Gamma-ray Self-Attenuation in Performance Demonstration Program Sources - 10480

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ABSTRACT

Encapsulated reference sources containing known amounts of special nuclear material (SNM) are routinely used to verify performance of Non-Destructive Assay (NDA) neutron and gamma-ray spectroscopy instruments used to measure radioactive waste. These artificial items are unlikely to be good physical analogs of the SNM present in the waste containers. In order to evaluate whether the mismatch is consequential or not it is necessary to understand two things: first the general nature of the SNM in the waste stream, and second the influence of self-attenuation and absorption of gamma rays in the reference source. In this work, we consider the second aspect of this problem using for illustration a type of Performance Demonstration Program (PDP) source. In particular the attenuation for key Pu lines are calculated as a function of mass loading and orientation. The results are discussed in the context of a typical 55 US gal. drum assay system – the Q^2 which has three high-purity germanium detectors evenly spaced and mounted close to the drum wall parallel to the axis. Data is acquired while the drum rotates. In this example, we demonstrate that the use of the WRM sources to represent a uniform "non-lump" container can, if uncorrected, lead to differences of 30-45% at 414 keV to factors of 2-6 at 129 keV, where the degree of difference is related to the amount of plutonium within the WRM source. The quantified understanding of these influences, allows one to demonstrate the overall performance of a system in the context of possible waste conditions that extend beyond standard system calibrations. Consequently, PDP program measurements stress NDA systems and results in improved performance and greater understanding of the total uncertainties in a given measurement.

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INTRODUCTION

Traceable Non-Destructive Assay (NDA) Working Reference Materials (WRM) are used as part of the qualification and approval process required before measurement systems can be used to quantify nuclear materials suitable for final disposal at the Waste Isolation Pilot Plant (WIPP) [1]. Successful participation in the Performance Demonstration Program (PDP) [2,3] is a requirement. The preparation and certification of such PDP sources is a complex task. An understanding of the source construction can be gained from the description given by Wong and Marshal [4]. The resulting assets are of enormous value in evaluating the performance of various kinds of instruments against different simulated waste forms.

In a recent article [5], we noted that the calibration reference sources are of a different physical structure to the Pu WRM sources that are typically used at waste handling facilities. This introduces a mismatch between the apparent efficiency versus energy curves due to the attenuation in the material of the sources and their containment. By convention the factory calibration is likely to have been made assuming that the source is homogeneously distributed within a uniform matrix of a similar composition and density as the unknown sample. Pu sources of the PDP type are made by dispersing plutonium dioxide in diatomaceous earth and sealing the blend in a substantial capsule. For a particular (65 g Pu) source we performed measurements which allowed us to construct an accurate mathematical model of the source attenuation [5]. In this work we extend these previous calculations, benchmarked as they are, to consider the impact of a calibration intended to model a uniformly distributed source activity compared to the PDP test geometry.

It is understood that PDP test measurements should be taken in the context of validating a system under conditions that are expressly different from the calibration conditions, such that the overall performance is evaluated beyond the simple validation of calibration. It is also understood that PDP test measurements do not necessarily represent the performance of a system under all possible conditions. Therefore, it is necessary to quantifiably understand the overall test conditions so that systems can be fully evaluated in the full context of their designed performance.

We consider here the dependence of the source attenuation on orientation. Usually the Pu WRM sources are used with the cylindrical axis vertical, but depending on where they are in relation to the detector(s) the range of slant angles through which the gamma rays must pass varies. For illustration, in this document, we will use the example of a 55 US gallon drum being measured in a Q^2-style assay system [6], but these results should be generally applicable for any NDA system or method with rotating drums. We illustrate how these test conditions improve the overall performance by requiring advanced analysis techniques that improve the accuracy of the systems, and further motivate innovative approaches to handling complex waste streams.

