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Nuclear Material Matrices from Future Fuel Cycles

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TOMOGRAPHIC GAMMA SCANNING (TGS) TO MEASURE INHOMOGENEOUS NUCLEAR MATERIAL MATRICES FROM FUTURE FUEL CYCLES

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ABSTRACT

Current methods for the non-destructive assay (NDA) of special nuclear materials (SNM) in 208-L drums can give assay errors of 100% or more when the drum matrix and/or radionuclide distribution is nonuniform. To address this problem, we have developed the tomographic-gamma-scanner (TGS) method for assaying heterogeneous drummed SNM. TGS improves on the well-established segmented-gamma-scanner (SGS) method by performing low-resolution tomographic emission and transmission scans on the drum, yielding coarse three-dimensional images of the matrix density and radionuclide distributions. The images are used to make accurate, point-to-point attenuation corrections. The TGS geometric counting efficiency is 60% that of a typical SGS device, allowing a TGS assay time of only 28 min per drum with a one-detector system. TGS may also be useful for non-destructive examination (NDE). Currently, TGS is the only practical method of imaging SNM in drums.

I. INTRODUCTION

Future nuclear fuel cycles are expected to generate 208-L waste drums that have no high-level contamination and are classified as "contact-handled," but that contain unknown amounts of ^{235}U , ^{239}Pu , and other special nuclear material (SNM) and transuranic (TRU) isotopes. The safe and economic disposition of these drums will require non-destructive assay (NDA) to measure the amount of SNM/TRU waste in each drum. Moreover, in the light of current regulatory trends, it seems likely that future regulations will dictate that all such drums be well characterized. This is already the case with TRU waste, which is subject to a growing assortment of rules that either presume a knowledge of the TRU loading or explicitly require some form of NDA.

Current methods for the NDA of SNM/TRU waste in 208-L drums rely on the assumption that both the drum matrix and the SNM/TRU radionuclides are homogeneously distributed within the drum. When this condition is not met and the matrix is non-benign, large assay errors can result. This is true for neutron-based NDA methods as well as for gamma-ray spectroscopic methods. The problem in either case is that the matrix effects are substantial and depend sensitively on the actual distribution of radionuclides and matrix materials, so a homogeneous drum assumption is not justified in general. To address this problem, we have developed the tomographic-gamma-scanner (TGS) method for assaying heterogeneous drummed SNM/TRU waste.

Like the well-established segmented-gamma-scanner (SGS) method, the TGS method uses a high-purity germanium (HPGe) detector to count gamma-ray emissions from the drum and to measure the transmission of gamma rays through the drum from an external source. The passive drum emissions (typically from ^{239}Pu and ^{235}U , although nearly any gamma-emitter can be assayed) are the basis for the assay, while the transmission measurements are used to correct for the attenuation of gamma rays in the drum matrix. The SGS method makes a single count for each gamma ray of interest in each of several horizontal layers of the drum and estimates attenuation corrections based on a uniform-layer assumption. The TGS method improves on the SGS method by performing low-resolution tomographic emission and transmission scans on each layer, yielding coarse images of the matrix density and radionuclide distributions. The matrix density image is used to compute point-to-point attenuation corrections for the emission image. In other words, the gamma-ray attenuation corrections used in the TGS method are based on the actual distribution of radionuclides and absorbing matrix, rather than on a one-case-fits-all assumption about the distribution. The result is a significant improvement in assay accuracy for heterogeneous drums.

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We reported earlier on results obtained with a one-sixth-scale drum scanner¹ and on the construction of our full-scale 208-L-drum experimental prototype scanner.² Since that time, the scanner configuration has been modified to give significantly improved counting efficiency. In this paper, we report on the performance of our prototype device as it is now configured. We are currently constructing a well-engineered, field-ready TGS system that has the same basic configuration as our prototype. This turnkey scanner, which is scheduled to be ready for testing and evaluation in the summer of 1993, will have essentially the same accuracy as our experimental prototype.

