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A DEMONSTRATION OF THE GROSS COUNT TOMOGRAPHIC GAMMA SCANNER (GC-TGS) METHOD FOR THE NONDESTRUCTIVE ASSAY OF TRANSURANIC WASTE

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Abstract
We examined the accuracy and sensitivity levels for three variations on the TGS method: the original TGS method using a high-purity germanium (HPGe) detector to measure net areas of full-energy gamma-ray peaks; a modified HPGe-detector method that uses net areas for the transmission analysis and the gross count TGS (GC-TGS) method for the emission analysis; and a NaI-detector method that uses the GC-TGS method exclusively. We found that while the accuracies of the methods were comparable, the GC-TGS method boosted the sensitivity per detector by a factor of approx. two for the HPGe GC variation and four for the NaI method. The implications for improved TGS scanner design are discussed.

INTRODUCTION

The tomographic gamma scanner (TGS) method for the nondestructive assay (NDA) of transuranic (TRU) waste in 55-gal drums[1] has proven to be accurate for assaying a wide range of heterogeneous waste forms, but is limited in usefulness by its moderate sensitivity and 1-h assay time. Using multiple high-purity germanium (HPGe) detectors in place of the single detector in current systems can reduce the assay time, the minimum detectable activity (MDA), or both. Because the MDA decreases as the square root of the number of detectors, significant improvements in sensitivity are difficult to achieve. For example, reducing the MDA by a factor 10 would require 100 detectors. Considering the complexity and high cost of HPGe detectors and the logistical problem of supplying them with liquid nitrogen, arrays of more than a few HPGe detectors may not be feasible.

NaI detectors are well-suited for use in large arrays of 100 or more detectors because of their simple operation and low unit cost, but the net-area techniques used in TGS do not work well with NaI because of its poor energy resolution. Whole-spectrum methods such as spectrum stripping or response-function fitting generally give good results, but until recently there was no simple method for measuring and applying gamma-ray attenuation corrections to whole spectra. As is described in ref [2] and [3], the material basis set (MBS) formalism used in ordinary TGS[4] can be applied to response-function fitting of NaI spectra both to solve for MBS transmission images and to apply the corresponding attenuation corrections in the emission imaging problem. In this report we evaluate the effectiveness of the gross count tomographic gamma scanner (GC-TGS) method, a modification of the TGS method that utilizes whole-spectrum techniques derived from the MBS model.

BACKGROUND

While the GC-TGS transmission analysis involves logarithmic response-function decomposition of transmission-source spectra — a subtractive process that amplifies statistical error — the emission analysis uses the summed counts from the emission spectra directly, with only ambient background subtracted (hence the term "gross count"). For this reason, the GC-TGS approach improves the sensitivity per detector compared with net- area methods, so that a lower MDA is achieved with a given number of detectors. This is true for both NaI and HPGe detectors, and one application examined here uses the GC-TGS method to obtain better precision for existing single-HPGe-detector TGS systems. For similar sizes, NaI detectors are more sensitive than HPGe detectors, so the improvement in sensitivity per detector when replacing HPGe with NaI is even greater.

Rawool-Sullivan, et al.,[3] examined multiple scattering in GC-TGS emission imaging and found it to be a significant effect. Reflected gamma rays are (mostly) downscattered into the continuum and are automatically eliminated when using conventional HPGe detector net area methods. When using whole spectra, however, scattered gamma rays are counted and would be interpreted by image reconstruction algorithms as radioactive sources proximal to the scattering object. Rawool-Sullivan, et al., found that a simple single-scatter model based on the Klein-Nishima formula[5] was sufficient to account for these reflected gamma rays, and this model has been implemented in recent versions of the TGS_FIT image reconstruction software[6].

The methods outlined here have been applied with encouraging results to the correction of NaI spectra transmitted through unknown absorbers in handheld monitor applications.[8,9] In particular, the success of the GC-MBS* method in determining the isotopic composition of binary mixtures of radionuclides transmitted through absorbers of unknown composition[9] is significant for the GC-TGS method. As was noted in ref [2], a potential limitation on the usefulness of GC-TGS as it is currently formulated is that the isotopic composition of the material being assayed is assumed to be known. The concern was that at a Pu mass loading below approx. 1 g (the nominal lower limit for gamma isotopics measurements with an HPGe detector) all gamma-emitters will look the same and contamination with non-TRU isotopes such as ^{60}Co or ^{137}Cs could inflate the Pu mass estimates significantly. However, the work of Miko, et al.,[9] suggests that many types of isotopic measurements will be feasible with the GC-TGS approach down to a Pu loading of much less than 1 g using data collected routinely during the assay.

