay of Pu-bearing waste by gamma-ray spectrometry. Differential absorption of the 129keV and 414keV lines can be used as a means of determining self-absorption and applying a correction. With a view to refining this technique, the behaviour of the 129keV and 414keV lines from ²³⁹Pu as they emerge from lumps of Pu has been studied using a point-kernel point-detector theoretical model for a range of lump shapes, densities and masses. The shapes include right circular cylinders, rods and rotating square plates and the densities range from those typical of PuO₂ to pure metal. The mass ranged studied covers 1µg to 350g.

Variation in shape and density for Pu lumps may be expected to complicate any correction method. However, from inspection of 3D plots of total lump mass against apparent 129keV and 414keV masses it is observed that points on a plot of apparent masses of the 129keV against the 414keV line of constant total mass lie on continuous curves, irrespective of the shape or density of the lump. This observation has been used to derive a lump correction approach by a 3D fitting algorithm to the computed apparent mass data.

A model has been devised to determine the self-absorption corrected mass from the observed apparent 129keV and 414keV line masses. The paper will present a description of the model, results in the form of 3D plots, a description of the fitting method and results obtained for the corrected mass for theoretical samples and for real samples of known total Pu mass for which the 129keV and 414keV lines have been measured. A discussion of the applicability of this method for multiple lumps is presented.

Keywords: NDA, Plutonium, Self-Absorption

INTRODUCTION

During previous tests at Canberra an issue with the plutonium self-absorption correction was highlighted; for our AE 4043/5 Pu sample [1], a consistent over-correction of 50%

was observed. The possibility of inaccurate measured line intensities as the source of the problem was ruled-out by comparing uncorrected 129keV and 414keV assay results with calculated SAC (self absorption corrected) emission rates for this sample. This prompted detailed analysis of the Pu self-absorption behaviour for various lump shapes and materials (densities) and hence the search for a more accurate method. One resolution to this problem is to calculate self-absorption factors using analytical methods [2]. Due to the extreme non-linearity in the nature of gamma attenuation these methods generally require some degree of approximation.

The standard SAC algorithm offered in the NDA2000 data acquisition and analysis suite [3] was developed empirically for the assay of canned radioactive waste containing a distribution of lump sizes. The nature of the empirical approach involved comparing gamma-assay result against calorimetric based assays of the same item. The form of the correction is shown below:

$$M_i = M^o e^{\beta/E_i}$$

where M_i and E_i are the activity and energy of the i 'th line, M^o is the corrected activity of the nuclide and β is a model parameter. This correction can be derived from first principles under the assumptions that the self-absorption is not severe and therefore the method may be less appropriate for single, mass and encapsulated sources. An option to the engine allows for the possibility that only a fraction of the Pu is affected. A further obvious limitation of the NDA2000 approach which shall not be addressed further here is the embedded $1/E$ dependence appearing in the exponent. The motivation behind the present study was to find a more generic approach with broader applicability.

DESCRIPTION OF THE MODEL

To perform these calculations a point-source point-detector model has been used. The model breaks the lump up into a specified number of voxels (volume elements), defined by radial, polar and height integration intervals for cylindrical

lumps, and lateral, depth, height and rotation integration intervals for cuboidal lumps.

The model sets the gamma-ray production in each voxel as the volume of the voxel and computes the probability of the gamma-rays reaching the detector without scattering or absorption. The voxel contributions are integrated over the whole lump and the calculation repeated with the density in the attenuation function set to zero, the ratio of these results giving the SAF (self-attenuation factor) for that gamma-ray line. The scattered component of the gamma-ray flux reaching the detector can safely be ignored where high resolution Ge detectors are to be used in the measurement if the 129keV and 414keV lines since the scattered photons are almost invariably energy degraded to the continuum below the photo-peak.

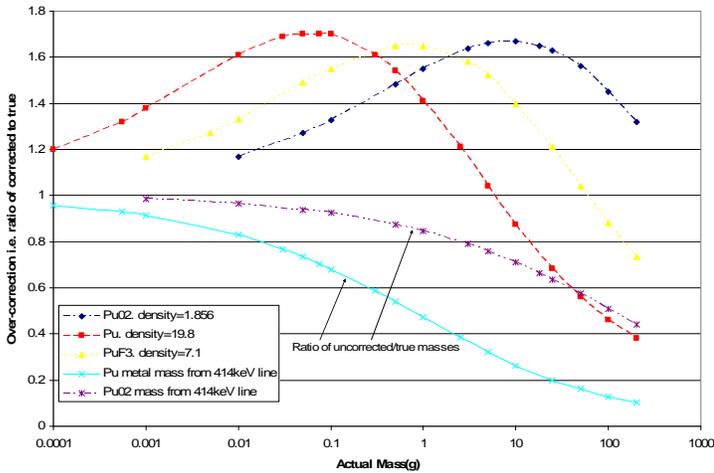


Figure 1: The original SAC mass vs true mass for Pu metal, PuO₂ and PuF₃ cylinders

