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

INTRODUCTION

Future nuclear fuel cycles are expected to generate 08-L waste drums that have no high level contamination and e classified as "contact handled," but that contain unknown nounts of ^{239}U, ^{239}Pu, and other special nuclear material SNM) and transuranic (TRU) isotopes. The safe and eco nomic disposition of these drums will require non-destructive ssay (NDA) to measure the amount of SNM/TRU waste in ch drum. Moreover, in the light of current regulatory trends, seems likely that future regulations will dictate that all such nms be well characterized. This is already the case with RU waste, which is subject to a growing assortment of rules at either presume a knowledge of the TRU loading or ophcitly 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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cale drum scanner[1] and on the construction of our full-scale 208-L-drum experimental prototype scanner.[2] Since that time, he scanner configuration has bee n modified to give signifiantly improved counting efficie: cy. In this paper, we report n the performance of our prototype device as it is now onfigured. We are currently constructing a well-engineered, ield-ready TGS system that has the same basic configuration s our prototype. This turnkey scanner, which is scheduled to e ready for testing and evaluation in the summer of 1993, vill have essentially the same accuracy as our experimental rototype.

I. 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-hoton emission computed tomography, or SPECT). With this ow-aspect collimator, our prototype TGS has 60% of the ounting efficiency of a typical SGS using an equivalent HPGe detector. Thus, a 28-min TGS assay will have the same ensitivity as an analogous 17-min SGS assay. (With its 70%-fficiency HPGe detector, our newer TGS unit will actually ave a better overall sensitivity - in the same assay time - than any existing SGS units.) Other recent changes in the scan onfiguration include (1) the collection of 150 two-thirds-econd counts on each layer instead of 100 one-second counts; 2) the reduction of the layer thickness to 5.7 cm, giving 16 yers per drum instead of 15; and (3) the use of an improved nage reconstruction algorithm. Otherwise, the scanner onfiguration is the same as described earlier.[2]

We used the computer code TGS_FIT[3] to reconstruct GS images and obtain radionuclide masses. TGS_FIT offers number of image reconstruction options. The approach llowed here uses the algebraic reconstruction technique ART) to reconstruct transmission (density) images and the pectation maximization (EM) method to reconstruct the temation (corrected emission (radionuclide intensity or ass) images. The images produced have a resolution of one ndred 6.1 - by 6.1 - by 5.7 cm volume elements (voxels) per yer. We normally scan an additional layer below the drum, ving a total of 17 layers, or 1700 voxels.

B. Mock Waste Drums

To evaluate the accuracy of the TGS, we made a series assays of a single 98.9 g metallic ^{239}Pu source placed at fferent heights and radial positions within mock waste rms of varying densities and degrees of homogeneity. The sumption behind this test is that a single point source is the 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 ^{239}Pu 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 ^{239}Pu 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 homogeneous damp sand layer in the lower third of the drum (ρ = 2.0 g/cm^3), a homogeneous polyethylene bead layer in the middle third (ρ = 0.9 g/cm^3), and air in the top third.

Case III: A heterogeneous, moderate density drum (average layer ρ = 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 ρ = 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 for sources within 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)