EXPERIMENTAL DETAILS

We evaluated three variations on the TGS method:

- The HPGe method. This is the "standard" TGS method using continuum-subtracted net areas of full-energy gamma-ray peaks.

* In the context of handheld monitors, the similar method is called the gross count material basis set (GC-MBS) technique rather than GC-TGS.

- The HPGe/GC method. This uses an HPGe detector with a net-area analysis for the transmission scan and the GC-TGS analysis for the emission scan. It is done simultaneously with the HPGe method by collecting sum data on additional regions-of-interest (ROI) during the emission scan.

- The NaI method. This uses a NaI detector and applies the GC-TGS analysis to both the transmission and emission data.

The remainder of this section describes the experimental arrangements used in these evaluations.

THE TGS PROTOTYPE SCANNER

The original TGS prototype scanner from TA-18 at Los Alamos was used to perform all scans. The scanner has been modified since it was last described in ref. [1]. Only the modifications are described here. The most important change was the replacement of the source-holder arm, a U-shaped piece of tubing that extended around the drum and held the transmission source in a fixed position relative to the detector during scans, with a separate up-down/left-right scanner that moves in parallel with the identical detector scanner on the opposite side of the drum. This enabled the use of lead shielding and collimation on the transmission sources, which allowed us to safely use stronger sources than were used in ref. [1] and a correspondingly shorter transmission scan time. All transmission scans were done for 28 min (100 s per layer, 17 layers per scan) with a combined transmission source of 10 mCi ^{133}Ba plus 10 mCi ^{137}Cs. Emission scans were done the same way, but with the transmission sources removed.

The maximum load that can be handled by our detector scanner is approx. 75 kg, so a minimal amount of lead was used in building the detector collimator. The collimator opening was the same diamond shape described in ref [1], with a 7.2-cm diagonal (50 cm^2 area) opening. The depth of the collimator (from face to detector) was 14.3 cm and the distance from the collimator face to the drum center was 34.6 cm. This is a high-efficiency geometry compared with other Los Alamos TGS systems, which are more tightly collimated and are sized to handle 84-gal overpack drums. The collimator had wall thicknesses of 5.1 cm of lead on the sides and 2.5 cm on the top and bottom. There was no front or back shielding. All measurements were performed in the basement of the Accelerator Development Laboratory (ADL) at TA-18, which has low to moderate ambient background rates, mostly from the concrete in the floor and walls. A TGS scanner operating in a high background environment would require significantly more shielding to achieve the sensitivity levels described here. Conversely, a highly shielded system operating in a moderate- or low-background area should achieve even better sensitivities.

The 35%-efficient Ortec GEM-type HPGe detector used in the HPGe and HPGe/GC methods was interchangeable in the scanner with the 5.1-cm diameter by 5.1-cm thick Bicron model 2M2/2 NaI detector used for the NaI method. The NaI detector was

wrapped with a 0.3-cm thickness of copper sheet, around which was wrapped heating wire, then additional copper sheet to match the diameter of the HPGe detector housing. The heating wire was used with a temperature probe and thermostatic control to maintain the detector at a constant temperature just above that of the room in order to minimize gain shifts. The geometries for the HPGe, HPGe/GC, and NaI methods were equivalent except for the slightly smaller diameter of the NaI detector crystal (5.1-cm diameter versus 5.7-cm for the HPGe detector). Data acquisition and scanner control were accomplished using the WIN_TGS[10,11] software. The signal processing electronics were the same as described in ref. [1]. An Ortec model 918 MCA set for 4K conversion was used to collect spectra from both detectors.

