Meeting the Challenge: How to Assay Diverse Drummed Waste Types with a Flexible Tomographic Gamma Scanner - 9472

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ABSTRACT

A new extended-range multi-modality tomographic gamma scanner (TGS) system is presented. New features for this system include a dual mode transmission beam that extends the dynamic range of TGS assays to drum densities that were previously unfeasible. The details of this system are described with particular emphasis placed on the unique elements of the system. Representative data highlighting the performance of the system with high-density drums are presented and discussed. These results demonstrate that extending the range of applicability for the TGS methodology is achieved.

INTRODUCTION

Gamma-emitting radioactive waste can show tremendous variability both in terms of nuclide composition and physical make-up and form. For facilities needing to assay or investigate small numbers of drums coming from diverse origins, a high resolution gamma spectroscopy instrument which offers a flexible approach is beneficial. A computer configurable tomographic gamma scanner (TGS) fitted with a dual intensity transmission source mechanism is one approach to providing high-performance multi-modality operation at reasonable cost of ownership.

In this work we describe a TGS solution designed to meet a need identified at Nuclear Engineering Seibersdorf. A TGS with a Programmable Logic controlled interleaved finger aperture, detector stand on

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a controllable slider, and a transmission source with a low-beam/high-beam attenuator was selected to give the range of sensitivities, modality options and density range sought.

In addition to the traditional TGS data acquisition and analysis option, in which spectral grabs are collected layer by layer as the item is rotated and scanned in front of the detector/Transmission beam and reconstruction algorithms are used to make maps of the linear attenuation and gamma-line activities present, this instrument also supports:

- TGS with uniform matrix and uniform matrix/activity analysis which is useful for highly attenuating items and the reconstruction algorithms account for layer cross talk explicitly
- SGS data acquisition which is useful for improving throughput on wastes where the degree of heterogeneity is known to be limited.
- Far field integrated gamma assays which can be compensated for matrix contents using, for example, net weight and can be applied to relative isotopic composition studies and to familiar items.
- A dual intensity transmission source in which the matrix density range can be extended by performing two transmission scans: one with a calibrated attenuator in place and one with it removed.
- Automated supervisor functions allowing the drums staged to be assayed automatically according to preloaded table or according to logic set-up ahead of time based on factors such as measured surface dose rate and drum weight.

Several other developments were undertaken in delivering this system. The Materials Basis Set (MBS) approach, in which the transmission versus energy for each voxel is represented by effective density of a few basis materials, was extended below the previous limit of 120 keV (which was due to the complication of the K-edges of the typical special nuclear materials U and Pu) to allow the 59.5 keV line from Am-241 to be covered. A methodology was developed, by which the high and low beam transmission data can be combined to create a hybrid transmission map that contains the best features of both source characteristics. Also ISOCS models (see later discussion) were developed so that a sourceless calibration transfer could be applied between measurement configurations.

In this paper we report the salient features of the measurement challenge and how this fed into the selection of the key functional capabilities of the system. Results generated by the TGS system are presented along with the uncertainties.

DESCRIPTION OF THE SYSTEM

The TGS is a system that combines high-resolution gamma-ray spectroscopy for net full-energy peak determination with three-dimensional single-photon attenuation coefficient images and three-dimensional single-photon emission images to quantitatively compute the activity of radioactive materials within drums in which the sources and/or matrix may be significantly heterogeneous [1]. In addition to this capability the system also has the capability to perform assays in the simple segment gamma scanner (SGS) or integrated gamma scanner (IGS) modes to increase throughput for drums in which the contents are better known and homogenously distributed. The transition from TGS to SGS to IGS effectively represents a coarsening of the voxel size used in the analysis.

Figure 1. Photograph of the TGS system during factory assembly. The transmission assembly is viewable on the left and the beam modulator and shutter mechanisms are visible. To the right is the germanium detector and surrounding aperture box. The drum is shown on the translation assembly in the raised position of the rotator above the roller conveyor.

The new features of this system over previous assemblies are the extension of the TGS technique to drums with average densities of greater than 1.5 g/cc, as well as the inclusion of the capability to assay down to energies as low as 45 keV for low-density low-Z drums. A photograph of the system with a drum in the assay position is shown in Fig. 1.

