Self-Absorption Corrections for Plutonium Measurements With the Tomographic Gamma Scanner – 17325

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ABSTRACT

The Tomographic Gamma Scanner (TGS) is a Non-Destructive Assay (NDA) instrument that combines high-resolution gamma spectroscopy with low spatial resolution three-dimensional image reconstruction in order to reduce bias and improve precision for measurements of drummed waste. The TGS technique is especially suited to measure containers with non-uniform source distributions and low-to medium-density heterogeneous matrices, compared with traditional NDA methods.

Canberra recently built, installed, and commissioned a TGS system, intended primarily for the measurement of plutonium (Pu) bearing waste, at a European nuclear fuel reprocessing facility. Pu assays using the TGS technique present technical challenges distinct from those typically encountered in traditional LLW applications. Among these is the need to apply corrections for self-absorption effects that occur on scales smaller than the TGS spatial resolution, and can compromise the assay result if not included in the TGS imaging methodology. We present a brief review of the Self Absorption Correction (SAC) technique, and discuss the complications involved with its application to the TGS methodology with examples from our recent experience.

This work continues a Canberra history of further developing and refining a technique to solve emerging problems, having commercialized the original methodology by partnering with the research institution that pioneered it. The TGS and the SAC methodologies were originally developed by Los Alamos National Laboratory (LANL) and John Fleissner respectively, and this work illustrates how Canberra has implemented an advancement of these techniques in a new TGS system.

INTRODUCTION

The Tomographic Gamma Scanner (TGS) is a nod-destructive assay (NDA) instrument for the identification and quantification of radioisotopes in drummed waste. The TGS technique provides improved accuracy over traditional gamma-ray waste assay methods. By combining High Resolution Gamma Spectrometry (HRGS) with low spatial resolution imaging on a three dimensional grid of discrete volume elements or “voxels”, attenuation corrections can be applied to the emission data on a voxel-by-voxel basis to better account for source and matrix heterogeneity than in other gamma waste assay techniques. The method has been fully described previously in the literature [1].

Whenever HRGS methods are applied to the assay of high-density radionuclides such as plutonium, self-absorption effects due to lumps of active material can pose a source of severe bias in the final mass quantification. While the TGS method is specifically designed to correct for the variability of attenuation effects due matrix heterogeneity, it does so on roughly the scale of the image voxel size. Lumps of plutonium significantly smaller than the resolution of the TGS (*e.g.* 1 voxel is approximately $[4.9\ \text{cm}]^3$ for a 120 liter drum using the standard 10×10 voxel per layer resolution), will not be properly corrected for by the TGS transmission image reconstruction, leaving some residual fraction of the attenuation uncorrected. In plutonium assays, metallic lumps as small as 1 mm are typical and can produce substantial attenuation because they are essentially invisible to the transmission imaging technique.

The energy dependence of the attenuation can be exploited to identify assay data with significant lumping, and to correct the results accordingly. Figure 1 shows an example of the effects of self-absorption on the reconstructed Pu-239 masses for a set of several gamma-ray lines measured in one TGS assay in the 129 to 451 keV region. A review of various approaches to Self-Absorption Correction (SAC) can be found in [2].