MODEL DEVELOPMENT

For the present study, we investigate the consequence of utilizing WRM sources as part of a PDP qualification process, by comparing various sensitivities utilizing mathematical modeling techniques to compute distributed source efficiencies and gamma-ray attenuation through the materials. We use the In-Situ Object Counting System (ISOCS) [7] and Laboratory Sourceless Calibration Software (LabSOCS) [7] to define the source geometries and compute efficiencies. These software packages offer a solution to this problem by using semi-analytical techniques to accurately determine the gamma-ray efficiency for virtually any source configuration that is routinely encountered [8].

The base model of the WRM comes from descriptions by Wong and Marshall [3]. The well-quantified plutonium dioxide material of the source is uniformly mixed with diatomaceous earth (DE) and doubly encapsulated in cylindrical stainless steel canisters. For the purposes of this analysis, we adopt the composition of DE in weight per cent as SiO_2 (86.0%), Na_2O (5.0%), MgO (2.5%), Al_2O_3 (2.5%), Fe_2O_3 (2.7%) and CaO (1.3%), with an estimated density of (0.45±0.05) g/cc. The density of PuO_2 crystal is assumed to be 11.45 g/cc [4]. The differential self-attenuation in the individual particles of PuO_2 may be important depending on the distribution of grain size; however, the bulk density of the powder is considerably reduced by distributing the particles in the DE substrate.

The capsule material is 304 stainless steel. The adopted composition is by weight percent is Fe (69.5%), Cr (19.0%), Ni (9.5%), and Mn (2.0%). The internal diameter is 4.32 cm and the side wall (considering both the inner and outer capsules) is 0.28 cm thick. The base thickness is 0.53 cm thick and the lid of the PuO_2-DE blend is 1.04 cm thick. There is one 0.64 cm thick 4.76 cm diameter graphite felt frit at the top of the cylindrical space to hold the PuO_2-DE blend in place; The weight of the graphite piece is (0.91±0.3) g (i.e. the apparent carbon density is quite low (0.080±0.026) g/cc). The length of the PuO_2-DE mixture (between the base & graphite cap) is approximately 20.65 cm giving a volume of about 302 cc (within a few cc subject to the degree of compression of the frit).

This model was validated in Ref. [5] by comparing the modeled efficiency to the measured efficiency for a 65 g (Weapons Grade Pu) WRM source. In this case the PuO_2-DE mixture was modeled as a uniform matrix of 33% PuO_2 by mass to DE with an average density of 0.66 g/cc.

For PDP certification, sources of the above configuration are inserted in to specially designed drums that have three open tubes to contain the WRM sources. These tubes are located along the central axis of the drum and at radii approximately 50% and 80% of the drum radius respectively. Up to three WRM sources can be inserted in each tube with approximately 7 cm spacing between each source. Thus a total of nine WRM samples can be placed within a PDP certification drum.

In this study, we consider a Q^2 drum assay configuration. In this configuration, the drum to be assayed is placed in front of three shielded but uncollimated high-purity germanium detectors placed with their end-caps about 6 cm from the drum wall and aligned vertically

with the central detector approximately at the center plane of the drum and the other two detectors approximately 36 cm above and 36 cm below the middle detector respectively. Between the detector end-caps and the drum are additional absorbers of 0.11 cm 304 stainless steel and 0.08 cm of PVC. The drum is on a rotator so that radial biases are averaged across the entire rotation. The spectra from each of the three detectors is summed and then analyzed. A Q^2 system is typically heavily shielded to reduce ambient background for maximum sensitivity to drum activities. This shielding does not reduce the efficiency, and therefore is not relevant to this exercise. The position of the WRM sources in the test drum, with respect to the Q2 detector locations is presented in Figure 1.