One of the key elements of a TGS system is the capability of creating a three-dimensional energy-dependent attenuation map of the assayed container. This is accomplished by measuring the transmission of a narrow gamma-ray beam passing through the container at multiple locations. Typically there are sixteen vertical layers in which there are 150 transmission measurements per layer at locations from the edge of the drum to the center while the drum is rotated.

The source of the transmission gamma rays is a highly collimated radioactive source that is located on the side opposing the germanium detector. Traditionally, the source of choice has been Eu-152 with typical activities of about 10 mCi [1]. With such a source, one can assay 208 liter waste drums up to densities of about 1.5 g/cc. Even for these moderate density drums the low-energy components (e.g. 122, 244, and 344 keV) from Eu-152 do not effectively penetrate the container. At higher densities the attenuation is such that a stronger source is required because even the high-energy components (1112 and 1408 keV) do not penetrate the drums with sufficient intensity. Thus for extending assays to drums with densities greater than 1.5 g/cc, one requires a stronger source and yet, due to the extreme attenuation, low-energy activity is highly non-penetrating and consequently not useful. To this end, a 250 mCi nominal activity ^{60}Co source was chosen for the system described here.

While a source of this strength is sufficient to penetrate drums up to high densities, it is also sufficiently strong that, when weakly attenuated, the intensity will saturate the detector signal due to excessive dead-time and pile-up. It is therefore necessary to have a low-intensity transmission mode. This is achieved by adding a movable attenuating block in front of the transmission source effectively creating a two configuration transmission source. A high-beam configuration (attenuator removed) for high-density applications, and a low-beam mode (attenuator added) applicable for density ranges from zero to about 1.5 g/cc. The transmission source assembly can be seen on the left-hand side in Figure 1. The large tungsten cylinder observable in this photograph is the beam stopper to put the source into a "safe" configuration. The attenuator is located between the stopper and the large source shield.

Since it is not known, *a priori*, the degree of attenuation for any particular view, a three-pass scanning protocol was established. In addition to the standard emission scan, there are separate low-beam and high-beam transmission passes. The decision of which beam intensity to use in the tomographic reconstruction is made on a view-by-view (i.e. data grab by data grab) basis at the time of analysis dependent on various acceptability criteria.

Prior to every TGS assay, a transmission normalization measurement is performed. This is performed with the source in low-beam mode only since it is impossible to measure directly the unattenuated source in the high-beam mode. The zero density high-beam transmission is related to the low-beam transmission by a previously determined ratio between the two settings. The method for determining this ratio is presented in Ref. [2]. This initial measurement also serves as a front-line quality-control check for the germanium detector operation.

While the use of a ^{60}Co transmission source is an effective method for extending the TGS method to higher density drums, the lack of low-energy lines limits the accuracy in determining the source attenuation at low energies. Traditionally, the TGS analysis uses a material basis set (MBS) analysis [3, 4] method to determine an effective Z (Z_{eff}) mass attenuation function based on a linear combination of a low-Z and high-Z mass attenuation function to observed gamma ray transmission data. The two high-energy gamma rays from ^{60}Co decay (1173 and 1332 keV) are generally insensitive to the Z of the matrix which reduces the effectiveness of the MBS approach; therefore, the user must select at the time of analysis the representative material based on the accepted knowledge of the drum. The material is represented in the analysis as the average proton number of its elemental components or Z_{eff} of the material. For energies above 200 keV, the choice of Z_{eff} is not a major contributor to the overall total measurement uncertainty.

In addition to tomographic scanning, it is possible to perform segmented gamma scan (SGS) assays. The SGS assay can be performed using either the high-beam or low-beam transmission measurement to perform a layer-by-layer density correction. For extremely dense drums (> 5 g/cc) in which even the high-beam transmission source cannot effectively penetrate, the system includes a multi-energy and density calibration in which the efficiency correction is performed based on the average density of the drum. The SGS efficiency calibrations are computed using Canberra Industries' In-situ Object Counting System (ISOCS) software [5]. With this software, it is possible to quickly compute the SGS efficiency over a wide range of energies, densities, and drum configurations.

