Segmented Gamma Scanner for Small Containers of Uranium Processing Waste- 12295

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ABSTRACT

The Segmented Gamma Scanner (SGS) is commonly utilized in the assay of 55-gallon drums containing radioactive waste. Successfully deployed calibration methods include measurement of vertical line source standards in representative matrices and mathematical efficiency calibrations.

The SGS technique can also be utilized to assay smaller containers, such as those used for criticality safety in uranium processing facilities. For such an application, a Can SGS System is aptly suited for the identification and quantification of radionuclides present in fuel processing wastes. Additionally, since the significant presence of uranium lumping can confound even a simple 'pass/fail' measurement regimen, the high-resolution gamma spectroscopy allows for the use of lump-detection techniques. In this application a lump correction is not required, but the application of a differential peak approach is used to simply identify the presence of U-235 lumps.

The Can SGS is similar to current drum SGSs, but differs in the methodology for vertical segmentation. In the current drum SGS, the drum is placed on a rotator at a fixed vertical position while the detector, collimator, and transmission source are moved vertically to effect vertical segmentation. For the Can SGS, segmentation is more efficiently done by raising and lowering the rotator platform upon which the small container is positioned. This also reduces the complexity of the system mechanism.

The application of the Can SGS introduces new challenges to traditional calibration and verification approaches. In this paper, we revisit SGS calibration methodology in the context of smaller waste containers, and as applied to fuel processing wastes. Specifically, we discuss solutions to the challenges introduced by requiring source standards to fit within the confines of the small containers and the unavailability of high-enriched uranium source standards. We also discuss the implementation of a previously used technique for identifying the presence of uranium lumping.

INTRODUCTION

The computer-controlled Segmented Gamma Scanner (SGS) has been in use for the non-destructive assay (NDA) of radionuclides in drummed waste for almost 40 years. The technique uses high resolution gamma spectroscopy to identify radionuclides based on their gamma-ray signature. Gamma spectroscopy acquisition is performed during rotation of the drum, which provides for the uniform sampling of the material present in the drum and smoothes the effects of radionuclide and attenuation inhomogeneity. Strict vertical collimation allows the drum to be divided into several segments thereby easing the requirement of matrix homogeneity to the confines of an individual layer [1] .

The SGS is most frequently employed for the characterization of large drummed waste typically stored in 55 gallon (208 L) drums. Although the principle of the technique is applicable to any size container, the mechanism and collimation must be altered in order to provide the correct

detector field of view and measurement efficiency appropriate to the container size. Additionally, the system calibration must take these variations into account.

In uranium processing facilities, characterization of wastes is often required for material control and accountancy (MC&A), disposal, and occasionally criticality safety purposes. Criticality safety controls are often implemented on the mass of the material contained within a given area or container [2, 3, 4], as mass is a direct factor in producing a critical or supercritical reaction. A natural extension of controls of mass is the control of volume, which can be applied to any enrichment or density of material. In the case of uranium-processing waste, volume is limited by restricting the waste to containers of only a few liters, essentially eliminating the risk of a criticality accident. These containers must still be characterized for MC&A and disposal, hence the need for a small-scale SGS such as the Can SGS described here.

The Can SGS system comprises a detector and collimator assembly, a transmission source assembly, and a small container rotator with vertical drive mechanism for scanning. The mechanical movement of the SGS system is controlled by a Programmable Logic Controller (PLC) in conjunction with Canberra's NDA 2000 software. The NDA 2000 software also performs data acquisition and analysis.

The detector and collimator assembly consists of a detector, detector shield, and collimator. The detector is a broad-energy High Purity Germanium (HPGe) detector with a 28 cc surface area crystal that is 2.5 cm thick, on a Cryo-Cycle hybrid liquid nitrogen and electric cryogenic system. The detector shield consists of a cylindrical steel weldment of poured lead, 53.2 mm (2.1 inch) thick. The inner diameter of the detector shield is lined with 2.3 mm (0.09 inch) of cadmium. The collimator portion of the assembly consists of a rectangular steel weldment of poured lead. The collimator has a fixed opening of 12.7 mm (0.5 inch) high by 177.8 mm (7.0 inch) wide by 152.4 mm (6.0 inch) deep. The opening of the collimator is lined with 3.3 mm (0.13 inch) thick tungsten.

Figure 1. Photograph of the Can SGS as built in 2011 at Canberra's Meriden Facility.