An advantage of having a physically-based mathematical approach is that it is possible to quickly compute "realistic" efficiencies for configurations that are difficult to produce physically. In this paper we consider the cases in which the WRM is loaded with a 33% PuO_2 / 67% DE 0.66 g/cc matrix (as in Ref. [5]) to represent a highly-self-absorptive Pu sample, a sample with 100% DE at 0.46 g/cc to represent the attenuating conditions of sample source container only, and an unattenuating configuration (all materials set to air with density 0.0013 g/cc) to represent the geometric efficiency of each of the source positions. A fourth configuration in which activity is uniformly distributed throughout a non-interfering drum volume is computed to represent a typical (idealized) calibration geometry. These configurations are summarized in Table 1. In this table are identified names that will be used in the following discussions to simplify descriptions.

For these simulations, each of the detector-to-WRM source positions are simulated individually, and consequently effects related to the shielding of one source by another is not considered, but this limitation is not considered to be significant. To show the effect of drum rotation on these non-uniform sources, the efficiency was computed for each detector for each of the six outer radial source positions (3 at 50% and 3 at 80% drum radius) at drum rotation angles of 0, 45, 90, 135, and 180 degrees. Zero degrees corresponds to the case with the outer radial source position closest to the detector. The efficiency at source positions 225, 270, and 315 degrees are determined from the symmetry with 135, 90, and 45 degree positions respectively. The efficiency for the three detectors for three central axis source positions were also individually computed. Because of symmetry no rotation is necessary for these three positions. In this paper, we focus only on the deviations due to the WRM source packaging, and therefore do not consider any interfering matrices or the internal source support structures between the sources and the detectors.

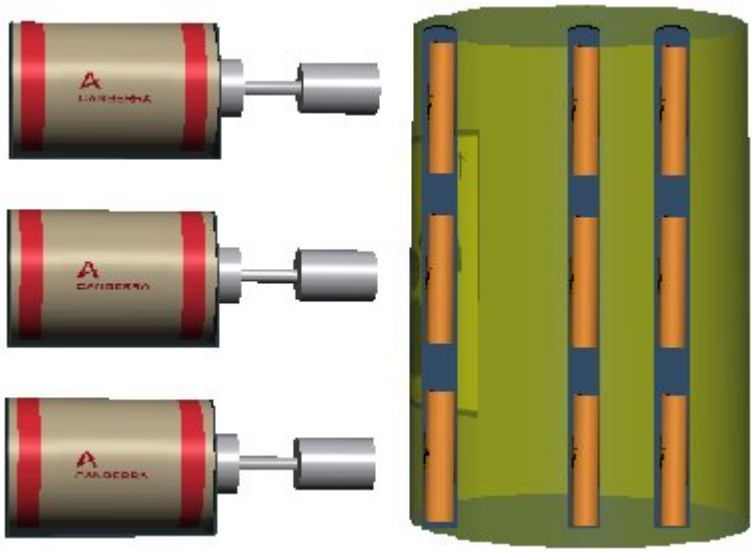

Figure 1. Schematic figure illustrating the configuration of the three germanium detectors in a Q^2 system with respect to the assayed drum. Also shown are the relative locations of the nine WRM sources (orange cylinders) within a PDP certification drum. The picture represents the zero degree rotation position. The scales are approximate.

Table 1. Summary list of the model configurations used in these computations.

Source Name	Source Form Factor	Source Composition	Density	Comment
PuDE	WRM	33% Pu 67% DE	0.66 g/cc	65 g Pu source with Canister
DEonly	WRM	100% DE	0.45 g/cc	Canister only
AIR	WRM	Air	0.0013 g/cc	Only geometrical effects
DRUM	Drum	Air	0.0013 g/cc	Typical calibration geometry

RESULTS

To understand the influence of the WRM form factor on the gamma-ray efficiency we consider the computed efficiency of the attenuating model (PuDE or DEonly) with respect to the non-attenuating model (AIR or DRUM). These ratios are reported in terms of "Correction Factors" that correspond to the multiplicative factor necessary to return the attenuated model value to the unattenuated value; consequently we report the inverse of the attenuation: $(\varepsilon_{attn}/\varepsilon_{air})^{-1}$. In this discussion, we consider the influence of rotation angle, source height, and finally the full volume averaged efficiency.