This system also includes a software-integrated weigh scale, dosimeter, and bar code scanner. Based on the results of measurements from these components, the system can automatically set and perform any of the assays based on previously entered selection rules. A conveyor loading system allows up to six drums to be pre-loaded and automatically assayed, each with possibly a different method, without operator intervention.

DUAL-BEAM TRANSMISSION IMAGE RECONSTRUCTION

Key to extending the dynamic range of the TGS system is the capability to combine the measurement results of the low-beam and high-beam transmission measurements to form an accurate measurement of the container composition.

To illustrate this, the results of three assays are presented in this section. The first assay is a drum with homogenously distributed silica sand through the interior. It has an average density of about 1.6 g/cc. The second assay is a drum with a homogenously distributed matrix of concrete mixed with 20% by mass of steel shot. The drum has an average density of approximately 3.4 g/cc. The third drum is an in-homogeneous sampling of steel objects. While the drum has an average density of about 1.1 g/cc, the steel structures create a matrix with significantly higher local densities.

All the measurements performed using a set of gamma standard sources containing a mixture of nuclides that emit gamma-rays spanning most of the energy range of interest. The sources are in the form of rods and include the following nuclides: ^{133}Ba, ^{137}Cs, and ^{60}Co. The activity is uniformly distributed in an epoxy matrix and cast in a 9.53 mm outer diameter by 813 mm long aluminum tube with 0.89 mm thick walls. Both ends of the tube have 2 mm end plugs, thus making the active length of the sources to be 809 mm. For the sand-matrix and concrete-steel-matrix drums eight such rod source standards were distributed in a spiral pattern from the center of the drum to near the edge to simulate a volumetrically uniform distribution of activity. The sample-steel matrix drum has three co-linear source tubes. Three rods were placed in the central tube, three in a tube at 50% radius, and two in the outside tube at about 75% radius. The total activity for the eight rods is 249, 41.0, and 39.8 □Ci for ^{133}Ba, ^{137}Cs, and ^{60}Co respectively with a 95% confidence level uncertainty of about 4% in each case based on the manufacturer's specifications.

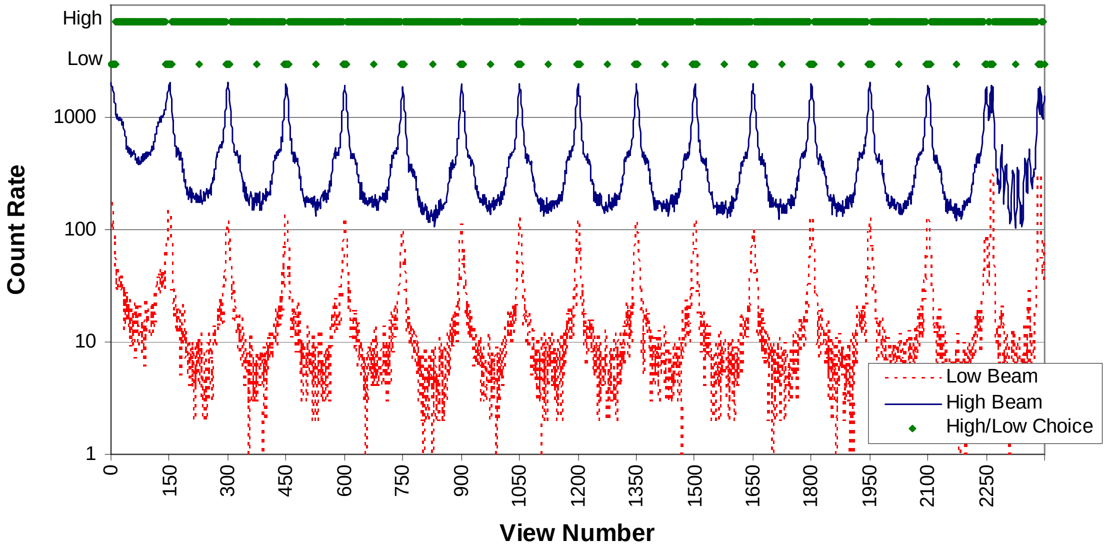

Figure 2. View-by-view count rate for the 1332 keV line from ^{60}Co transmission source passing through a sand matrix drum with an average density of 1.6 g/cc. The solid line represents the rates in high-beam mode, while the dashed lines are from the low-beam mode. The diamond symbols above the lines, indicate which mode was selected for a particular view. A high point indicates a high-beam mode selection, and a low point represents a low-beam selection.