METHOD

The behaviour of Pu bare cylinders (i.e. not encapsulated within stainless steel) of diameter equal to height have been calculated for three different densities: pure Pu at $\rho = 19.8\text{g/ml}$, PuO₂ at $\rho = 1.856\text{g/ml}$ and PuF₃ at $\rho = 7.1\text{g/ml}$, over the mass range 0.1mg to 200g. In the model it is assumed that the cylinder is viewed in the far field by a detector located along a mid-plane diameter. The over-correction (the ratio of the corrected mass to the true mass for the original NDA2000 engine) is displayed on Figure 1. The graph shows that the original SAC method would not only over-correct by a factor of 1.5 at intermediate masses, but also severely under-correct at higher masses for Pu metal lumps. For example, a 100g Pu metal lump would appear as only 50g, a 50% under-determination. In many routine waste assay situations this is not a serious concern because the maximum single lump size is known to be far smaller than this. For example, in MOX fabrication facilities the waste is restricted to items such as fine oxide powder, grindings and pellet chips. Similarly in a glove box clean-out exercise visual inspection, screening and packing records are often available. However, historical waste from weapons production facilities may pose particular concern.

The graph also shows curves that would be obtained from the 414keV line with no SAC correction. These indicate that for a large range of masses (particularly below 100mg for Pu known to be in the form of oxide) it might be better to offer no correction at all rather than to use the default NDA2000 approach.

The analysis of the uncorrected behaviour of self-absorption for cylinders has been extended to a number of other geometries over the range 1 μg to 350g:

- Cylinders with diameter=height (as shown above)
- Square faced plates with a fixed depth of 1mm and the square side ranging from 0.2mm to 433mm
- Rods with a fixed length of 100mm, with diameters ranging from 25 μm to 49mm
- Cubes of side length ranging 0.3mm to 57mm
- Plates of fixed square face length 3mm, with heights ranging from 0.5 μm to 840mm
- Plates of fixed square face length 30mm, with heights ranging from 0.2 μm to 209mm
- Plates of fixed square face length 100mm, with heights ranging from 0.2 μm to 19mm
- Zero attenuation geometry (129keV mass = 414keV mass = true mass)

RESULTS

Figure 2 shows M_2 (the apparent mass of the 414keV line) plotted against M_1 (the apparent mass at 129keV) for a selection of these materials/shapes with lines of constant true mass in the range 0.001-350g shown in black, creating an interesting and useful 3D surface whose properties may be used to approximate a more accurate mass based upon the measured (observed

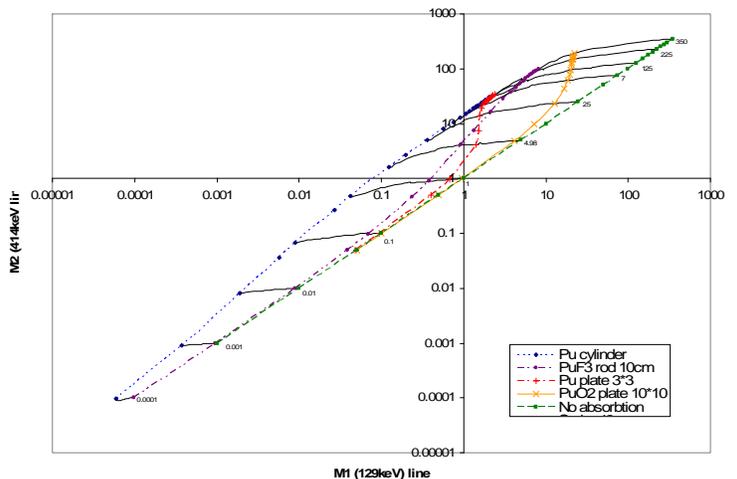


Figure 2: The 414keV line vs the 129keV line masses for a selection of lump shapes and densities

apparent) masses of the 129keV and 414keV lines.

The diagonal line $M_1=M_2$ in Figure 2 is the case of no attenuation, where M_1 , M_2 and the true mass are all equal in value. It is physically impossible for a lump to produce a data point in the region below this line. The curve at the furthest

distance from the no-attenuation line is the case for Pu cylinders (the most attenuating material and shape considered). Tiny massed lumps of Pu that do not experience attenuation will lie on the no-attenuation line. As their mass increases and the 129keV and 414keV lines become more attenuated, the curve migrates towards the highest attenuation curve. As the mass increases further they saturate and the points bunch together on the most attenuating line. No data point should reside beyond this curve.