Effect of Rotation Angle

The correction factor for the WRM when the source is in the outer radial position (80% or the drum radius) and the drum is at different rotation angles is presented in Figure 2. In this figure, the correction factor for each drum rotation angle is computed, $(PuDE/AIR)^{-1}$,

and plotted relative to the correction factor for the source position at the 180 degree drum rotation angle. This 180 degree position is the farthest from the detector, and consequently has the shortest path lengths through the PuDE matrix and WRM canister. As the WRM rotates closer to the detector, the detector presents a larger solid angle with respect to the source. While the absolute efficiency increases at the closer positions, the larger scattering angles are more attenuated resulting in larger correction factors for source positions closer to the detector. In this particular example the correction factor is 1.15 at the 0 degree position compared to 1.0 (by definition) for the 180 degree position for the 129 keV gamma ray from Pu-239.

For full assays, drums are rotated to reduce the biases induced by activity concentrations within the drum. The results show that there is an additional induced bias from the rotational position of the source due to the additional attenuation, but the rotational average of these biases produces deviations that are typically less than 5%. In the context of the total measurement uncertainty of most NDA systems, an error of this magnitude is typically negligible. This is in sharp contrast to the effect of source height or the self-attenuation of the WRM source matrix and canister itself. These two effects are presented and discussed in the following sub-sections.

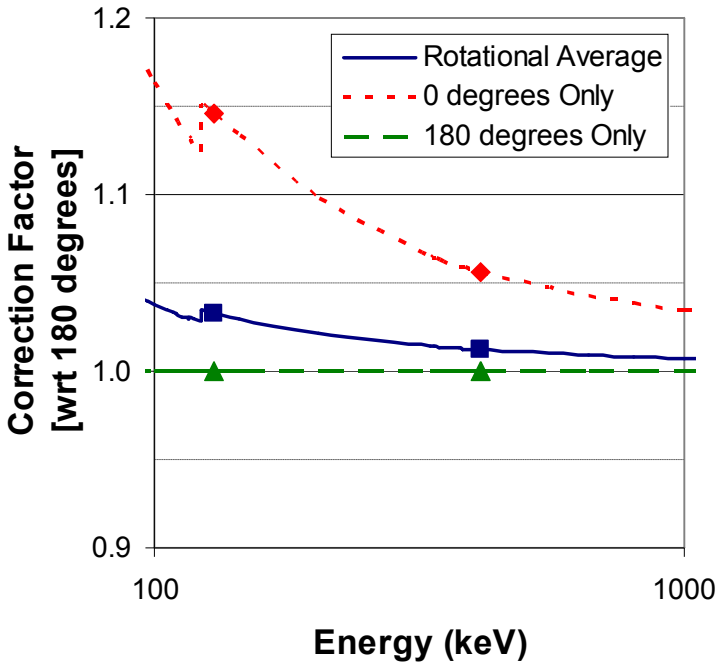

Figure 2. Relative (PuDE/AIR)$^{-1}$ correction factors with respect to the 180 degree position for efficiencies in the Top detector for the WRM located at the top level at 80% radial position at 0 (closest) and 180 (farthest) degree drum rotation angles. Also included is the average normalized correction factor over all ration angles. The points on each line represent the gamma-ray energies 129 and 414 keV respectively. These are two key gamma-ray emission lines from Pu-239 decay.

Effect of Source Height

To investigate the effect of source height on the source attenuation, we consider the WRM source positioned at the 80% radial position at each of the three vertical source positions and averaged over all drum rotations and viewed from the top detector position. The condition of attenuation for a WRM that is loaded only with DE (DEonly), and presumably a very small amount of PuO_2 is presented in Figure 3. The computed correction factors for the PuDE WRM are presented in Figure 4.