II. EXPERIMENTAL DETAILS

A. Scanner Configuration

In its current configuration, our experimental prototype TGS uses a 15.2-cm-deep collimator with a 2.5:1 aspect ratio (compared to the 9:1 aspect ratio ordinarily used in single-photon emission computed tomography, or SPECT). With this low-aspect collimator, our prototype TGS has 60% of the counting efficiency of a typical SGS using an equivalent HPGe detector. Thus, a 28-min TGS assay will have the same sensitivity as an analogous 17-min SGS assay. (With its 70%-efficiency HPGe detector, our newer TGS unit will actually have a better overall sensitivity - in the same assay time - than many existing SGS units.) Other recent changes in the scan configuration include (1) the collection of 150 two-thirds-second counts on each layer instead of 100 one-second counts; (2) the reduction of the layer thickness to 5.7 cm, giving 16 layers per drum instead of 15; and (3) the use of an improved image reconstruction algorithm. Otherwise, the scanner configuration is the same as described earlier.²

We used the computer code TGS_FIT³ to reconstruct TGS images and obtain radionuclide masses. TGS_FIT offers a number of image reconstruction options. The approach followed here uses the algebraic reconstruction technique (ART) to reconstruct transmission (density) images and the expectation maximization (EM) method to reconstruct the attenuation-corrected emission (radionuclide intensity or mass) images. The images produced have a resolution of one hundred 6.1- by 6.1- by 5.7-cm volume elements (voxels) per layer. We normally scan an additional layer below the drum, giving a total of 17 layers, or 1700 voxels.

B. Mock-Waste Drums

To evaluate the accuracy of the TGS, we made a series of assays of a single 98.9-g metallic ²³⁹Pu source placed at different heights and radial positions within mock-waste drums of varying densities and degrees of homogeneity. The assumption behind this test is that a single point source is the

most difficult case, because it has the highest degree of heterogeneity of the emitting radionuclide and, thus, is the distribution most likely to result in a large assay bias. This is without doubt true for SGS assays; whether it is strictly true for TGS assays is a matter of current study. The few assays we have performed using multiple (2-4) sources showed noticeably better accuracy than similar assays of a single source, which tends to support the assumption.

We used a relatively large ²³⁹Pu source to obtain good counting statistics in all measurements, as we are interested in gauging the accuracy of the method without the complicating effect of poor statistics. As is well documented elsewhere,⁴ metallic ²³⁹Pu particles are subject to self-attenuation (the so-called "lumping" problem), which results in a low assay bias when the average particle size exceeds ~ 1 mm. This can be corrected for (to a point) in both SGS and TGS assays using the differential absorption technique. However, our 98.9-g source is too large for this correction method to be used. The apparent mass of the source, based on the intensity of the 413.7-keV gamma-rays used for assays, is only 13.0 g. Since we are only concerned here with matrix corrections, we will ignore this difficulty and treat the source as having a mass of 13.0 g; that is, references to the "true" mass should be understood to mean the apparent mass of 13.0 g.

The mock-waste forms that we used are described below:

- Case I: No drum (i.e., source mounted on a free-standing, low-Z source holder).
- Case II: A three-layer drum, with a homogenous damp sand layer in the lower third of the drum ($\rho = 2.0 \text{ g/cm}^3$), a homogeneous polyethylene bead layer in the middle third ($\rho = 0.9 \text{ g/cm}^3$), and air in the top third.
- Case III: A heterogeneous, moderate-density drum (average layer $\rho = 0.2$ to 0.9 g/cm^3) containing aluminum scrap, slabs of 5.1-cm-thick polyethylene, and large styrofoam blocks.
- Case IV: A heterogeneous, high-density drum (average layer $\rho = 0.3$ to 1.5 g/cm^3) containing assorted electronic scrap mixed with rags, lab coats, booties, and cardboard boxes.

Each of the 208-L drums had thin-walled, upright aluminum tubes imbedded in the matrix at different radial positions to allow reproducible insertion of a source (or sources) into the matrix. In a series of assays involving different positions of the source, vertical spacings of 5.1 cm were used so that the source would not always be at the same relative position within a layer (recall that the layer thickness is 5.7 cm).