The transmission source assembly is located on the opposite side of the container from the detector and consists of a highly collimated 5 mCi Eu-152 transmission source. The purpose of the transmission measurement is to provide an attenuation correction due to the waste matrix inside the container [5]. The transmission source shield is a 203 mm (8.0 inch) diameter lead shield used to hold and position the transmission source, as well as provide approximately 100 mm (4 inches) of lead shielding for personnel protection. A 4.8 mm (0.19 inch) hole through the center of the shield provides the beam path for the transmission source when the shutter is open. The shutter assembly contains a 76.2 mm (3 inch) long by 12.5 mm (0.498 inch) diameter tungsten shutter plug mounted to a slider assembly. For fail-safe operation upon loss of power, the tungsten shutter plug drops to block the transmission source beam.

Figure 2 shows the top view of the system geometry. The height of the rotator is 279.4 mm and the distance from the detector endcap to the center of the container is 308 mm.
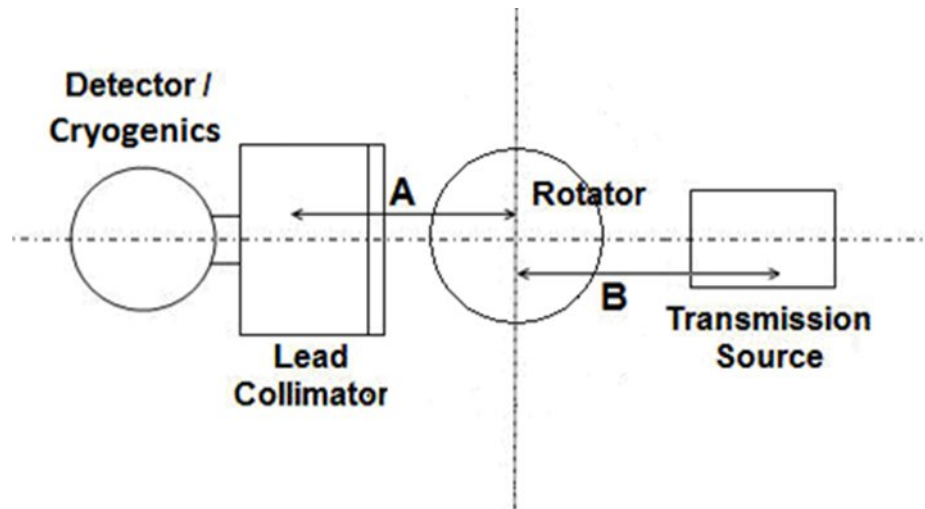

Figure 2. The Top View of the Can SGS System Geometry (drawing not to scale). The distance from the center of the rotator to the detector face (labeled A) is 308 mm and to the location of the transmission source within the transmission source assembly is 407 mm (labeled B). The width of the rotator is 178 mm.

The Can SGS differs from the current drum SGS designed for measurement of 55 gallon drums in the mechanism, collimator opening, and size of the transmission source. In terms of differences in the mechanism, for the Can SGS the rotator/container platform is moved vertically to achieve segmentation rather than the detector and transmission source assemblies in the traditional SGS. The detector and transmission source assemblies are positioned on fixed tables. The elimination of the vertical drive mechanisms on the detector and transmission assemblies simplifies the mechanism design and control.

The collimator size and system configuration were designed for high throughput of 2 liter bottles, while still maintaining adequate segmentation. For this reason, the collimator opening was reduced from 101.6 mm to 12.7mm. The assay item was divided into eight segments, each approximately 1 inch high.

For smaller containers, a smaller transmission source can be used and still yield sufficient statistical count rates through dense matrices. The transmission source used with the Can SGS is a 5 mCi Eu-152 source, reduced from the 10 mCi Eu-152 source used in a current drum SGS. The Eu-152 source provides attenuation correction values across a broad range of energies (121 keV – 1408 keV), encompassing gamma-ray signatures from a broad range of nuclides. The broad energy range also ensures attenuation correction is still performed even when highly attenuating materials reduce low energy peaks beyond reasonable statistics. Barium-133 is a commonly used transmission source when measuring special nuclear material as its key energy lines range from 80 to 356 keV. However, Eu-152 was chosen due to the prevalence of high density matrix materials present in uranium processing wastes. The 5 mCi Eu-152 source produced unattenuated real-time transmission count rates of approximately 1750 cps and 170 cps for the 121 keV and 1408 keV lines, respectfully.