In the case of the DEonly matrix, one can observe that the source has significant correction factors even for the case where the source is at the same height as the detector (the top position). When the source is in the bottom position the correction factor can be as great as about 4.5 at 129 keV. At the bottom positions, the gamma-rays from the source must pass through more substantial components of the canisters, as well as, having longer path lengths through the sides of the canister due to the shallower angles and lower efficiency due to more gammas entering through the sides of the detector.

In the case of a large mass loading of PuO_2 in the WRM source, the correction factors become even more substantial. For example, in the case of the PuDE matrix for the source in the bottom position, a correction of nearly 20 is required for the top detector at 129 keV. This is almost a factor of 5 greater than the DEonly matrix. Thus the

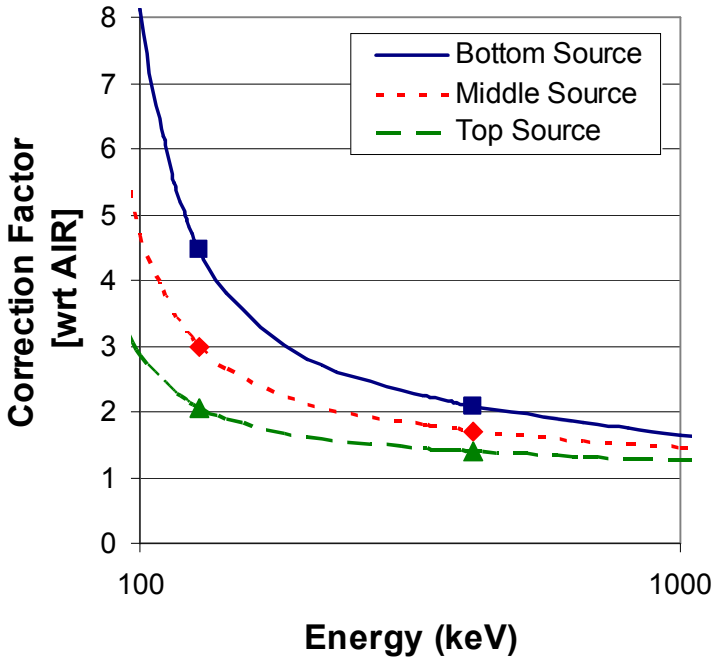

Figure 3. (DEonly/AIR)$^{-1}$ correction factors with respect to the vertical source position for efficiencies in the Top detector for the three WRM sources located at the 80% radial position on each level and averaged over all drum rotation angles. The points on each line represent the gamma-ray energies 129 and 414 keV respectively. These are two key gamma-ray emission lines from Pu-239 decay.

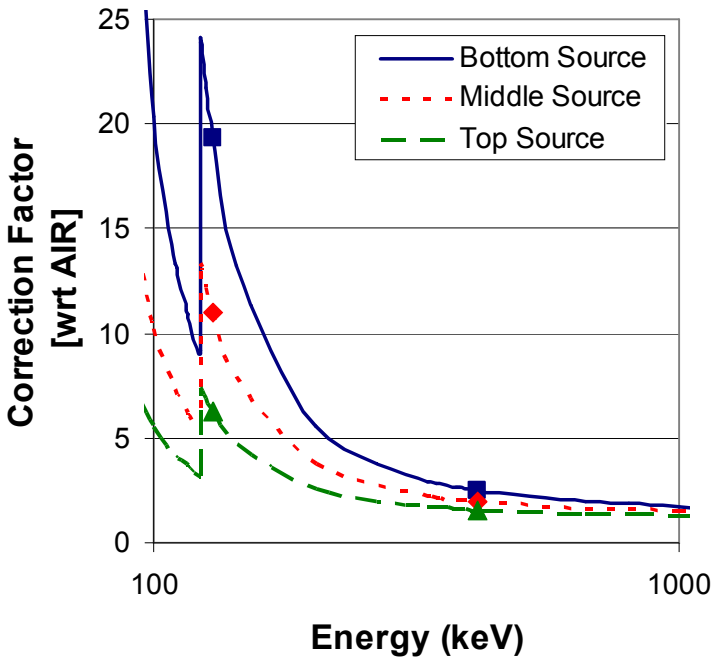

Figure 4. (PuDE/AIR)$^{-1}$ correction factors with respect to the vertical source position for efficiencies in the Top detector for the three WRM source located at the 80% radial position on each level and averaged over all drum rotation angles. The points on each line represent the gamma-ray energies 129 and 414 keV respectively. These are two key gamma-ray emission lines from Pu-239 decay.