To put the TGS performance into perspective, we compare the TGS assay results for Cases II and IV with SGS assay results for the same source positions in the same drums. We obtained the SGS measurements by modifying our experimental prototype TGS unit to perform SGS assays. All this required in terms of hardware modification was replacing the TGS collimator with a 15.2-cm-deep SGS-type slit collimator. We then used a separate software package to drive the scanner and collect the data in SGS mode. The data were analyzed using standard SGS methods.⁵

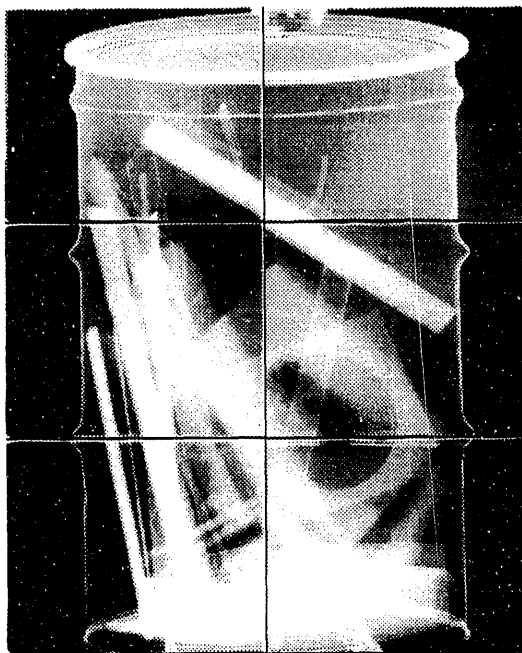
Our experimental prototype uses a relatively weak ^{133}Ba transmission source (~ 0.5 mCi, compared to the preferred source strength of ~ 10 mCi) for analysis of the 413.7-keV peak in ^{239}Pu . To compensate, we used a two-pass approach, with a 28-min emission scan and a separate, 9-h extended transmission scan. The assumption here is that a 9-h scan with the weak source is essentially equivalent to a “normal” 28-min scan with a full-strength transmission source. For multiple assays of the same drum, a single transmission scan was used for all the (emission) assays in a series. This probably represents the largest departure from realistic conditions in our experimental data. A “real” TGS device, like current SGS devices, would use a stronger source, probably ^{75}Se instead of ^{133}Ba . The normal mode of operation would be a one-pass scan, in which the transmission and emission data are collected simultaneously.

III. RESULTS

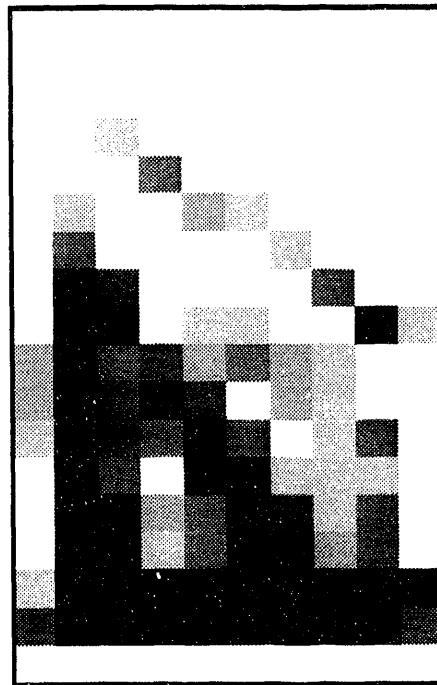
Figure 1(a) shows a ^{60}Co radiograph of the aluminum scrap mock-waste drum (Case III in the text); Fig. 1(b) shows a tomographic projection (summed side view) of the TGS transmission image for the same drum. This projection is equivalent (in concept) to the radiograph in 1(a), so the TGS image quality can be judged by comparison. We can see that while the TGS density image is too coarse to observe details, it matches the radiographed image quite well and accurately reveals the gross features of the matrix. In addition, the corresponding emission image in Fig. 1(c) clearly shows the position of the ^{239}Pu source inside the drum.

A. Case I: Assays of a Free-Standing Source

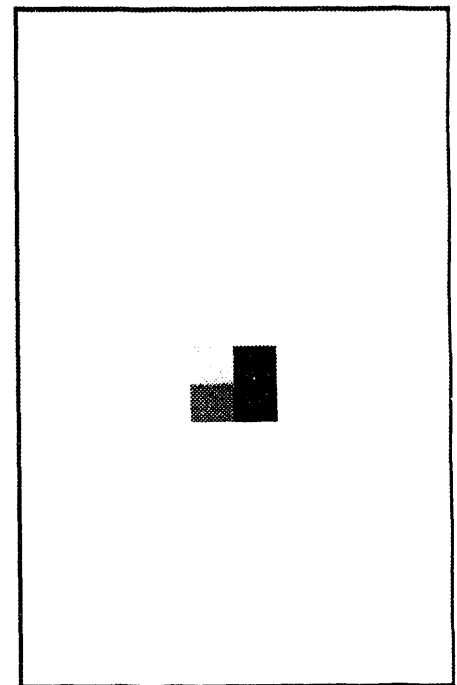
An important design goal was to achieve a nearly uniform point-source response throughout the active assay volume of the TGS in the absence of any gamma-ray attenuation; that is, for a free-standing point source. This condition does not hold for SGS, which will give different results for a point source at the center of the active volume than for a source at the periphery. Also, for point sources near the drum periphery, there will generally be a 10 to 15% assay difference caused by vertical variations; that is, a source halfway between two layers will assay 10 to 15% higher than one in the center of a layer. Nor can one assume that these problems will disappear in a tomographic assay.