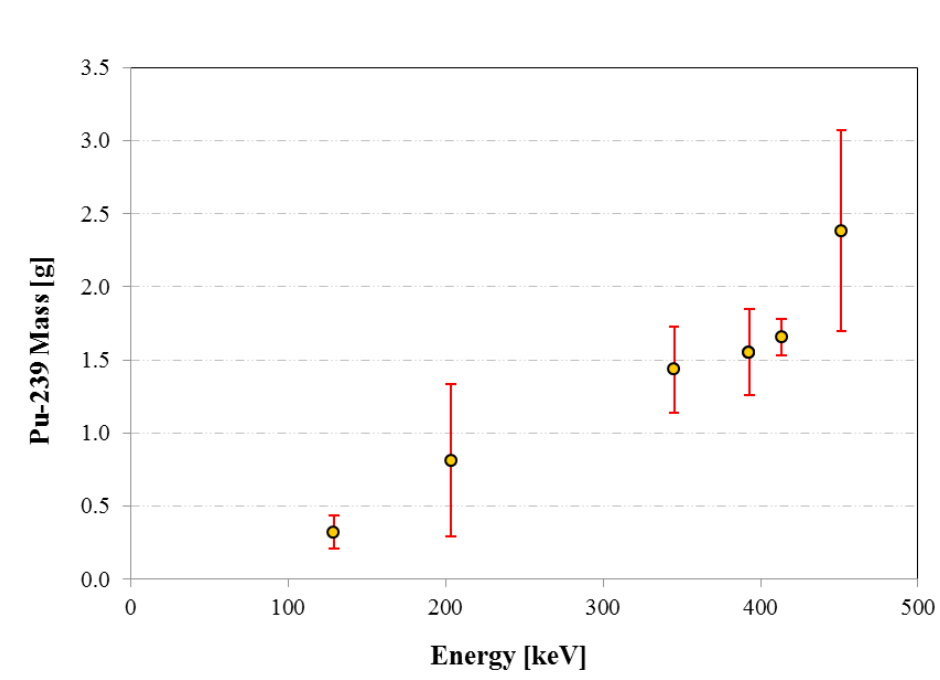

Figure 1 Example of experimentally recovered mass vs. energy for observed Pu-239 gamma ray lines

In the present work, the application of a post-analysis SAC methodology is described for a TGS system recently deployed to a European fuel reprocessing facility for the measurement of Pu bearing drummed waste. Several variations of underlying the attenuation models are evaluated.

METHODOLOGY

Overview

To correct for the self-absorption in plutonium, we apply an attenuation model based on first principles, as well as some straightforward assumptions which we describe below, following more or less the method attributed to John Fleissner at the Rocky Flats plant in the U.S. A similar methodology is described by Prettyman in [4]. The most general form of the attenuation model employed in this method is given by:

$$m_i = m_0\left(\alpha + (1-\alpha)e^{-\beta/E_i^n}\right). \quad (1)$$

This model requires a nonlinear fit to obtain four parameters (α, β, m_0, and n) from the set of masses m_i obtained from the measurement of multiple plutonium gamma rays at different energies E_i. These parameters represent respectively the un-attenuated mass m_0, the fraction of the mass not subject to self-absorption α, as well at the effective attenuation coefficient of the self-absorbing material which is parameterized in terms of a power law,

$$\frac{\mu}{\rho}(E) \cdot \rho \cdot T = \frac{\beta}{E^n}. \quad (2)$$

Here ρ is the density in g/cm^3, and T is the effective thickness of the self-absorbers in cm. $\mu/\rho\,(E)$ is the energy dependent mass attenuation coefficient for the self-absorbing material, which is postulated to follow a simple inverse power law over the energy region of interest. Fitting all four parameters requires that a minimum of four gamma lines can be measured with sufficient statistics to obtain a good fit.

Technical Challenges

In attempting to apply this formula in the context of TGS assays, a number of technical challenges are encountered. The foremost challenge is that, because of limited assay time and matrix attenuation effects, it is often the case that fewer than the minimum of four lines will be observed the necessary precision.

A further complication is the fact that the form of the attenuation model in equation (1) implies two assumptions: first, that all of the self-absorption can be ascribed to lumps of the same size and density; second, since the TGS methodology combines data from multiple measurements taking different views through the assay item, that the lumps must be spherical, so that the attenuation thickness is unchanged in different views. To the extent that either of these implicit assumptions are violated by the reality of the assay item, our attenuation model is no longer strictly correct, since in general it is not true that

$$\sum_i e^{-x_i} = e^{-x_{eff}}. \quad (3)$$

While these assumptions are unlikely ever to be exactly met in a real measurement, we trust that the measurement conditions can still be reasonably approximated by a single effective exponential decay model.

A final technical challenge, specific to the TGS, is that by design the TGS method corrects for absorption effects in the matrix. For large self-absorbers, approaching the voxel size, the self-absorption effect may already be partially corrected by the transmission image.

For all of these reasons, it is imperative that the SAC should not be applied blindly; expert review is always recommended.

Implementation

The SAC engine currently implemented in NDA 2000 was historically intended for use with the Segmented Gamma Scanner (SGS) and/or Q^2 methods, both non-imaging methods that are expected to perform assays on higher density drums than are typically recommended for the TGS. In these applications it is not uncommon that only two Pu lines may be usable (129 and 413 keV). Thus, two simplified versions of equation (1) were implemented in the NDA 2000 software itself. In the first version, the exponent of the energy *n* set to unity, so that the attenuation model becomes

$$m_i = m_0\left(\alpha + (1-\alpha)e^{-\beta/E_i}\right). \quad (4)$$

This form still requires at least three lines to be detected, however. If the minimum number of lines are not detected, the software automatically defaults to the second version, in which the non-attenuated mass fraction α is additionally set to zero, thus requiring only two lines:

$$m_i = m_0 e^{-\beta/E_i}, \quad (5)$$

Setting the energy exponent parameter n to unity will almost universally overestimate the unattenuated mass m_0. In many applications, this assurance of a conservative error is desirable behavior, and for that reason this is deliberately selected in the NDA 2000 software. The more significant the self-absorption effect, the greater the overestimation of mass will tend to be.