The Can SGS is configured for two types of SGS measurements, SGS with and SGS without transmission. For assays without transmission correction, the NDA 2000 software uses a multi-curve efficiency calibration for nuclide quantification. A multi-curve efficiency is a group of density and energy dependent efficiency curves spanning the expected density and radionuclide gamma energies expected in the waste stream. It allows for interpolation between known efficiencies allowing measurement of any item within the energy and density bounds. For assays using transmission correction, the NDA 2000 software uses the transmission calibration to calculate the item transmission ratio and then applies a correction for matrix attenuation to the zero-density efficiency calibration obtained from the multi-curve efficiency calibration. A typical NDA 2000 report gives the radionuclide activities for each segment and for the entire container called here the Combined Segment NID. This reporting stage is configured to report either the average of the segment results or the sum of segment results.

As high throughput is often a requirement, the SGS assay was limited to a total assay time of 20 minutes. Therefore, an SGS with transmission count measured each segment for 110 seconds emission only and 36 seconds for transmission segment. To reduce biases for the very short count times, relative to the current drum SGS, each acquisition is constrained to be comprised of whole segment rotations, i.e., the container must complete an integer number of rotations. Biases due to partial container rotations become less significant as the total number of container rotations per count increases.

LUMP DETECTION

The correction of self-absorption in special nuclear material is a common problem in NDA. One method used for self-absorption correction is called differential attenuation analysis. It takes advantage of the difference in attenuation between the varied Pu gamma-ray peaks to determine the total mass. The technique works well for lumps of Pu because the gamma-ray peaks are broadly spaced in energy (129-414 keV). [6]

Although the principles of the technique are applicable to any nuclide emitting several gamma rays, in practice a self-absorption correction based on differential attenuation is not reliable for U. This is because of the low energies and close proximity of the U-235 gamma ray peaks, which reduces its sensitivity to self-absorption [7]. However, this does not mean that the analysis cannot be adapted to the detection, or qualification, of large amounts of self-shielding resulting from lumps of material.

This is not a novel concept; the solution employed on the Can SGS is based on previous predictions by Gillespie [8] and implementation of lump detection by Rackham [9]. Rackham

employed the differential attenuation, alternatively called Differential Peak Absorption technique, to the assay of Uranium in waste drums. Based on the ratios of the transmission-corrected net peak areas of the 143 keV, 163 keV, and 205 keV peaks to the transmission-corrected net peak area at 186 keV, the measured uranium could be classified within two categories: infinitely dilute or significant self-shielding.

Canberra's implementation of lump detection compares the line activities of U-235 at 143 keV, 186 keV, and 205 keV. The line activity provides an advantage over peak area as line activity already accounts for attenuation correction and peak intensity. If the ratio of activity for the 143 keV peak compared to the 186 keV peak is less than a user-defined limit, then a lump is identified. Similarly, if the ratio of activity for the 205 keV peak compared to the 186 keV peak is greater than a user-defined limit, then a lump is identified. The line activity ratio limits are editable by the customer and will need to be optimized for the material being assayed.

CALIBRATION METHODS

For a standard segmented gamma scanner system, the total system calibration is comprised of many separate, although often interrelated, calibrations. In this discussion, we limit our scope to the transmission and efficiency calibration approaches. Specifically, we focus on variations from standard SGS calibration approaches for small containers and for expected activity lumping in the waste stream.

The transmission calibration provides a reference count rate for transmission assays to determine the appropriate self-attenuation correction for a given sample matrix. To generate the calibration, a measurement is collected with the empty sample container positioned on the rotator and with the transmission source exposed. Then, during an item assay with transmission, the transmission source is again exposed and the count rate from the transmission source beam attenuated by the sample matrix is detected and recorded. The transmission fraction relative to the empty container is calculated and used to determine the matrix-specific attenuation correction factors based on the approach developed by J. L. Parker [10, 11]. In that approach, the attenuation correction factor (CF) is determined empirically based on the following equation for a cylinder in far-field geometry:

$$CF = \frac{-\ln(T^{\kappa})}{1 - T^{\kappa}}$$

where T is the transmission fraction relative to the empty container and the factor κ (kappa) is a geometry dependent parameter defined during calibration.

For standard SGS calibrations performed of 55-gallon drums, the most common container type used with the SGS approach, κ has been established to be 0.823. For small containers, which have a larger ratio of detector distance to container radius, this correction factor must be revisited. In Parker's derivation of self-attenuation correction, the most significant parameters are listed as (1) the linear attenuation coefficient of the material, (2) the volume and shape of the sample material, (3) the linear attenuation coefficient of the sample container, (4) the size and shape of the sample container, (5) the position and orientation of the sample relative to the detector, and (6) the size, shape, and efficiency of the detector. While parameters 1 and 3 are consistent, the sample volume is reduced, container size and thickness are less, the detector is positioned closer to the sample, and the reduced detector collimator opening effects a different detector shape and efficiency. Using an empirical optimization approach, it was determined that for the specific geometry described for this system, κ is 0.700.