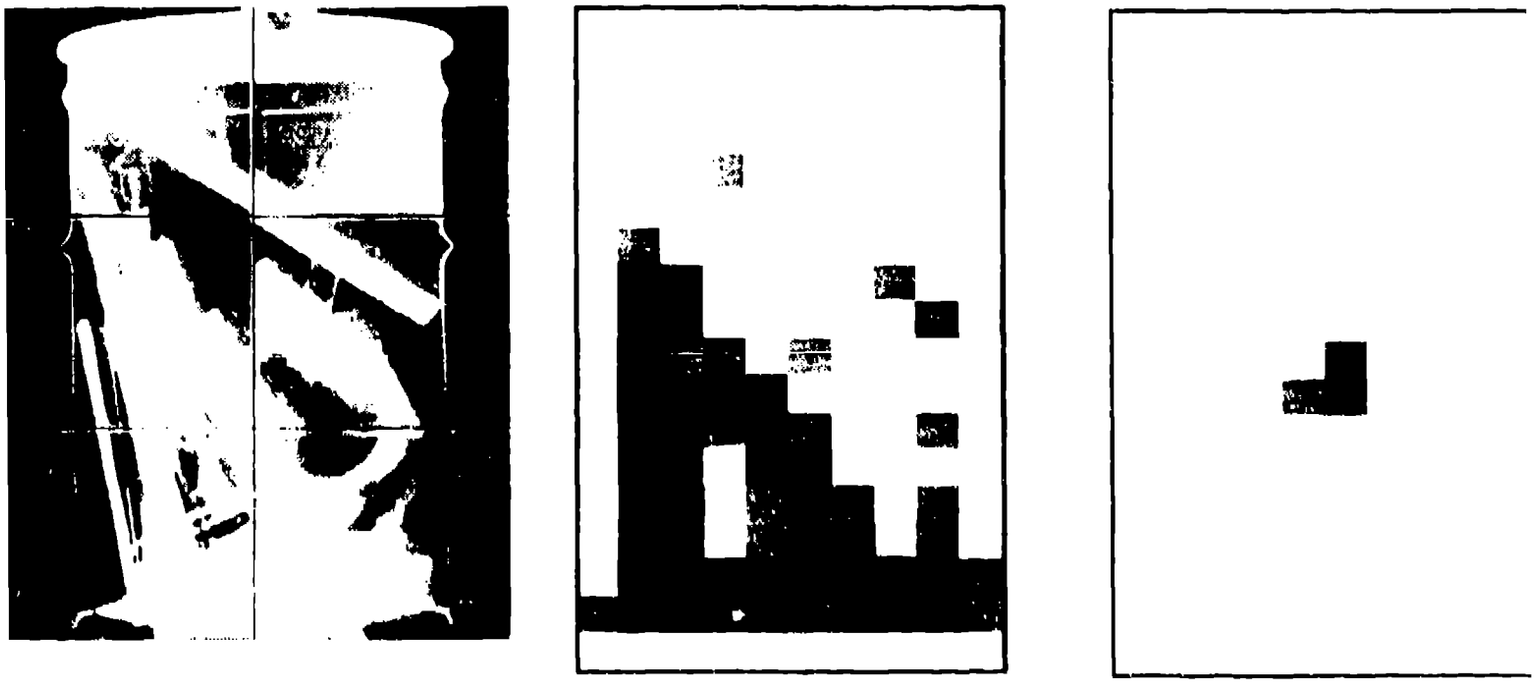
compare the TGS assay results for Cases II and IV with SGS assay results for the same source positions in the same drums. We obtained the SGS measurements by modifying our experimental prototype TGS unit to perform SGS assays. All this required in terms of hardware modification was replacing the TGS 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.

Figure 1(a) shows a ^{57}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) ^{57}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 ^{57}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.

GS, we made 52 assays of a free-standing source (Case I ove) at different positions within the TGS assay active lume (that is, within the volume defined by a 208-L drum). e horizontal positions were more-or-less uniformly distrib-d radially, with some at the (horizontal) center of a voxel, ne in-between two voxels, some at the corner of four xels, and others placed simply at random. For each of these rizontal positions, assays were made at four different tical positions in increments of one-eighth of a layer 76 cm). The assays involved between 14,000 and 20,000 al net counts per assay, with corresponding standard viations (in the total counts) between 0.88% and 1.04%. us, with a uniform spatial response and with no statistical or amplification in the image reconstruction process, the 52 ays in this series would be expected to exhibit a standard viation of ~ 0.95% (the average).