A three-pass assay is performed in which 112.5 seconds are spent acquiring data at each of the 16 layers in each pass (low-beam transmission, high-beam transmission, and emission), for a total assay time of 1.5

hours per drum. For each segment, 150 individual measurements, each 0.75 seconds long, are acquired while the collimated detector and transmission source scan from the outer edge to the center of the drum and back to the same outer edge. During this scan the drum is fully rotated five times.

At the TGS analysis stage the corresponding view for each of the low and high-beam assays are compared, and the beam mode selection is made based on comparison of the statistical precision of each measurement, including the consideration of the measurement dead-time. A hybrid view-by-view is then produced as the combination of the two modes. A low-beam selection is added directly to the matrix, while a high-beam selection is scaled to the low-beam intensity by the predetermined energy-dependent attenuation factors. This hybrid view matrix is then used in the standard TGS analysis [6].

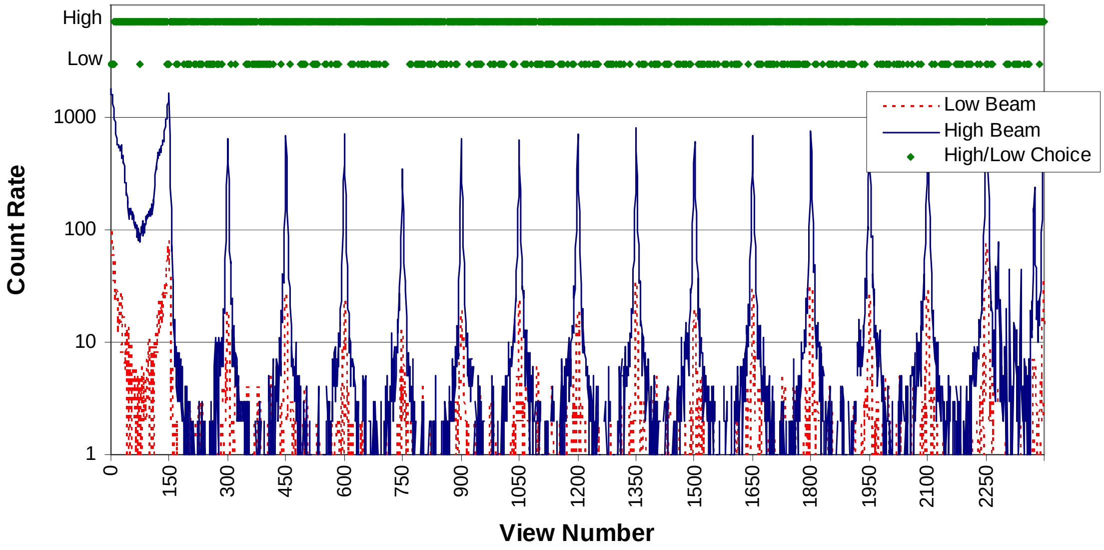

Figure 3. View-by-view count rate for the 1332 keV line from ^{60}Co transmission source passing through a Concrete/Steel matrix drum with an average density of 3.4 g/cc. The solid line represents the rates in high-beam mode, while the dashed lines are from the low-beam mode. The diamond symbols above the lines, indicate which mode was selected for a particular view. A high point indicates a high-beam mode selection, and a low point represents a low-beam selection.

Examples showing the comparison of the view-by-view low and high-beam transmission count rate sinograms for the three measurements presented here are illustrated in Figs. 2, 3, and 4, for the sand-matrix, concrete-steel-matrix, and sample steel-matrix drums respectively. These figures show both the low-beam and high-beam count rates for the 1332 keV line from the Co-60 transmission source. Also shown above these lines are the points that indicate whether the low or high beam was selected for a particular view.