correction factor is variable and significant depending on the PuO_2 load of the WRM. For reference, for the Weapons Grade Pu (WGPu) PDP WRMs the Pu content varies between approximately 0.02 g and 75 g.

Average Effect

The Q^2 system is designed in such a way that errors due to source non-uniformities are reduced by having three detectors distributed over the height of the drum, and the assays are performed while the drum is rotated. Consequently, comparing the correction factors required for only a few selected geometries may not provide of proper overall impression of system response. In this section, we consider the full assay condition in which the WRM is at each of the nine positions within the PDP drum (as indicated in Figure 1). The drum is rotated so all radial positions are averaged and the efficiency of all three detectors are combined.

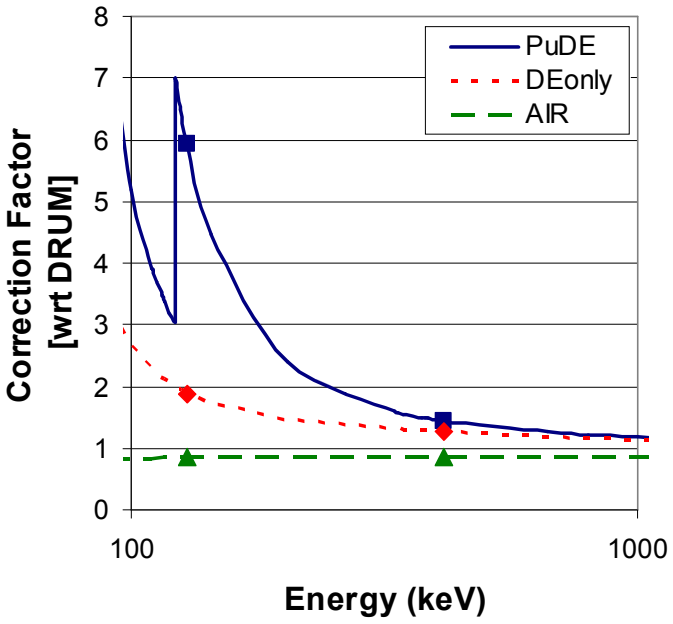

Figure 5. $(PuDE/DRUM)^{-1}$, $(DEonly/DRUM)^{-1}$, and $(AIR/DRUM)^{-1}$ correction factors averaged over all detectors, source positions, and drum rotation angles. The points on each line represent the gamma-ray energies 129 and 414 keV respectively. These are two key gamma-ray emission lines from Pu-239 decay.

It should also be noted that Q^2 systems are not calibrated to WRM sources, but rather under the assumption that the source activity is distributed uniformly through the volume of the 55 gallon drum. In addition, it is assumed that there is negligible Pu self-attenuation effect; i.e. it is assumed that the calibration source does not contain "lumps" of Pu. In this regard we consider the correction factors for PuDE, DEonly, and AIR all with respect to the uniformly-distributed DRUM efficiency with a non-interfering matrix of air with density 0.0013 g/cc (see Table 1). These correction factors are presented in Figure 5.

In the case of the $(AIR/DRUM)^{-1}$, this shows primarily the effect of the source geometry compared to the typical calibration geometry. The $(AIR/DRUM)^{-1}$ correction factor is less than one with a value of approximately 0.85 independent of energy. This discrepancy is due only to the geometric difference of activity located at the nine WRM source positions (no attenuation from the WRM canisters or drum matrix), compared to activity distributed throughout the entire drum.