(a) ^{60}Co radiograph



(b) density tomogram



(c) emission tomogram

Figure 1 - Side views of the 208-L aluminum scrap mock-waste drum (Case III). All views were taken from the same angle. (a) A ^{60}Co radiograph of the drum. (b) A tomographic projection (summed side view) of the TGS transmission image; this projection is equivalent to a coarsely digitized radiograph and should be compared to the radiograph in (a). (c) A tomographic projection of the TGS emission image of a 98.9-g ^{239}Pu source placed in the center of the drum; the darker voxels indicate regions of higher radioactivity.

To evaluate the uniformity of response of our prototype TGS, we made 52 assays of a free-standing source (Case I above) at different positions within the TGS assay active volume (that is, within the volume defined by a 208-L drum). The horizontal positions were more-or-less uniformly distributed radially, with some at the (horizontal) center of a voxel, some in-between two voxels, some at the corner of four voxels, and others placed simply at random. For each of these horizontal positions, assays were made at four different vertical positions in increments of one-eighth of a layer (0.76 cm). The assays involved between 14,000 and 20,000 total net counts per assay, with corresponding standard deviations (in the total counts) between 0.88% and 1.04%. Thus, with a uniform spatial response and with no statistical error amplification in the image reconstruction process, the 52 assays in this series would be expected to exhibit a standard deviation of $\sim 0.95\%$ (the average).

Figure 2 shows the measured error distribution (frequency histogram) for the series of free-standing-source assays, compared with the statistical-error-only distribution.