Fig. 2 shows the sinograms for the sand drum matrix. In this figure one can observe spikes in the data a every 150 views. These spikes represent the increase in rate that occurs as the transmission/detector view approaches the edge of the drum. The trough between correspond to rates obtained for transmission/detector positions between the edge and center of the drum. There are a total of 16 trough regions which correspond to the number of vertical segments performed in the measurement with lower to higher segments proceeding from left to right. The somewhat increased rate in the lowest segment is due to the curvature of the drum bottom being sufficiently high to reduce the attenuation. The highest segment views show more variability because of the distribution of matrix material at the top of the drum.

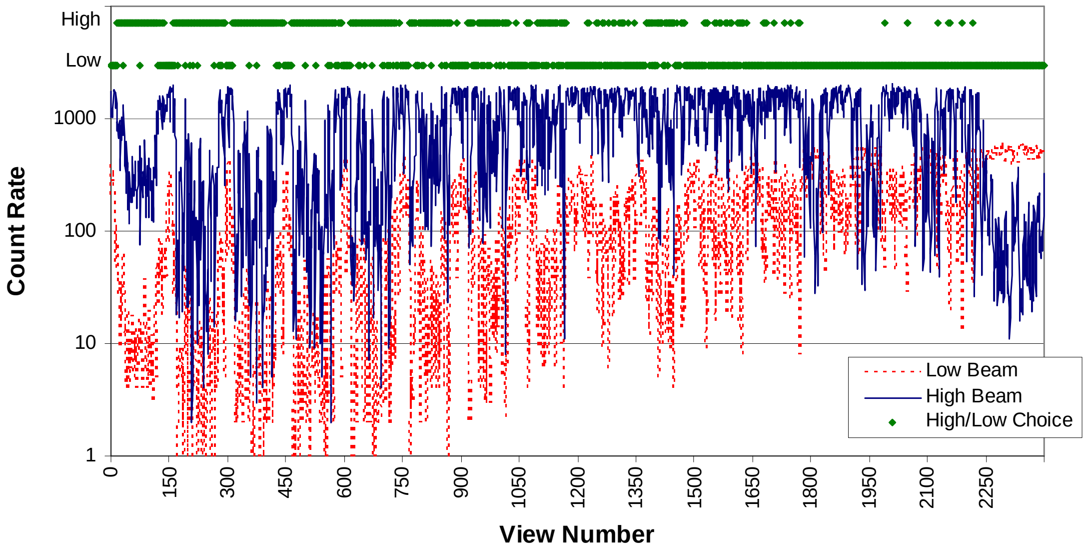

Figure 4. View-by-view count rate for the 1332 keV line from ^{60}Co transmission source passing through a heterogeneous steel matrix drum with an average density of 1.1 g/cc. The solid line represents the rates in high-beam mode, while the dashed lines are from the low-beam mode. The diamond symbols above the lines, indicate which mode was selected for a particular view. A high point indicates a high-beam mode selection, and a low point represents a low-beam selection.

By design the low-beam transmission is about 36 times smaller in this system than the high beam rates, but, as can be seen in Fig. 2, the trends for the low-beam views are very similar to the high-beam views. It should also be noted that while it appears that the high-beam count rates are reasonable for all views, when the rate exceeds about 1000 counts per 0.75 second view, the performance of the system is severely degraded due to pile up and dead time effects. Consequently, the results at these high rates do not accurately reflect the true transmission for these particular views.

Shown at the top of Fig. 2 are points that indicate which mode was chosen for a particular view. For almost all views, the high-beam was the selected mode; the only exceptions were for the views near the edges of the drum and the most central view at each segment.