If one considers the situation where the PuO_2 source is located within the WRM canister, one observes the strongly energy dependent correction factors represented by the DEonly and PuDE trends in Figure 5. The DEonly trend represents the "minimally" attenuating condition in which there is little plutonium self-absorption (i.e. close to the lowest mass loading of about 0.02g WGPu). The PuDE represents the situation in which there is significant plutonium self-absorption due to the high mass loading of the WRM (i.e. 65g

WGPu close to the upper mass limit of 75g). At 414 keV the correction factors for the PuDE and DEonly are 1.44 and 1.27 respectively. At 129 keV the corrections factors cover a very significant spread of 5.9 to 1.9 from the high Pu load to low load respectively. Thus not only do all WRM sources have significant self-attenuation due to the structure of the canisters, the amount of self-attenuation can vary strongly between sources dependent on the amount of plutonium within the source.

DISCUSSION

Such deviations illustrate the potential challenges related to the assay of waste containers in general. While typical calibrations for waste assay systems assume the radio-isotopic contents are uniformly distributed throughout a container of uniformly distributed inert matrix material, it is also known that this is only an approximation to the actual radio-isotopic distribution which, in general, is unknown and potentially highly non-uniform. The matrices themselves can be highly variable and it is not unlikely that the radio-isotopic contents could be readily shielded. This shielding could come in the form of self-attenuation, in the form for example of plutonium lumping, or by shielding within the matrix contents itself.

The conditions presented by PDP style assay with WRM's certainly does not represent the ideal calibration configuration, but these PDP containers indeed do not misrepresent the potentially difficult conditions of a "true" waste drum. The successful measurement of a PDP style drum often requires the implementation of advanced analysis techniques to account for conditions beyond the standard calibration.

The PDP program has had the beneficial effect of further defining and probing the total measurement uncertainties (TMU) of systems, while encouraging developments in measurement systems that ultimately reduce the TMU in waste applications.

By way of example, most plutonium waste NDA systems include a mechanism to perform a "self-consistency" check on the analysis results by comparing the computed Pu-239 mass from the measured rate of the 129 keV to that from 414 keV. It is well known that the analysis of these two lines should produce statistically the same result following appropriate corrections for decay yield and other physical conditions. A difference in these yields indicates a condition in which the calibration is not consistent with assumed source distribution.

One common technique to correct for this inconsistency is to apply an energy-dependent correction based on the function, $e^{B/E}$, in which "*B*" is a fitting parameter and "*E*" is the decay energy. An elementary approach is to fit the parameter "*B*", such that the results computed Pu-239 mass for 129 keV and 414 keV transitions produce the same result.

The application of this approach is illustrated in Figure 6. In this figure is represented the fitting of the above exponential function to the correction factor data presented in Figure

5. While the application of this exponential correction does not completely eliminate the biases in each of the test conditions, they biaes are significantly reduced. For example,