Based on this database of calculated results a Pu SAC method (a software engine) has been developed to calculate the mass of Pu from the two apparent masses M_1 and M_2 , and has been used to demonstrate the effectiveness of the new self-absorption technique. The new method is described briefly below.

A NEW SAC ALGORITHM

For each material and shape combination, 6th order log polynomial curves for the true mass (M_T) versus M_2 , and M_1 (129keV line) versus M_2 (414keV line) have been approximated using Microsoft Excel™, to obtain the self-absorption corrected mass and M_1 as functions of M_2 . The functional representation is purely empirical. Using these equations, for every M_2 we may take a slice of the surface with varying true mass and M_1 and observe a range of M_1 , M_T pairs which are plotted against one another. By using the measured value of M_1 , and treating the locus between successive points as a straight line, the SAC mass may be found. If the M_2 co-ordinate lies outside the 3D surface a least squares fit is used with the nearest 4 points.

The engine tests the limits of the data to ensure there are enough points to perform the calculation correctly. Since the individual materials' curves are fitted only to the maximum point used in the development stage, the 3D shape is undefined beyond these points (which differ between materials). There must be at least 4 points in order for the program to process the least squares fit, therefore the maximum inputted value of the 414keV line mass is 216.84g. Similarly, there is a minimum limit below which the curve is undefined, which is 0.000097g. Since at very low masses the attenuation is negligible, if the 414keV line mass is below this value the program will accept the inputted 414 mass. In practice the lower limit plays little role since the correction is negligible and the signal will ultimately fall below the detection limits.

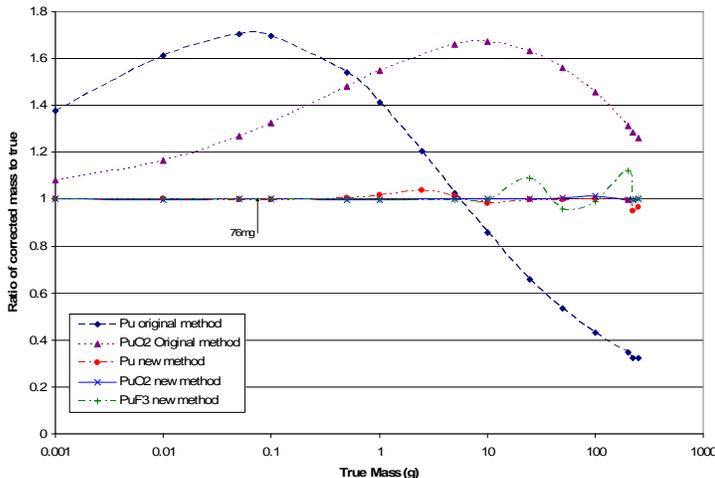


Figure 3: Comparison between the old SAC method and modelled results

COMPARISON TO EXPERIMENTAL RESULTS

This method is compared to the original SAC method in Figure 3 for a mass range of 1mg to 250g which can be considered to be an extreme range for a single lump in most waste assay scenarios. The new engine provides a SAC mass which is much closer to the true mass than the original technique.

The UK ILW/LLW limit for a 60kg gross weight drum is in the range of 50-90mg depending on the isotopics (relative isotopic composition) and the new SAC technique is seen to work accurately without over-reporting in this range which will significantly improve sentencing. The fluctuations at higher masses are due to the least squares approximation being used since the M_2 co-ordinate lies outside the 3D surface, or there not being enough points to plot the curve. It has been decided, therefore, that due to these fluctuations (which increase beyond the 250g limit) and the maximum 414keV input mass, that the engine will display a maximum value of 216.84g. However this limit is not an inherent limitation of the general approach just described. It is rather a pragmatic limit set, at this time, by the amount of modelling work undertaken as part of this study to define the 3D surface.

It can be appreciated that at low masses the counting precision may be limiting but that the corrections are relatively small. As noted above the 414keV apparent mass may be an adequate default value for the true mass. At high masses on the other hand the lump may be close to saturation, meaning that only the surface regions are viewed and that a large fraction of the mass plays little part in the measurement. In this case the statistical accuracy may appear to be much better, but by the same token the demands on the data to distinguish different massed bodies by on small fractional changes in size is also increased.

The uncertainties on the corrected mass arising from the uncertainties in the measured apparent masses, based on the assumption that a single lump is present, have been implemented within the engine using the following equation:

$$\sigma_{m_r}^2 = \left(\frac{\partial m_r}{\partial m_1} \Delta m_1 \right)^2 + \left(\frac{\partial m_r}{\partial m_2} \Delta m_2 \right)^2$$

Δm_n is the inputted uncertainty in the 129keV or 414keV masses from NDA2000. The partial differentials are found by propagating small changes in M_1 and M_2 through the calculation process.