Fig. 3 shows the transmission sinograms for the concrete-steel-shot matrix drum. Many of these features in this figure are similar to those from the sand-matrix drum (Fig. 2). One can easily observe the peaks due to the drum-edge measurements, as well as, the troughs for the mid-drum measurements. The transmission, however, is significantly less than what is observed for the sand drum. This naturally is due to the increased density of the drum. One can see from these results that even at the high-beam setting, there are mid-drum views that have little or no counts. As with the sand-matrix drum, the majority of views selected are from this high-beam mode. Perhaps surprising, however, there are more low-beam mode selections for this drum than for the sand-matrix drum. This is because in the mid-drum regions the transmission attenuation is so extreme that even the high-beam transmission views can have few to zero counts. In this situation, it is possible for an occasional random count or two in a low-beam view to produce a better uncertainty than the corresponding high-beam view and consequently be selected over the high-beam. In these extreme situations, the occasional selection of a low-beam view has negligible influence on the final results. Regardless, the high-beam mode is selected in over 80% of the views.

The third measurement presented in the paper is the result of the measurement on a drum heterogeneously filled with various steel components. While the drum itself has a moderate overall density of 1.1 g/cc, the steel constituents produce very high local densities (~ 7 g/cc). The transmission sinograms for the measurements on this drum are presented in Fig. 4.

Compared to the previous examples, the sinograms for this sample steel drum are much more complicated. There are numerous views in which the high-beam measurements exceed 1000 counts, and these correspond to "shine paths" which offer little attenuation. Even in these regions of low attenuation, there are views in which the count rates are strongly suppressed. These suppressed views correspond to cases in which a significant chunk of steel blocks the view.

For this example, the high-beam mode is selected for about 40% of the views, with the majority of these being in the lower portion of the drum where the number of steel components is greater. It is also interesting to note that at the top of the drum (the right most views in Fig. 4), the low-beam views have actually greater count rates than the high-beam views. This is because the drum is not fully filled and the transmission at the top of drum is essentially unattenuated except for the drum wall. In this case the detector is exposed to the full intensity of the source in the high-beam mode and the detector is completely saturated. While the data for that particular view in the high-beam mode is compromised, the detector recovers quickly from this condition when the incident rate is reduced, and because rate loss corrections are applied on a view-by-view basis using the reference pulser method, data in neighboring views with greater attenuation are valid.

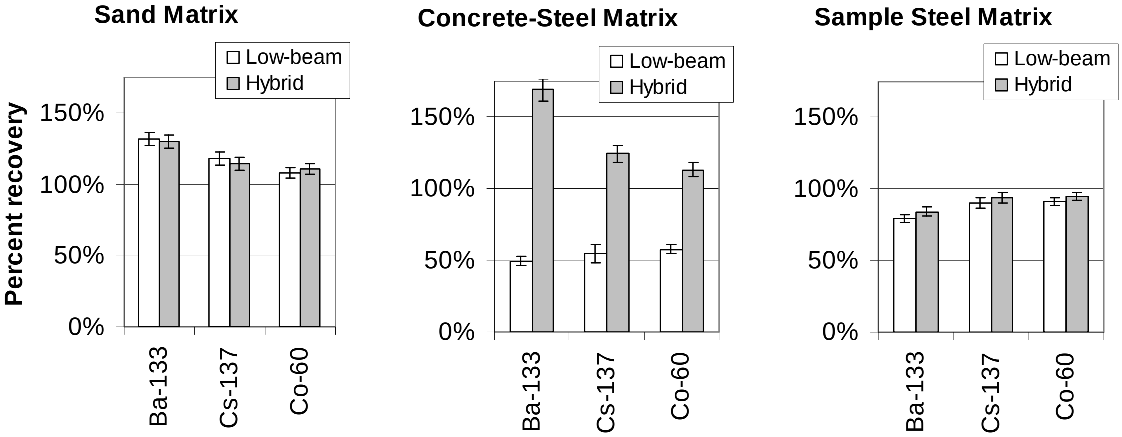

Figure 5. Comparison of the measured activity to the expected activity, represented as a percent recovery, for three drum measurements. The three drums have matrices of sand, concrete-steel, and a sampling of steel with densities of 1.6, 3.4, and 1.1 g/cc respectively. For each drum the percent recovery for ^{133}Ba, ^{137}Cs, and ^{60}Co are shown when TGS data are analyzed using only the low beam transmission data (Low-beam) or when using a hybrid of the low and high-beam transmission data (Hybrid).