REGIONS OF INTEREST FOR GC SCANS

The WIN_TGS data acquisition program allows three ROIs to be set — a peak ROI and two continuum ROIs — for each full-energy peak to be assayed. An unlimited number of these 3-ROI sets can be specified for transmission or emission scans. In each tomographic view, the sum of the counts within each ROI is saved in the output data file in raw integer form without conversion to net areas or count rates. The C utility program MAKE_ROI.EXE was used to automatically generate ROI files readable by the WIN_TGS program to be used in the GC methods. For the HPGe/GC method, our procedure was to use the WIN_TGS ROI editor to create a regular full-energy-peak ROI file with ROI sets at 414 keV and other emission energies for ^{239}Pu emission scanning. This "normal" ROI file was then modified by MAKE_ROI to fill in the gaps between existing ROIs with additional ROIs of up 32 channels width. This in effect saves an irregularly compressed but complete spectrum while preserving all the ROI sums needed for assays based on the full-energy peaks, so that both the HPGe and HPGe/GC analysis can be done on the same scan.

The ROI files used with the NaI detector used no regular peak ROIs, but were compressed uniformly by 32 channels (to 256-channels) across the entire spectrum. Not all of the ROI sums were used in later analyses (the TGS_FIT software allows selection of the ROIs to be used).

SURROGATE DRUMS AND EMISSION SOURCES

We performed transmission and emission scans on a set of 5 metallic weapons grade (WG) Pu sources at different radial positions inside two surrogate drums. The Pu source masses and shapes were 1) 0.228-g disk; 2) 0.620-g disk; 3) 3-g cylinder; 4) 30-g cylinder; and 5) 100-g cylinder. Because of self-shielding, these exhibit lower effective masses in gamma-ray assays. Assuming no there is no self-shielding in the 0.228-g source, the effective masses based on emission of the 414-keV gamma ray are 1) 0.228 g, 2) 0.380 g, 3) 0.93 g, 4) 5.24 g, and 5) 11.0 g. The effective masses are somewhat higher for the GC methods because of the greater penetration, an effect also observed with matrix corrections. With the NaI method, the 100-g Pu source has an effective mass of 18.7 g based on no self-shielding with the 0.228-g source.

The surrogate drums had vertical aluminum tubes placed at radii of 0 cm, 12 cm, and 25 cm from the drum center to allow positioning of the sources at any height at those radii. The first surrogate was filled halfway with SiO_2 beads of average density 1.04 g/cm^3. The unfilled upper half of the drum was used to represent an empty drum (i.e., the non-interfering matrix case), while the bottom half was used to represent high-density uniform matrices such as uniform sludge. Measurements in this drum were made at two heights only: at the center of the SiO2 half and at the center of the empty half. Henceforth, we will refer to these two cases as the "empty drum" and the "SiO_2-beads" drum. The second drum was three-fourths filled with loosely stacked blocks of high-density polyethylene. Measurements were made at four heights in this drum, which represents a non-uniform moderate-density matrix. This will be referred to as the "polyethylene-blocks" drum.

DATA ANALYSIS

All scan data were analyzed using version 2.0 of the TGS_FIT software, which has been modified since its use in ref. [1] to support MBS multiple-energy and gross-count analysis, among other improvements. For all three TGS methods we used the NNLS (non-negative least-squares) algorithm of Lawson and Hanson[11] for transmission imaging and single-layer emission pre-imaging and the EM (expectation maximum) algorithm of Shepp and Vardi[12] for three-dimensional fitting of the full emission image. All three methods used a two-material basis set of elemental Pb and Al. In the HPGe method, calculated attenuation coefficients for Pb and Al at the transmission and emission peak energies were used as bases. The two gross count methods (HPGe/GC and NaI) used basis spectra of ^{133}Ba+^{137}Cs and WG Pu transmitted through 0.64 cm of Pb and through 15.2 cm of Al. No attempt was made to find optimal basis materials or thicknesses. The fitting region for spectral analysis omitted the low-energy and x-ray region below 200-keV in transmission imaging and 300-keV in emission imaging of WG Pu. No doubt better choices of basis sets and fitting regions will emerge once more experience is gained.