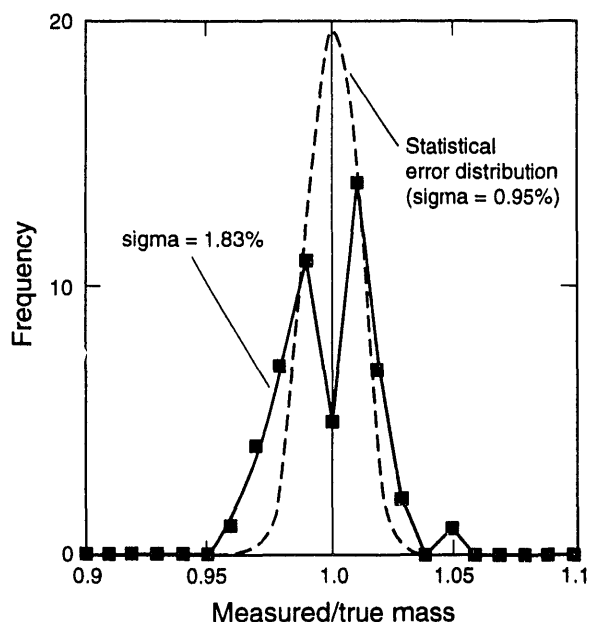


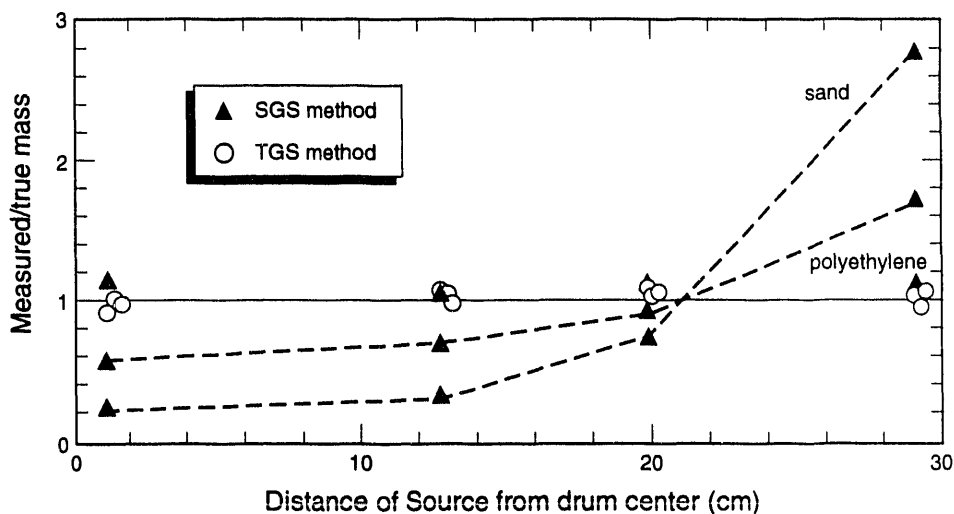
Figure 3 - Comparison of TGS and SGS assay accuracy, expressed as the ratio of the measured-to-true mass, as a function of source position for a single 98.9-g ^{239}Pu source in uniform layers of sand ($\rho = 2.0 \text{ g/cm}^3$), polyethylene beads ($\rho = 0.9 \text{ g/cm}^3$), and air. This three-layer drum is referred to as Case II in the text.

The observed distribution, which is not quite gaussian (it appears to be slightly bi-modal), has a standard deviation of 1.83%. Assuming that the statistical and systematic errors combine in quadrature, we can estimate that the maximum standard deviation attributable to positional variation - in the absence of any gamma-ray attenuation - is 1.55%.

B. Case II: Assays in Uniform Matrices

Figure 3 shows the results (expressed as a ratio of the measured-to-true mass) of TGS and SGS assays of the 98.9-g ^{239}Pu point source in homogeneous matrices of sand, polyethylene beads, and air, as a function of the distance of the source from the drum center (Case II). The SGS assay values for the sand and for the polyethylene bead layers are connected with dashed lines to emphasize the upward trend as the source is moved from the center of the drum to the outside. This trend is easily understood in terms of the SGS homogeneous drum assumption.

Figure 2 - The error distribution of 52 TGS assays of a free-standing source at various positions (Case I). The purely statistical error distribution, based on nuclear counting statistics in the raw data, had an average standard deviation of 0.95%, as indicated by the overlaid gaussian function. The additional error is a measure of the uniformity of the spatial response.



In the SGS assays, the same attenuation correction factor is applied to all scans of a given layer, regardless of the radial position of the source. In the sand matrix, a source is attenuated by a factor of 12 more in the center of the drum than at the outside, so with SGS - no matter how good the correction - there must always be a factor of 12 difference between the assays for these extreme cases. A uniform distribution requires a correction factor somewhere in-between the extremes, and so the SGS assay under-corrects sources in the center and over-corrects sources at the outside of the drum.

The attenuation of gamma rays in polyethylene beads is less severe than in sand; the variation in assay value as a function of position is roughly half that seen in the sand matrix. The general trend, however, is the same. As would be expected, the SGS assays with no matrix (air layer) show no significant bias as a function of position.

In contrast to the SGS results, the TGS assay results are uniformly accurate at all positions in all three matrices. This is because the TGS method applies attenuation corrections that are specific to the source positions, as determined by the image reconstruction process. It must be stressed that sand is a difficult matrix and requires longer than normal assay times for good counting statistics (we used 9-h emission counts for the 3 inner positions in the sand matrix to obtain 1% or better statistics). Even so, the accuracy obtainable is impressive. Consider that the TGS assay for the center of the sand layer applied an attenuation correction factor of 25 and gave a result within 8% of the true value.

C. Case III: Assays in the Aluminum Scrap Drum

Gamma-ray attenuation in complex materials is non-averaging, in that an absorber composed of alternating zones of high- and low-density material attenuates less than a uniform absorber having the same average density. Thus, it could be argued that matrices with a fine-grained, complex structure (that is, with variations on a smaller scale than the TGS resolution) will not be correctly assayed. The simple response to this (for both TGS and SGS assays) is that the non-averaging effect should be approximately the same for the transmission source as for the gamma rays coming from inside the drum. This is probably true for many drums. However, it is easy to imagine cases where the proximity of an internal source makes the matrix appear either more or less attenuating than it does to the more distant external source.