For applications in which higher precision estimates of plutonium mass are a priority, a tradeoff against the risk of underestimating the mass can be made by applying the SAC in offline analysis, keeping the power-law version of the exponential decay model as originally presented in equation (1). For the measurements reported here, this offline analysis approach was utilized. In addition, for all of the measurements reported here, it was observed that the un-attenuated mass fraction α never differed from zero by an amount greater than it's uncertainty, and that including that additional parameter in the fit significantly increased the fitted uncertainty in the other parameters. As a result, the attenuation function ultimately used had the form

$$m_i = m_0 e^{-\beta/E_i^n}, \quad (6)$$

with the energy exponent n set to a fixed value greater than unity which was derived on the basis of a-priori process knowledge. This retains the robustness of the truncated fit given in (5) in the sense that it can still be fit with only two good lines, while vastly improving the quality of the correction compared with the $n = 1$ version.

One additional SAC form worth mentioning is the so called “infinite energy extrapolation”, which can be understood as a first-order Taylor series expansion of the simplest exponential form of the attenuation function given in equation (5). In this form a linear fit of the mass *vs.* the inverse energy is performed for two or more detected lines, using

$$m_i = m_0(1 - \beta/E_i)\,. \tag{7}$$

This method has been found to give good results particularly in cases where the attenuation is fairly small [3]; it tends to over-estimate the mass less than the exponential version in (5) since it truncates higher order terms which tend to have a net additive effect.

ANALYSIS RESULTS

The performance of the SAC as described in the previous section is evaluated here for two different test items. The first test item consisted of a well-quantified sample of PuO_2 powder in a small, cylindrical plastic container suspended near the center of a 120 L drum. This item was specifically constructed as a calibration standard for waste assay systems although it is one that is not particularly favorable to the TGS methodology, as we will describe below. The second test item consists of PuO2-contaminated air filter material, loosely packed into a 120 L drum. This drum more closely approximates the real waste stream items expected for assay by this TGS system.

In the discussion below, Pu-239 mass results for the uncorrected analyses, which utilize a simple weighted mean of all detected lines, are compared with corrected mass results using: the “power-law” exponential attenuation model in equation (6), the “1/E” exponential attenuation model in (5), and the linear “infinite-energy extrapolation” in (7).

Assay Item One

Five assays of the first assay item, consisting of a PuO_2 cylinder suspended near the center of an empty 120 L drum, were made with different assay times ranging from just under two hours to 25 hours. The uncorrected Pu-239 mass results are listed in Table 1, along with the corrected mass results using each of the various attenuation models. The actual Pu-239 mass was previously determined by mass spectrometry to be 2.31 g. No uncertainty was given for this value, but typically mass spectrometry uncertainties are expected to be small compared with those of gamma measurements, on the order of 0.1% or so.

As can be seen in the plot of these results in Figure 2, there is significant residual attenuation in the uncorrected mass values which is not accounted for by the TGS analysis; the uncorrected results are 88% low on average compared to the known mass value. In this high attenuation case, the simple 1/E exponential decay model is seen to overcorrect for the self-absorption significantly, by an average of 65%. Better results are obtained from both the linear infinite energy extrapolation and the $1/E^n$ or “power law” exponential model. In both models, the un-attenuated

mass is underestimated. The power law method understates the true mass by 20% on average, while the infinite energy extrapolation results are around 10% low.

Assay	Total assay time [hrs]	Uncorrected Pu-239 Mass [g]	1/E model SAC Mass [g]	Infinite Energy extrapolation SAC mass [g]	Power Law model SAC mass [g]
1	1.875	0.21 ± 0.02	4.08 ± 0.68	2.04 ± 0.22	1.86 ± 0.21
2	11.25	0.25 ± 0.01	3.50 ± 0.30	1.96 ± 0.11	1.72 ± 0.10
3	5.825	0.32 ± 0.02	3.98 ± 0.39	2.11 ± 0.13	1.93 ± 0.12
4	25	0.27 ± 0.01	3.94 ± 0.26	2.05 ± 0.08	1.86 ± 0.08
5	1.865	0.39 ± 0.04	3.50 ± 0.55	2.21 ± 0.23	1.91 ± 0.21

Table 1 Assay results for PuO2 cylinder suspended in center of 120 l drum.