A source-based efficiency calibration using rod sources has been the historically proven method of calibration of drums, and is accepted as the industry standard for Efficiency Curve Calibrations [12]. A set of 6 - 7 vertical rod sources are placed at specified radii within the drum, that, when rotated, approximate a uniform distribution of activity. The radial placement *s* of *n* calibrated rod sources of equal strength to create a uniform distribution of activity follows

$$s_i = \left(i - \tfrac{1}{2}\right) \cdot \frac{a^2}{n} \;, \quad i = 1 \text{ to } n$$

where *a* is the internal radius of the drum and *i* indicates an individual rod source. The result is that each rod rotates through an equal volume distribution [13].   However, with small containers, it is not possible to fit large numbers of rod sources, and in fact one or perhaps two sources are the maximum due to space constraints. Therefore, for a small container, to minimize the amount of displaced matrix, a single rod would be placed at a radius of 0.71 times the radius of the matrix. Further considerations place the location of a single source at 80 – 85% of the container radius [13]. The methodology of having a retaining cylinder for placement of the calibration source also has consequences, as the ratio of displaced matrix to sample matrix becomes more significant, recalling that the diameter of the rod source holders are constant and cannot be proportionally reduced in size to the small container. Alternatives to using rod sources for a source-based calibration include point sources or custom-made uniform activity distribution source standards. However, point sources will introduce other challenges to approximating a uniform activity distribution due to vertical segregation, and a volumetric source standard approach has a greater challenge in spanning a range of densities and matrix materials for a system calibration.

As a solution to these challenges, a mathematical-based efficiency calibration approach is another option and is used in place of a source-based approach for the Can SGS. Canberra's In Situ Object Counting Software (ISOCS) is used to generate uniform activity efficiency responses for a range of densities and energies for the small container of interest.   The efficiency response is calculated both for each segment, and for the summed spectra of all segments. This is then loaded into the software to create a multi-curve efficiency calibration.

Good agreement has been shown using this calibration approach for uniform activity distributions in homogeneous drums as well as for other large containers [14, 15, 16]. For non-homogeneous activity distributions, a Total Measurement Uncertainty (TMU) analysis is performed and captures maximum expected variations, and can be used to develop appropriate process decision limits.

The ISOCS mathematical efficiency approach can be and is directly applied to smaller containers with no loss in fidelity. The simple cylinder and complex cylinder templates, which are generally used for a 55 gallon drum, are also suitable templates for a small container measured by an SGS. Figure 3 shows a graphical rendering of an ISOCS model using the simple cylinder to calculate the efficiency response of a 2 liter container measured by the Can SGS.
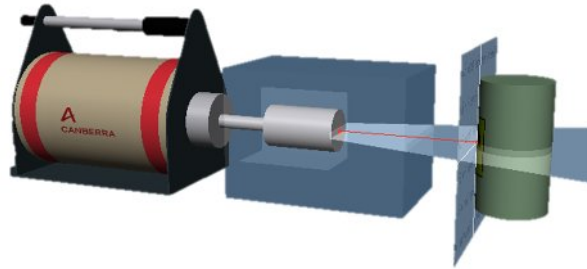
Figure 3 ISOCS model of a 2 liter container, Can SGS collimator, and BE2825 Ge detector. The template used is a simple cylinder, and in this model the full container is defined for a specific segment. The collimator is rendered in blue and made transparent to view the detector. Also rendered is the field of view of the collimator opening and the reference plane used to define the geometry parameters.

For a uniform activity distribution throughout the container, the most accurate and preferred approach to mathematical efficiency calibration is to define the full container for each segment measurement. One can then hypothetically determine the total container activity from any given segment measurement, and it follows that the Combined Spectra NID reports on the average of the activities determined by the individual segment measurements. This approach takes into account count rate contributions from other segments, and so the error introduced by segment crosstalk is minimized. The error associated with a non-homogeneous distribution is accounted for in a TMU.