RESULTS AND DISCUSSION

ESTIMATES OF ASSAY ACCURACY

To estimate the relative accuracies of the three methods we assayed the 30-g and 100-g Pu sources individually at several radial and vertical positions within the surrogate drums. Our reasoning was that statistical error is small in these assays, so deviations of the measured mass values from their true values represent systematic inaccuracies of one kind or another. We quantize this inaccuracy as the "bias" of the assay method, where bias refers here to the std. dev. for a set of standard measurements. While this non-universal measure of inaccuracy only has meaning for a particular set of measurements, it seems a reasonable basis for comparing the three methods to one another.

Figure 1 shows the ratio of the measured-to-true mass for assays of our 100-g and 30-g WG Pu standards at different radial positions inside the empty drum, the heterogeneous polyethylene-block drum, and the uniform SiO_2-beads drum. The results are equally spaced along the horizontal axis, ordered by increasing overall attenuation correction

factor for the 414-keV gamma-ray as measured in the HPGe-method (the correction factor is the inverse of the uncorrected measured-to-true mass ratio shown in the figure). At each horizontal position the assay results are shown for each of the three methods for the same drum and source position. Although the points coincide, the attenuation corrections required for the GC methods are generally much smaller than for the HPGe method. For example, the largest HPGe-method correction factor required was 14.9, for the center of the SiO_2-beads drum. The HPGe/GC- and NaI-method corrections for the same case were only 2.95 and 4.38, respectively.

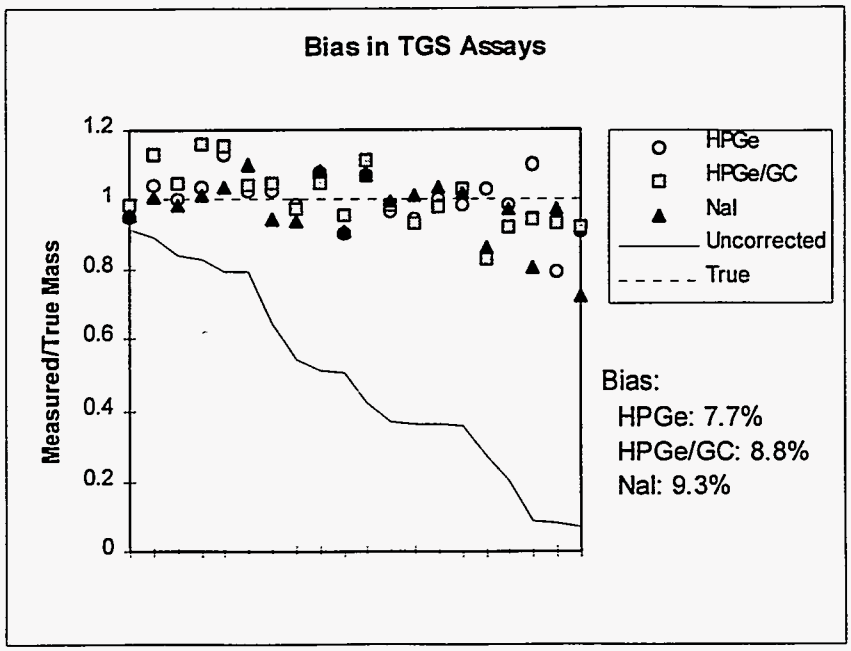

Fig. 1 Measured-to-true mass ratio for assays of the 30- and 100-g Pu source in the empty drum, the polyethylene-blocks drum, and the SiO_2-beads drum. The uncorrected value is for the 414-keV gamma-ray using the HPGe method, and is the inverse of the correction factor required for the assay.

What we see in figure 1 is that the assay bias, as measured by the std. dev. in the assay value over all drums and source positions, is only slightly larger for the HPGe/GC and NaI methods (8.8% and 9.3%, respectively) than for the HPGe method (7.7%). These differences are not significant in terms of potential NDA applications and may be attributable to our inexperience with GC-TGS methods.