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

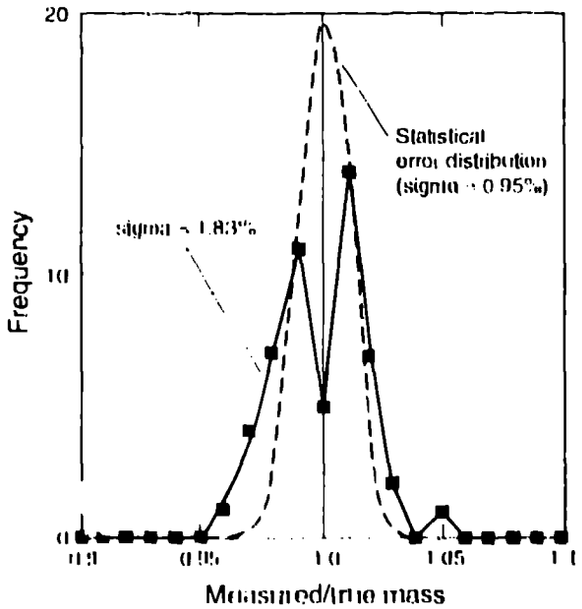
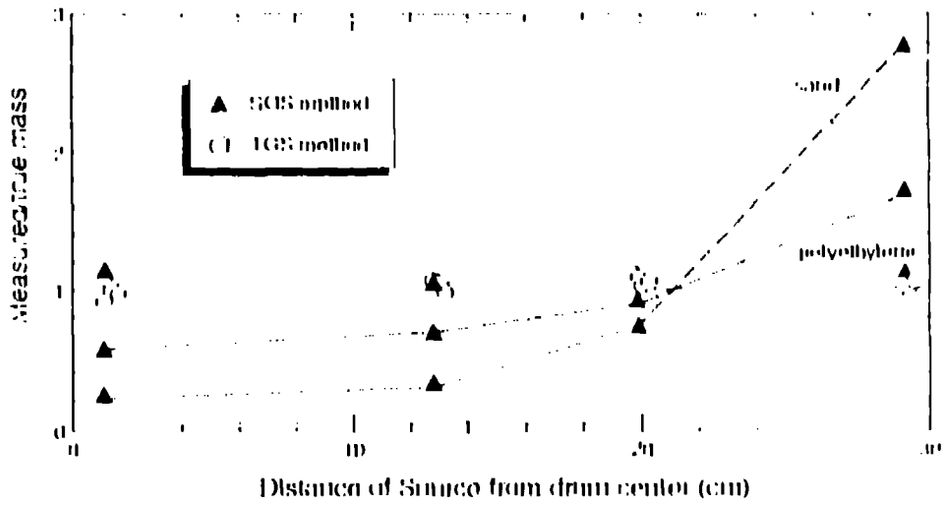


Figure 2 - The error distribution of 52 TGS assays of a free standing source at various positions (Case I). The purely statistical error distribution, based on nuclear counting statistics in the raw data, had an average standard deviation of 0.95%, as indicated by the overlaid gaussian function. The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

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 (ρ = 2.0 g/cm^3), polyethylene beads (ρ = 0.9 g/cm^3), and air. This three layer drum is referred to as Case II in the text.



applied to all scans of a given layer, regardless of the radial sition of the source. In the sand matrix, a source is attenued by a factor of 12 more in the center of the drum than at e outside, so with SGS - no matter how good the correction - ere must always be a factor of 12 difference between the says for these extreme cases. A uniform distribution reires a correction factor somewhere in-between the exmes, and so the SGS assay under-corrects sources in the nter and over-corrects sources at the outside of the drum.