This item represents a particularly challenging case for the TGS, for multiple reasons. For one, the size of the PuO_2 containing cylinder near the center of the drum is on the order of centimeters, approaching the voxel size and thus the TGS resolution. It is therefore to be expected that some of the attenuation due to the active material itself may already be partially corrected by the TGS transmission image, so that as a result the exponential power selected on the assumption of a pure PuO_2 sample may not be strictly correct. Furthermore, it is known that a sample placed on the axis of the drum presents a worst case scenario for the TGS analysis, due in part to the apparent spreading of the observed activity amongst the four center voxels. An overall low bias is typical in this unfavorable scenario, even for simple cases of single-line nuclides in non-self-absorbing configurations.

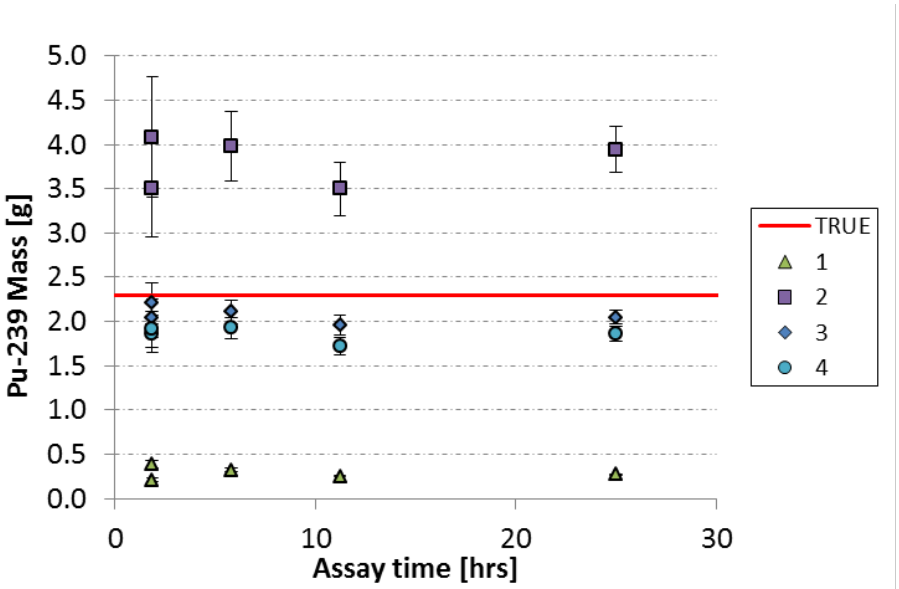

Figure 2 Measured Pu-239 masses for assay item one, by assay count time. The data series' shown represent the uncorrected (1), 1/E exponential SAC (2), Infinite-Energy Extrapolation SAC (3), and Power-law exponential SAC (4). The "true" Pu-239 mass is indicated by the horizontal line.

Assay Item Two

The second test assay item, consisting of PuO_2 laden filter paper material loosely packed in a 120 L drum, was measured once for a total assay time of 13.25 hours. The resulting uncorrected Pu-239 mass is given in Table 2, along with the SAC results using each of the attenuation models indicated previously. These results are compared with the "true" Pu-239 mass in Figure 3.

In this case, the "true" Pu-239 mass was previously determined by two methods: gamma assay using Canberra ISOCS efficiency modeling, and by passive neutron coincidence counting. At the request of the facility, we report here only the relative recovery obtained by the various methods.

SAC Method	Recovery	Unc.
Uncorrected	0.780	0.013
1/E	1.252	0.042
Power Law	1.021	0.021
Infinite Energy	1.117	0.029

Table 2 Pu-239 mass values using various SAC models.

For this assay item, the uncorrected result is 22% low of the true mass; while the result of the 1/E exponential attenuation model is 20% high. The linear infinite energy extrapolation, which was 10% low for the high-attenuation case previously described, is here 9% too high. The $1/E^n$ "Power Law" is seen in this case to give the best result, 2% low of the nominal true value and within the 1-sigma error bars of both the measurement and the established mass. It is worth noting that this represents an overall more favorable assay configuration for the TGS, where the active material is distributed throughout the drum rather than being concentrated near the center as in the previous example.