However, the challenge introduced by lump detection via a segment activity decision level is that (i) the segment activity must correspond to the observed activity in the given segment only and (ii) the assumption of a homogeneous activity distribution is in direct contrast to the condition being evaluated. Furthermore, the presence of lumps is itself a significant contributor to assay error using the SGS technique [12]. Considering the application of lump detection, we propose a different mathematical calibration approach for the efficiency calibration using ISOCS, which we will refer to as the "heterogeneous calibration" to distinguish it from the "homogeneous calibration" discussed above.

The key difference with the heterogeneous efficiency calibration is that the geometry associated with the segment efficiency calibrations is defined to be only the specified segment, rather than the full container. This is illustrated in Figure 4. As a result, the activity reported by any given segment measurement will be limited to only the activity in the given segment itself, and the Combined Spectra NID results will be the sum, rather than the average, of the activities observed in each segment. A uniform activity distribution is still assumed in a given segment, but this approach will greatly reduce the geometric error, typically captured in a TMU, of the worst case scenario in which a small lump is the single source of activity in the container. It also allows for a reasonable activity quantification of lumps in a given segment, which may be used for screening and determining decision limit in the waste stream. The drawback of this approach is that if activity exists in adjacent segments, the count rate contributions from other segments are not considered in the efficiency calibration and will result in an overestimation of the activity.
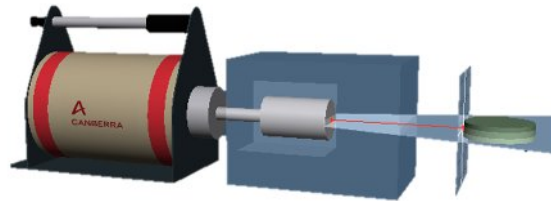
Figure 4 ISOCS rendering of the geometry defined for the efficiency calibration of a segment using the heterogeneous calibration.   As in Figure 3, the collimator is rendered transparent, and the detector viewing angle and geometry reference plane are illustrated.

RESULTS

Verification of the system calibration was performed with a 2 liter bottle and with a set of point sources bundled together. At the time of measurement these sources comprised of a single source of Co-57 with an activity of 26.88 µCi and four sources of Ba-133 with a total activity of 20.88 µCi. Each source is encapsulated in disc of 1 inch plastic, and the five sources are stacked together to create a vertical activity distribution of approximately 1 inch. The Co-57 is positioned in the middle of the stack, with two Ba-133 sources below and two Ba-133 sources above. Each of the Ba-133 sources have comparable activities.

Four different matrix materials were used for verification: air, scrap papers and vacuum cleaner bags, plastic gloves and plastic bags, and sand. For all but the air matrix, a plastic tube was placed vertically in the matrix at a radius of approximately 85% the container radius in order to insert the sources into the container matrix. The sources were typically placed at approximately half the height of the container.

Table 1 shows the activity results determined during a standard item count without transmission. The counts were analyzed with both approaches to the system efficiency calibration – the more common "Homogeneous Calibration", and our proposed "Heterogeneous Calibration". The activities were determined by taking the weighted average of the line activities, and for each count, only the activities reported by the two segments closest to the source are listed, termed "upper" and "lower" segments Activity reported for all other segments are close to the minimum detectable activity. The Combined Segment NID results, indicating the total measured activity in the container, are listed as well as how this value compares to the expected activity. The Combined Segment NID results for the Heterogeneous Calibration were determined by taking the sum of the individual segment activities, while for the Homogeneous Calibration they were determined by taking the average of the individual segment activities.