The moderate-density aluminum scrap mock-waste drum (Case III) was used to test the effect of a fine-grained, heterogeneous matrix on TGS assay accuracy. As can be seen (to an extent) in the radiograph in Fig. 1(a), the matrix for this drum is made up of small-diameter aluminum ($\rho = 2.7 \text{ g/cm}^3$) rods,

bars, and odd-shaped pieces supported in a matrix of lighter materials. In addition to the aluminum scrap, there are also a few steel and brass pieces. A matrix of this complexity might easily "fool" the TGS assay by virtue of the non-averaging effect just described.

Figure 4 shows the error distribution in 48 assays of the ^{239}Pu source at 16 vertical positions in each of 3 radial positions in the Case III (aluminum scrap) mock-waste drum. The average standard deviation in the raw data was 1.6% (as illustrated by the overlaid gaussian distribution). The standard deviation in the TGS assay values was 5.5%, implying a maximum of 5.3% systematic error. This is good accuracy and gives us some confidence that good attenuation corrections can be obtained even in a complex matrix. Still, the detailed distribution of errors appears too broad-tailed to be gaussian. It looks more like the 5.3% error is the sum of a narrower error distribution (say, 2 to 3%) and a smaller, broad outlier distribution (up to the largest error of 14%).

D. Case IV: Assays in the Electronic Scrap Drum

Figures 5(a) and 5(b) compare the results of 60 SGS and TGS assays of the 98.9-g ^{239}Pu source at various positions within the electronic scrap mock-waste drum (Case IV). The SGS transmission values for the external ^{133}Ba transmission source ranged from 0.017 to 0.41. The drum matrix, while complex, can be broadly broken down into a region of high

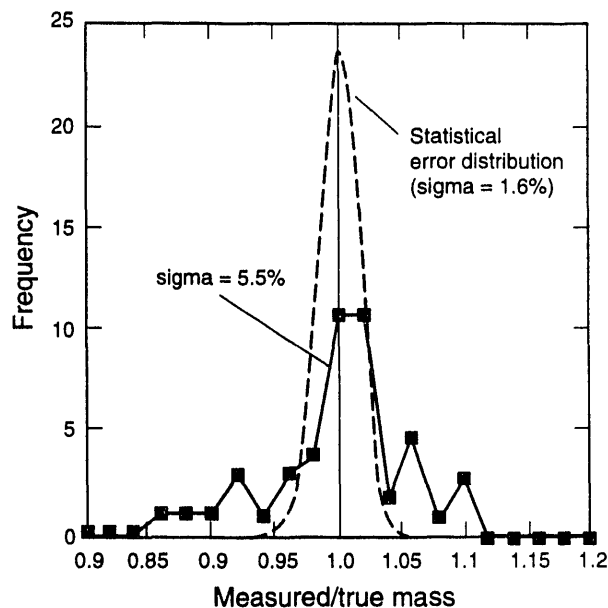


Figure 4 - Error distribution of 48 TGS assays of a ^{239}Pu source in different positions in a heterogeneous, moderate-density 208-L mock-waste drum (Case III). The purely statistical error distribution in the raw data averaged 1.6%, as indicated by the overlaid gaussian function. The additional error is a measure of the accuracy for this set of assays.