ESTIMATES OF ASSAY PRECISION

To estimate the precisions and MDAs of the three methods we made 5 to 15 replicate assays on each of the three smallest Pu sources (0.228, 0.620, and 3 g) at the centers of the empty drum, the polyethylene-block drum, and the SiO_2-beads drum. The standard

deviations for the replicate assays give a conservative estimate of the precision for the different masses at each measurement point. Figure 2 shows the measured RSD for the measurements on the three standards at the center of the empty drum. What we observe in the figure is that both the GC methods have significantly better precision than the HPGe method, and that the NaI method has better precision that the HPGe/GC method. The differences are more pronounced with the 3- and 0.62-g sources than for the smaller source. For the 3-g source, the HPGe, HPGe/GC, and NaI methods had RSDs of 6.2%, 2.8%, and 1.3%, respectively. This is reasonable, as the HPGe/GC and NaI methods had 5.6 and 19.6 times the net counts seen with the HPGe method.

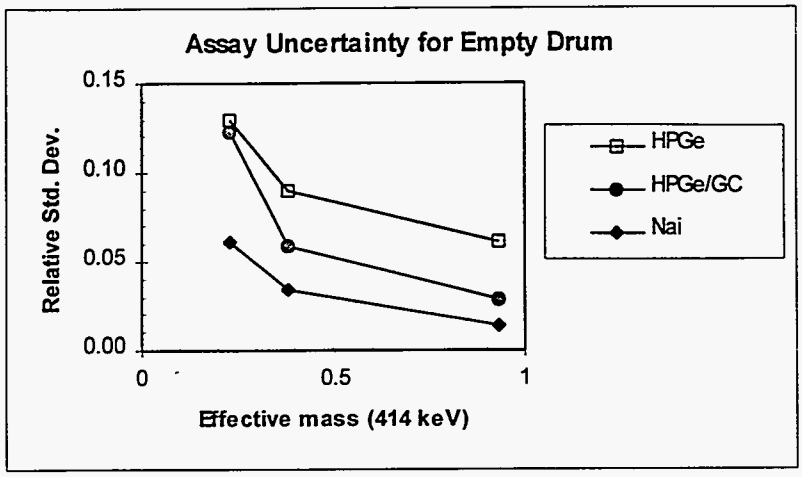

Fig. 2 Assay uncertainty (RSD) determined from replicate scans of the 0.288-, 0.620-, and 3-g Pu sources in the center of the empty drum. The RSDs are plotted against the effective mass of the sources based on the 414-keV line.

TABLE I. Assay Precision for Pu Sources Based on Replicate Measurements

Drum	**Effective mass**	**RSD, HPGe**	**RSD, HPGe/GC**	**RSD, NaI**
empty	.228 g	12.9%	12.3 %	6.13%
"	.38	8.96	5.92	3.36
"	.93	6.16	2.8	1.34
polyethylene	.228	(36.33)	17.7	16.7
"	.38	14.0	6.91	6.70
"	.93	6.17	5.48	1.17
SiO_2	.228	(68.5)	(139)	(27.04)
"	.38	(61.6)	(54.2)	5.98
"	.93 g	(34.8%)	5.04%	1.99%

* based on 414-keV gamma ray, HPGe method

( ) means the average assay result was low by 50% or more

Table I summarizes the RSD values measured in the empty drum, polyethylene-blocks drum, and SiO_2-beads drum. The general trends are the same for the polyethylene drum as for the empty drum, although the errors are much larger overall, exceeding the 3σ level at 0.228 g with HPGe method. The RSDs for the center of the SiO_2-beads drum illustrate the greater difficulty of assaying dense drums. The SiO_2 HPGe method RSDs are greater than 3σ for all three sources. The 3-g source RSDs were 34.8%, 5.0%, and 2.0% for the HPGe, HPGe/GC, and NaI methods, respectively, which implies there was some instability in the image reconstructions with the HPGe method that caused the error to become amplified. This may be attributable to the previously noted lower attenuation of the whole gamma-ray spectrum than of the 414-keV full-energy peak.

Statistical error in the transmission scan contributes to the final assay error. To measure the error introduced by the transmission image for the SiO_2-beads drum we analyzed the same emission scan of the 100-g source in the center of the drum paired with ten replicate transmission scans. Figure 3 shows the RSDs for the ten assay results for the HPGe and NaI methods for the 0.228-, .620-, and 3-g sources. As can be seen in the figure, the uncertainty due to the transmission part of the analysis is roughly a factor of ten less for the NaI method than for the HPGe method.