ACTIVITY COMPARISON

For each of the three drums presented in the previous section the view-by-view emission activity for gamma-rays from ^{133}Ba, ^{137}Cs, and ^{60}Co were analyzed and corrected using two different analysis modes. The first option uses only the low-beam transmission mode to calculate the attenuation matrix, while the second option uses the hybrid transmission data to compute the attenuation matrix. The results are presented in Fig. 5.

The sand matrix drum has a density, 1.6 g/cc that is at the upper limit of the average densities that can be handled by standard TGS assays using a "traditional" TGS system. For the present extended range TGS system, one can observe that hybrid results give slightly better results compared to the low-beam only analysis. Similar results can be seen for the sample steel matrix drum in which the density is a more moderate 1.1 g/cc. For these two lower-density examples, the hybrid approach is not strictly required, but the results presented here show that the hybrid analysis can provide an overall improvement in the measurement results.

The most significant difference is observed for the high-density (3.4 g/cc) concrete-steel shot drum. In this case, the low-beam only analysis computes a source activity that is only about 50% of the certificate activity of the sources. If the hybrid transmission is used one obtains a significantly improved result compared to the low-beam only anlaysis for nuclides with higher energy lines (i.e. ^{137}Cs and ^{60}Co). For ^{133}Ba, the results are significantly over corrected, which is an indicator that counts at the interior of the drum are over estimated for the lines associated with this particular nuclide. Because low-energy gamma rays are so weakly penetrating in high-density drums, it is seen that the results of the TGS analysis can still be compromised if the activity from the low-energy lines is insufficient to produce statistically significant peak over background events from the central portion of the drum.

To illustrate the difference between the low-beam only and the hybrid analysis, representative reconstructed images from the concrete-steel drum are presented in Fig. 6. In this figure, one can observe the reconstructed images from both the low-beam only analysis and the hybrid analysis for the 662 keV gamma ray from ^{137}Cs. One can see that the low-beam only transmission image has been computed such that the majority of the attenuating voxels are located at the surface of the drum. The corresponding emission image also in turn was computed to have the majority of activity near the surface of the drum. Because of these conditions the source activity is significantly under computed.

In counterpoint to the low-beam only analysis, the hybrid results (right side of Fig. 6) show a transmission image in which the attenuating voxels are homogenously distributed throughout the drum. This is a more accurate representation of the true matrix distribution. In addition, the emission image has activity more consistently distributed throughout the matrix, which is also more representative of the true distribution. Consequently, the computed activity from the hybrid analysis is significantly better than the low-beam only results.