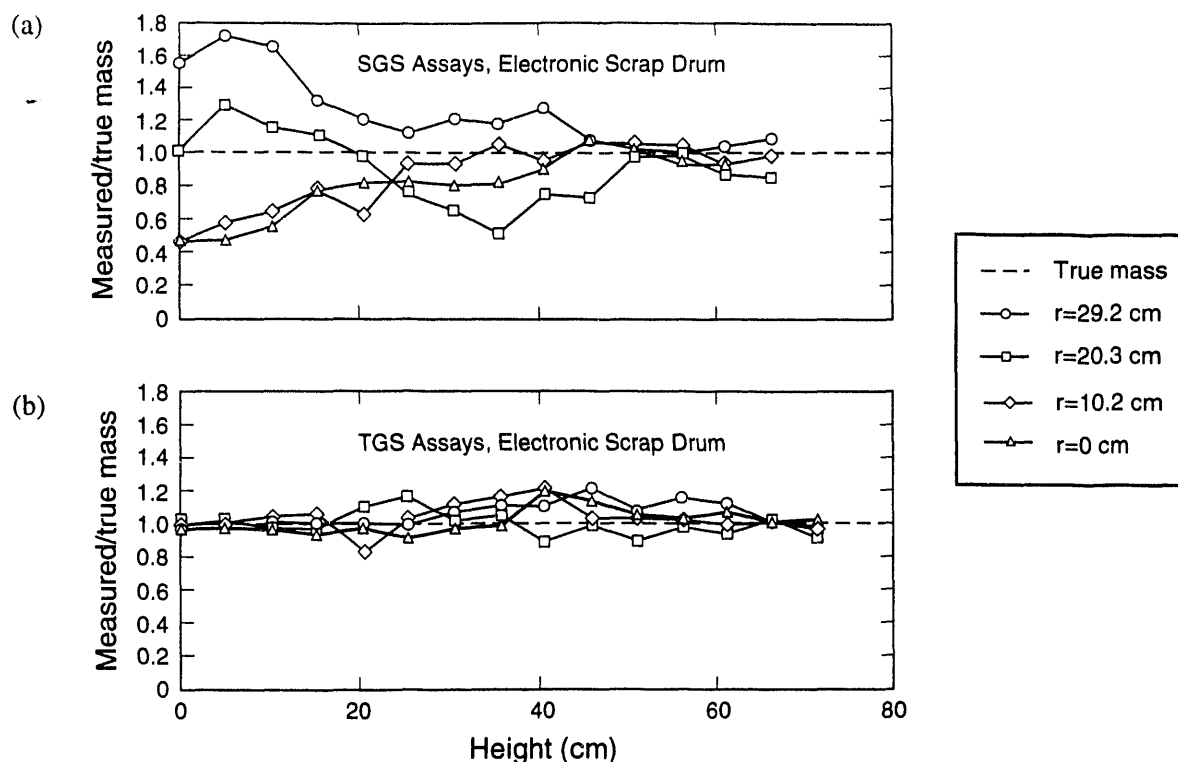


Figure 5 - Comparison of SGS and TGS assays of a ^{239}Pu source at 60 different positions in a heterogeneous mock-waste drum. This drum (Case IV in the text) contains dense electronic scrap filled in with labcoats, booties, and empty cardboard boxes. The (SGS) average transmission of 356-keV ^{133}Ba gamma rays through the drum ranges between 0.017 and 0.412. (a) SGS assay results as a function of height in the drum, for various radial positions (r). (b) TGS assay results for the same positions.

density (in the bottom), a region of moderate-to-high density (in the middle), and a region of low-to-moderate density (at the top). The magnitude of the variations in SGS assays closely follows this division, with the largest variations (from a factor of 2.2 too low to 1.7 too high) in the high-density region and the smallest variations in the low-density region. The standard deviation for the 60 SGS assays was 26.1%.

The TGS assay values for the same drum (Fig. 5b), in contrast, are clustered closely about the correct value, having a standard deviation for the 60 assays of 8.4%. The purely statistical errors in the assays (based on the error in the raw data) ranged from 1 to 5%, with most cases closer to 1%. We assume that the additional variation in the TGS results is mostly systematic error.

IV. CONCLUSION

The data presented here demonstrate the superior accuracy of TGS assays, compared with SGS assays, for very heterogeneous radionuclide distributions in moderate- to high-density matrices. Our intention was not to raise questions about the accuracy of SGS; in all likelihood, only a small

fraction of the SNM/TRU waste drums produced in present and future nuclear fuel cycles will be as heterogeneous as those used in this study. Moreover, the matrices studied included some extreme cases. A sand matrix would normally be rejected as being too dense for an SGS assay, and the density of the electronic scrap drum was at the margins of acceptability. Our intention, rather, was to illustrate that the TGS gives accurate results even in difficult cases. Because of the potential for errors, it is usually recommended that the SGS method be used only for drums with low-density matrices, or with moderate- to high-density matrices that are known to be uniform. The practical advantage of the TGS method is that it allows moderate- to high-density drums that are not definitely known to be homogeneous to be assayed with confidence, extending the range of drums that can be assayed.

An additional advantage over all other current NDA methods is that a TGS assay provides the operator with a visual image of the drum matrix and SNM distribution. The TGS emission image can be used, for example, to locate and quantify radioactive "hot spots" within a drum. This would be useful for repackaging drums that exceed SNM/TRU load limits.

The principle disadvantage of the TGS method, compared with the SGS method, is its longer assay time. Because even heterogeneous low-density drums (drums with a maximum layer density of 0.5 g/cm³ or less) can be accurately assayed with an SGS, we expect that future systems will combine the SGS and TGS methods in one unit.

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