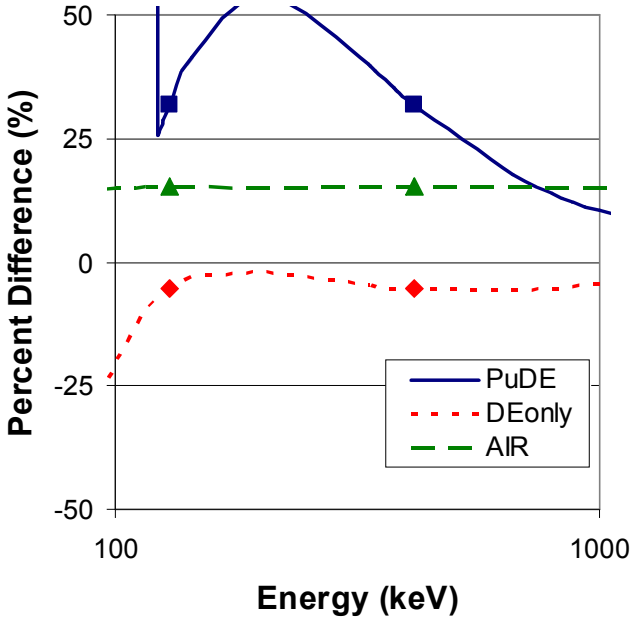

Figure 6. The percent difference for (PuDE/DRUM)$^{-1}$, (DEonly/DRUM)$^{-1}$, and (AIR/DRUM)$^{-1}$ correction factors averaged over all detectors, source positions, and drum rotation angles compared to an approximate $e^{B/E}$ correction in which the value of "*B*" is the result of a fit that produces the same net correction factor for the gamma-ray energies 129 and 414 keV.

the computed uncorrected bias of the 129 keV in the PuDE WRM source was a factor of 5.9 above the "uniform" calibration. Following this simple correction, the bias is only 30%. The 414 keV bias in this example is reduced from 44% to 30%.

The simple correction presented here is not intended to imply that this is the extent of corrections applied in the fielded systems, but rather provided as an illustration to highlight that such additional self-consistency and other more advanced methods are routinely applied in advanced NDA systems.

CONCLUSIONS

The practice of using Pu working reference standards is intended to exercise the capabilities of the NDA system with respect to known limitations of the NDA measurement. In the interpretation of the data from the assay of the WRM, it is important to remember that these materials are generally not intended to be ideal calibration standards. In order to handle the Pu safely, these WRM sources need to be encapsulated, but as we have shown the encapsulation can exert a significant influence on the emergent gamma-ray intensity. Additionally the substrate used to distribute the PuO_2 within the capsule and the mass of the PuO_2 itself gives rise to self-attenuation which is a function of that mass loading. While an ideal calibration standard might avoid these challenges, our results illustrate the need to properly account for these effects in both the analysis technique and in the total measurement uncertainty budget.

We have shown that mathematical modeling is a feasible method for quantifying the effects provided a description of the sources is available. Numerical results were provided for a particular instrument type (Q^2) to illustrate the magnitude of the effects. In this example, we demonstrated that typical methods to "average out" inhomogeneities by distributing the WRM sources at nine different radial and vertical positions and rotating the drum do not eliminate the interfering effects. These results show that there is a potential for a 15% bias at all energies due to the use of 9 discrete rotating sources to simulate a uniform matrix. Further we show that the use of the WRM sources to represent a uniform "non-lump" container can lead to errors of 30-45% at 414 keV to factors of 2-6 at 129 keV. The degree of error is related to the amount of plutonium within the WRM source. It is also emphasized that these biases must be taken in account for the successful assay of a PDP drum, and in fact general correction algorithms exist in most NDA systems which can also be applied to the results of PDP drum assays.

While this paper concentrated on the Q^2 geometry, the interfering effects of WRM sources in other gamma-ray spectroscopy NDA systems can have similar biases since the primary contributions are the interfering effect of the WRM canister with large variability due to the Pu mass load in the source. The combined influence of variability of WRM sources with different Pu masses in addition to the standard waste drum matrix corrections must be understood and accounted in any gamma-ray spectroscopy-based NDA system in which WRM sources are used to evaluate the system's proper operation. The PDP drums represent conditions that extend beyond routine uniform source and matrix distributions, and consequently beneficially stress NDA systems beyond standard calibrations.

We note that not all waste forms will be dilute and non-attenuating and nor will they be like the manufactured working reference materials available. Interpretation of measured data by subject matter experts in the context of what is known about the waste stream assisted by embedded algorithms to flag for self-attenuation and so forth remains a vital component for assuring quality results. The detailed understanding of the calibration and test conditions further contribute to the development of advanced NDA systems with increased accuracy for the waste streams they are designed to assay while avoiding the pitfalls that can arise from making corrections without a full understanding of the system performance.

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