CONCLUSIONS

The application of Self Absorption Corrections to gamma ray measurements of plutonium bearing waste is a challenging topic generally requiring expert analysis to obtain good results. There are a number of approaches which can be taken, each of which may have varying degrees of suitability for different measurement applications. Methods requiring larger numbers of parameters to be fit tend to be unreliable for TGS assays due to limitations of statistical precision and often a limited number of observed lines. If these fits can be performed at all, they tend to yield parameter values consistent with simpler versions of the model (e.g., no unattenuated mass component, or exponential power equal to unity) but with much higher uncertainties. Restricting the number of free parameters to be fit based on a-priori knowledge or by adhering to a conservative limiting case can improve the reliability of the results. We have shown that the $1/E^n$ exponential model with n judiciously chosen can provide excellent results under reasonable assay conditions. By setting the power parameter n to unity one obtains a reliably high (and thus conservative) estimate which may be desirable for some applications. Using this

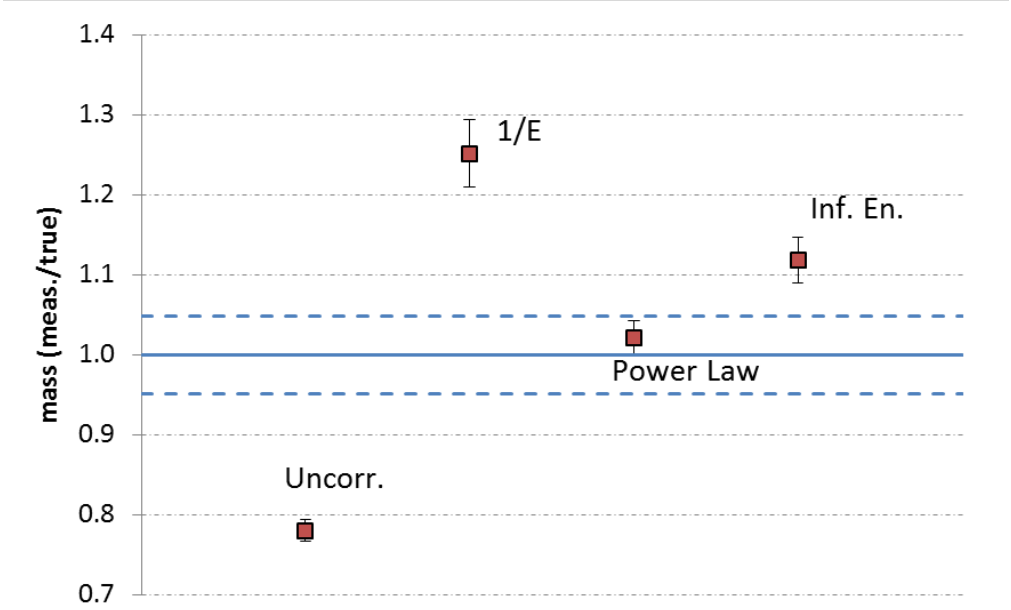

Figure 3 Relative Pu-239 mass results for filter paper drum, compared to previously established "true" value. Uncorrected mass is shown along with results of various SAC models: the 1/E exponential attenuation model, the power law exponential model, and the linear infinite energy extrapolation. The dashed lines indicate the uncertainty in the "true" mass value.

model the overestimation will tend to increase with the degree of self-absorption present. The linear extrapolation to infinite energy tends to over-estimate the mass by a more modest amount when correcting for low to moderate attenuation, but can under-estimate the mass at very high self-absorption. Care should be taken to select the appropriate correction approach for the assay configuration and requirements at hand.

REFERENCES

[1] R.J. Estep, *Assay of Heterogeneous Radioactive Wastes by Low-Resolution Tomographic Gamma Scanning*, Winter Meeting of the American Nuclear Society, 11-15 November 1990, Washington, DC, USA, Pub. as Transactions of the American Nuclear Society; 1990; vol.62, p.178-80.

[2] S. Croft, R. D. McElroy Jr., S. Philips, R. Venkataraman, and D. Curtis, J. Radioanal. Nucl. Chem. 276 3 (2008) 667-683

[3] R. Venkataraman and S. Croft, Nucl. Instrum. and Meth. A 505 (2003) 527

[4] T. H. Prettyman, L. A. Foster, and R. J. Estep, *Detection and Measurement of Gamma-Ray Self-Attenuation in Plutonium Nuclides*, 37th INMM, 1996