	Heterogeneous Calibration		Homogeneous Calibration	
	Co-57	Ba-133	Co-57	Ba-133

True Total Activity (µCi)	26.9 ± 0.99	20.9 ± 0.94	26.9 ± 0.99	20.9 ± 0.94
DENSITY 0.001 g/cc - Empty				
Lower Segment	0.40 ± 0.017	4.6 ± 0.15	2.7 ± 0.11	31 ± 1.0
Upper Segment	27.5 ± 0.33	17.0 ± 0.49	182 ± 2.2	113 ± 3.3
Combined Segment NID	29.0 ± 0.67	21.6 ± 0.52	23.2 ± 0.28	18.04 ± 0.433
Measured / True	1.04 ± 0.048	1.04 ± 0.055	0.86 ± 0.029	0.86 ± 0.038
DENSITY 0.189 g/cc – Cellulose				
Lower Segment	23.3 ± 0.28	14.9 ± 0.43	0.31 ± 0.049	6.2 ± 0.33
Upper Segment	1.7 ± 0.038	6.7 ± 0.21	176 ± 2.1	134 ± 3.7
Combined Segment NID	25.0 ± 0.30	21.6 ± 0.50	22.1 ± 0.27	18.1 ± 0.47
Measured / True	0.93 ± 0.034	1.03 ± 0.054	0.82 ± 0.026	0.87 ± 0.039
DENSITY 0.530 g/cc – Polyethylene				
Lower Segment	30.2 ± 0.39	20.0 ± 0.52	205 ± 2.67	135 ± 3.9
Upper Segment	0.08 ± 0.002	2.93 ± 0.097	0.52 ± 0.079	19.8 ± 0.79
Combined Segment NID	30.3 ± 0.39	23.1 ± 0.56	25.8 ± 0.34	19.5 ± 0.51
Measured / True	1.13 ± 0.050	1.10 ± 0.063	0.96 ± 0.036	0.93 ± 0.045
DENSITY 1.26 g/cc – Sand				
Lower Segment	32.6 ± 0.55	19.8 ± 0.59	227 ± 3.9	136 ± 4.6
Upper Segment	19.8 ± 0.59	0.20 ± 0.021	1.4 ± 0.17	23 ± 1.1
Combined Segment NID	32.9 ± 0.53	23.22 ± 0.60	28.6 ± 0.46	20.1 ± 0.54
Measured / True	1.22 ± 0.060	1.11 ± 0.064	1.06 ± 0.046	0.96 ± 0.048

Table 1. Can SGS emission verification results for a 2 liter bottle. Two multi-curve calibrations are compared – the "homogeneous calibration" and the "heterogeneous calibration".

The results show that a reasonable value of the segment activity is determined in the heterogeneous calibration, which is the primary motivation for employing the heterogeneous calibration approach.   It is seen that the segment activity is grossly overestimated in the homogeneous calibration, but that is not un-expected as in standard practice the segment activities are not used directly. When the overestimated segment results are averaged with the segments reporting no activity during the Combined Segment NID processing, the result is a total measured container activity that is comparable to the expected activity. Likewise, for the heterogeneous calibration, the Combined Segment NID also reports results that are comparable to the expected activity.

To demonstrate the accuracy of the transmission calibration for small containers, the results of a set of item counts with transmission are presented in Table 2. The Combined Segment NID results are reported, as is the comparison to the expected activities. These results are generated with the heterogeneous efficiency calibration and with κ = 0.7. For all matrices, the transmission Combined Segment NID results are very good and well within 2 standard deviations of the expected values.

	Co-57	Ba-133
True Total Activity (µCi)	26.9 ± 0.99	20.9 ± 0.94
DENSITY 0.001 g/cc		
Combined Segment NID	27.6 ± 0.69	21.5 ± 0.54
Measured / True	1.03 ± 0.047	1.03 ± 0.05
DENSITY 0.189 g/cc		
Combined Segment NID	26.7 ± 0.32	21.5 ± 0.56
Measured / True	0.99 ± 0.038	1.03 ± 0.05
DENSITY 0.530 g/cc		
Combined Segment NID	28.6 ± 0.37	22.6 ± 0.59
Measured / True	1.06 ± 0.044	1.08 ± 0.061
DENSITY 1.26 g/cc		
Combined Segment NID	25.5 ± 0.36	22.4 ± 0.67
Measured / True	0.95 ± 0.036	1.08 ± 0.063

Table 2: Activity results for the an Item Count with Transmission for a range of matrix materials in a 2 liter bottle

CONCLUSION

The SGS technique is a well-accepted NDA technique applicable to containers of almost any size. It assumes a homogenous matrix and activity distribution throughout the entire container; an assumption that is at odds with the detection of lumps within the assay item typical of uranium-processing waste. This fact, in addition to the difficultly in constructing small reference standards of uranium-bearing materials, required the methodology used for performing an efficiency curve calibration to be altered. The solution discussed in this paper is demonstrated to provide good results for both the segment activity and full container activity when measuring heterogeneous source distributions.

The application of this approach will need to be based on process knowledge of the assay items, as biases can be introduced if used with homogenous, or nearly homogenous, activity distributions. The bias will need to be quantified for each combination of container geometry and SGS scanning settings.

One recommended approach for using the heterogeneous calibration discussed here is to assay each item using a homogenous calibration initially. Review of the segment activities compared to the full container activity will signal the presence of a non-uniform activity distribution as the segment activity will be grossly disproportionate to the full container activity as seen in Table 1. Upon seeing this result, the assay should either be reanalyzed or repeated using the heterogeneous calibration.

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