It should be noted that there are also additional uncertainties introduced by the curve fitting technique. However, these errors (shown below) are negligible compared to the errors from the counting statistics etc.

The algorithm and software engine have been used to analyse results from simulated waste drums. Figure 4 shows an example of these results along with their 1 sigma errors bars for a 5g AE4043 sample located at various heights and radii within an otherwise empty 205 litre drum. The graph shows that the algorithm performs well at various positions within the drum. As an example, the height 2 and radius 2 (2/3 radius and 2/3 height) case would be observed as 2.24 +/- 0.02g in NDA2000 without any self-absorption correction.

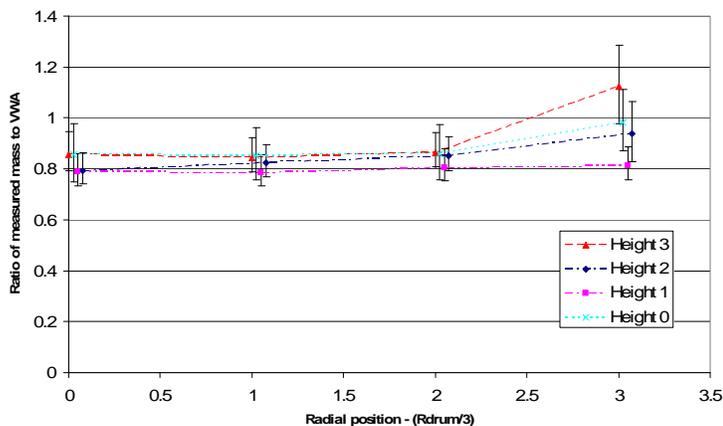


Figure 4: Spatial variation for 5g Pu sample in an empty drum.

EXTENSION TO MULTIPLE LUMPS

The extension to multiple lumps has been investigated. Due to the high degree of non-linearity observed in the model, the combination of a series of smaller lumps was expected to produce a smaller self attenuation factor than that expected for a single large lump of equal mass and therefore the model was expected to underestimate the total Pu in a sample.

The apparent 129keV and 414keV masses for a number of material/shape combinations were summed and used as input for the algorithm to see how it would behave for multiple lumps. Table 1 shows these lumps combinations along with the calculated true mass and the true to SAC mass factor mass.

As shown, the method appears to work well for multiple lumps of various compositions.

Table 1: Example multiple lump results

Material type	Lumps	Total Mass	SAC M_T	True/SAC Mass Ratio
Pu cylinder	0.1g x 10	1g	1.12	0.895
Pu cylinder	10g x 10	100g	128.02	0.781
Pu cylinder	25g x 10	250g	270.15	0.925
PuO2 cylinder	1g x 10	10g	10.30	0.971
PuO2 plate	0.1g x 1	1g	1.04	0.957
PuO2 plate	25g x 5	125g	124.77	1.002
Mixed		25g	20.95	1.191

CONCLUSIONS

In this work we have re-examined how the self absorption correction may be derived. An empirical approach was adopted where the nature of the empirical study involved calculating the behaviour of a large range of lump types (shapes, compounds and densities) and letting nature reveal the correlations with

promise. The results of these calculations have been fitted to a numerical SAC algorithm which has been developed into a functional self-absorption engine for Pu.

The correction method has been applied to lumps that are uniform and is based on the use of a single pair of lines. In practice the majority of drums will contain multiple lumps with a distribution of lump sizes. It is planned to study this effect by numerical simulation.

The method (to be reported) has also been successfully extended to the case of U using the 186keV and 1001keV lines for those cases where the $^{235}\text{U}:$ ^{238}U ratio is known or can be determined. Also the application of the method in real waste drums with inhomogenous matrices is being analysed.

It is recognised that implementation of a SAC in practice is a challenging undertaking. No method can be viewed as perfect. The expectation for how the SAC can improve the quality of the assay must therefore be realistic. One expression of this is in the uncertainty estimate assigned to the corrected mass. This must include the appropriateness of the underlying model for the unknown situation at hand. For example whether a single lump or a distribution of lumps may be the cause of the apparent mass discrepancy and whether all of the lumps present may be reasonably treated as being of the same shape, composition and density or not. Also the lump correction must be robust and quantified against imperfections in the gross matrix correction factors applied such as may be introduced by localised shielding effects. The latter situation could arise if a lump of fissile material had been pre-packaged before being placed into the disposal drum.

REFERENCES

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