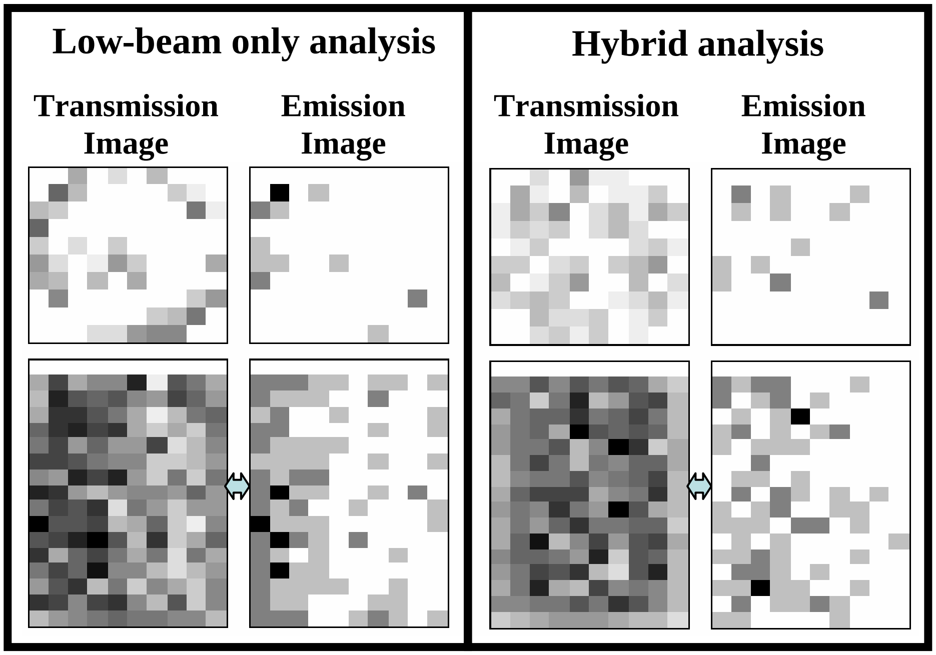

Figure 6. Transmission and emission images from the TGS analysis of the concrete-steel drum. The images are shown for the 662 keV gamma ray from ^{137}Cs, that result from a low-beam only transmission analysis (left panels) and from a hybrid transmission analysis (right panels). The four lower rectangular images are vertical projections of imaged results of the drum, while the upper square panels are horizontal slices through the drum at the point represented by the arrows in the lower panels. Note for the Hybrid analysis that the attenuation distribution in the transmission image is more properly homogenously distributed compared to the low-beam transmission image.

CONCLUSIONS

A multi-modal gamma-ray sensitive radioactive waste assay system has been developed based on tomographic gamma scanning principles. This system continues to provide the capabilities to automatically select and perform TGS assays, SGS assays with transmission, and/or SGS assays without transmission that are available in currently existing automated devices. The present system, however, extends the dynamic range of both the TGS and SGS to include the capability to effectively assay up to average drum densities of 3.4 g/cc for TGS and 6 g/cc for SGS. Key to this increase of the dynamic range is the utilization of a dual-beam ^{60}Co transmission source that can be run in a low-intensity attenuated mode, for low to moderate density drums, or a high-intensity mode, for high density drums.

Because the high-beam intensity is sufficient to completely saturate the detector, it was necessary to develop a process to combine the measurement results from low and high-beam transmission modes. The process of combining these modes to create hybrid view-by-view data has been demonstrated for three different drum configurations: a moderate density 1.6 g/cc sand-matrix drum, a high-density 3.4 g/cc concrete-steel-shot matrix drum, and a heterogeneous sample steel drum of 1.1 g/cc density. We have chosen to illustrate the transmission data using sinogram plots and find these to be highly diagnostic and visually intuitive to understand.

From these comparisons, it can be seen that at moderate densities the performance of the hybrid analysis is comparable to the low-beam only analysis, however, for the analysis of high-density drums it is critical to have the high-beam capability to be able to accurately assay waste drums in the TGS mode.

With the new extended range TGS system, it is now possible to assay a wider range of varying waste drums with a single system than what was previously available.

REFERENCES

[1] S. Croft, R. J. Estep, T. D. Anderson, R. J. Huckins, D. L. Petroka, and M. Villani, *A New Drum Tomographic Gamma Scanning System*, in proceedings of 25th Annual ESARDA (European Safeguards Research and Development Association) Symposium on Safeguards and Nuclear Material Management, Stockholm, Sweden, 13-15 May 2003. EUR 20700 EN (2003) Paper P096, ISBN 92-894-5654-X.

[2] S. C. Kane, S.Croft, P. McClay, R.J. Estep, W.F. Mueller, M.F. Villani, R. Venkataraman, *Extending the Dynamic Range of the TGS through the use of a Dual Transmission Beam*, Paper presented at the 47th Annual Meeting of the INMM (Institute of Nuclear Materials Management), Nashville, TN, USA, July 17-21, 2006.

[3] R.J. Estep, D. Miko and S. Melton, “Monte Carlo Error Estimation Applied to Nondestructive Assay Methods”, 7th NDA and NDE conference, Salt Lake City, May 22-26 2000.

[4] R.J. Estep, T.H. Prettyman and G.A. Sheppard, Comparison of Attenuation Correction Methods for TGS and SGS: Do We Really Need Selenium-75?, LANL report LA-UR-96-2575, INMM 37th Annual Meeting, July 28- August 1, 1996, Naples, Florida.

[5] *Model S573 ISOCS Calibration Software Users Manual*, Canberra Industries Document 9231013D V4.0, Meriden, CT, USA.

[6] R. J. Estep, *User’s Manual for TGS_FIT Version 2.2*, NIS6-QAP-00.33 Rev 2.2.