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

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

C. Case III: Assays in the Aluminum Scrap Drum

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

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

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

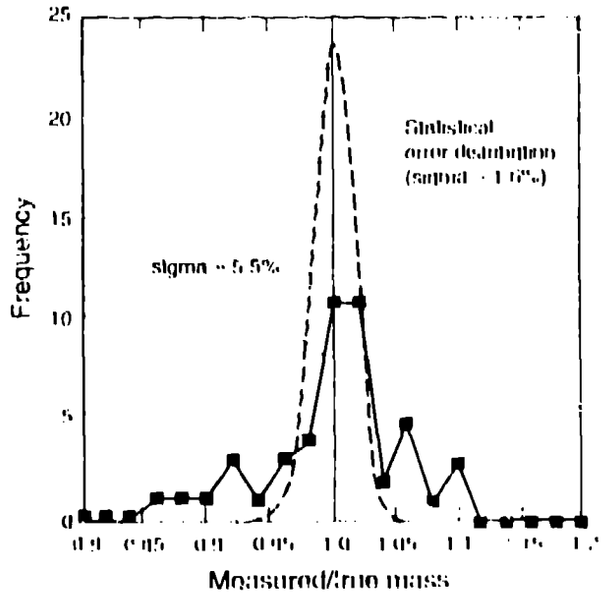
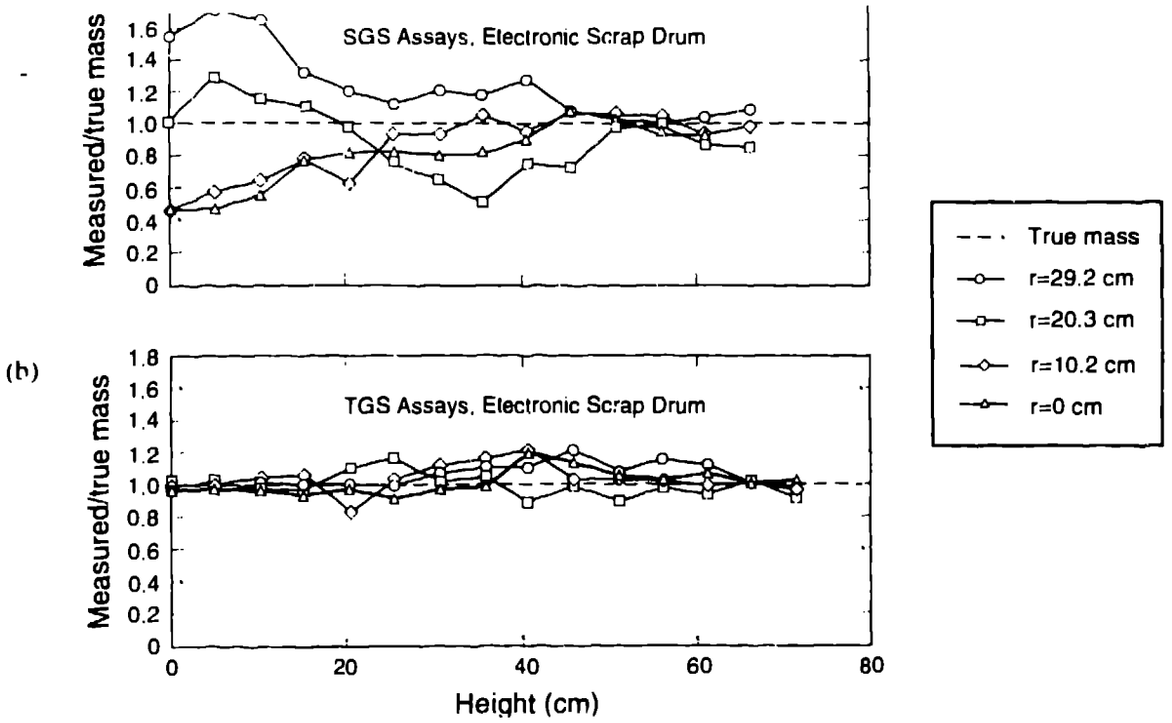


Figure 4. Error distribution of 48 TGS assays of a ^{239}Pu source in different positions in a heterogeneous, moderate density 208 L mock waste drum (Case III). The purely statistical error distribution in the raw data averaged 1.6%, as indicated by the overlaid gaussian function. The additional error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



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

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

The TGS assay values for the same drum (Fig. 5b), in trast, are clustered closely about the correct value, having andard devia.. n for the 60 assays of 8.4%. The purely istical errors in the assays (based on the error in the raw r) ranged from 1 to 5%, with most cases closer to 1%. We me that the additional variation in the TGS results is stry systematic error.

CONCLUSION

The data presented here demonstrate the superior racy of TGS assays, compared with SGS assays, for very rogeneous radionuclide distributions in moderate- to high sity matrices. Our intention was not to raise questions nt 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 homogenous 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.

ared with the SGS method, is its longer assay time. Because ven heterogeneous low-density drums (drums with a maxi-ium layer density of 0.5 g/cm^3 or less) can be accurately ssayed with an SGS, we expect that future systems will ombine the SGS and TGS methods in one unit.

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