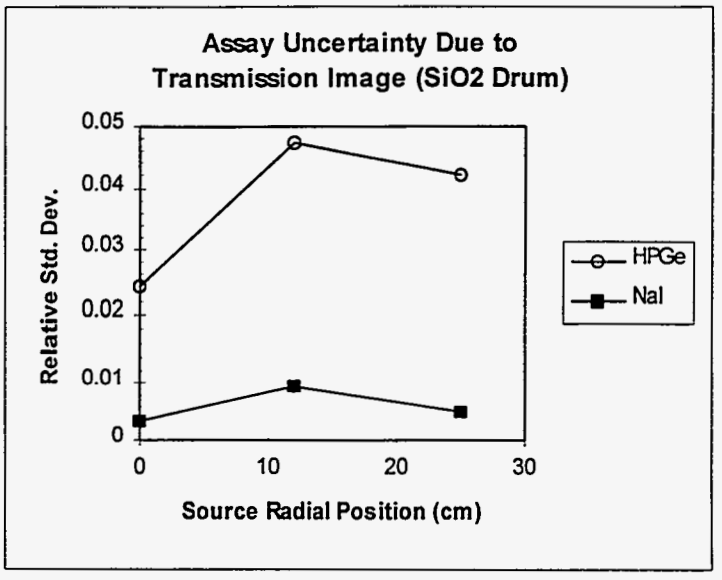

Fig. 3 The assay uncertainty due to the transmission image for the r=0 cm, r=12 cm, and r=25 cm radial positions in the SiO_2-beads drum.

DESIGN ISSUES

In ref [2] we estimated the MDAs attainable with the HPGe and NaI methods with varying numbers of detectors based a 0.6 g Pu sensitivity for the HPGe method and an assumed factor of 11 improvement in the sensitivity using the NaI method. This improvement was estimated using count rates for the entire spectrum, including the low-energy x-ray region. In our analysis here we used only the energy region from approx.

300-keV to 450keV for emission analysis, which contains approx. 20% of the counts in the spectrum. For design purposes we have revised our earlier estimates to use a nominal HPGe sensitivity of 0.5 g for one detector; a factor of 2 improvement using the HPGe/GC method; and a factor of 4 improvement using the NaI method. Based on these assumptions, Table II lists predicted MDAs for the 3 methods as a function of the number of detectors in an array for 1-h assays and for 10-min assays.

TABLE II. Estimated TGS Pu Minimum Detectable Activities for Various Array Sizes

	MDA for a 1-h Assay			**MDA for a 10-min Assay[a]**		
Number of detectors	**HPGe method**	**HPGe/GC method**	**NaI method**	**HPGe method**	**HPGe/GC method**	**NaI method**
1	500 mg	250 mg	125 mg	1580 mg	791 mg	395 mg
5	224	111	55	708	351	174
10	158	79	28	500	250	89
20	111	55	20	351	174	63
40	79	28	14	250	89	44
80	55	20	9.9	174	63	32
160	28 mg	14 mg	7 mg	89 mg	44 mg	22 mg

a. A 10-min assay may not be feasible with fewer than 5 detectors for mechanical reasons

Table II illustrates the difficulty of decreasing the MDA by increasing the number of detectors. Because of the square-root dependence, the incremental improvement becomes smaller with each detector added to the system. For the 1-h NaI assays, for example, going from 1 to 5 detectors drops the MDA by 70 mg (or 17.5 mg per detector), while going from 80 to 160 detectors drops the MDA by only 2.9 mg (.036 mg per detector). However, the use of arrays can simplify the mechanical design by eliminating motion modes, and for NaI arrays the resulting savings can offset the cost of the additional detectors. With multiples of 10 detectors, the left-right scan motion can be eliminated by using a fan-beam geometry. With 160 detectors, both the left-right and up-down scan motions can be eliminated, leaving only the relatively simple drum rotation. The entire assay would involve two rotations of the drum, once for transmission scanning, once for emission scanning. Our estimates are that the cost savings due to this simpler design would more than offset the cost of the detector array, which is estimated to be $175,000 (or about half of the cost of the ^{3}He detectors in a typical neutron assay system). More significantly, without the left-right/up-down motions, the 160-detector system would be compact enough to shield completely within a lead or steel enclosure, which could easily lower the MDA into the sub-milligram range.

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