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An integrated Tomographic Gamma Scanning system for non-destructive assay of radioactive waste

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Abstract

The Tomographic Gamma Scanning (TGS) technique is a relatively new method in the field of non-destructive assay (NDA) of radioactive waste. The TGS technique combines High Resolution Gamma Spectrometry (HRGS) with Three-Dimensional (3-D) low spatial resolution transmission and emission image reconstruction techniques to achieve assay goals. When compared to the traditional methods such as Segmented Gamma Scanning (SGS), the TGS technique can yield better accuracies for cases where the radionuclide is distributed non-uniformly in a heterogeneous matrix. The TGS technique is ideally suited for low-to-moderate density waste matrices, say 1.0 g cm^{-3} or below for 55 US gal. drums, although it can be extended to higher densities by using alternative approaches to the design or analyses. Recently Canberra Industries designed, built and characterized four such TGS systems for nuclear power plant applications. Many of the design features and the end application itself set these TGS systems apart from the others that had been built previously. The four TGS systems are the first commercial grade systems that could quantify radionuclides contained in nuclear power plant waste, using the TGS technique. The TGS systems featured two different combinations of collimator and source–detector distance; a "near" geometry with a collimator aperture of 50.8 mm and a "Far" geometry with a narrower collimator aperture of 40.6 mm. For assaying drums with matrix densities greater than 1.0 g cm^{-3} and/or dose rates greater than 6 mSv h^{-1} the system could be configured as a SGS. In the SGS mode, five different assay geometries could be configured using different collimator, source-to-detector distance and absorber combinations. During operation, the appropriate assay geometry was selected automatically based on the drum weight (density) and dose rate measurements. The characterization and performance of the one of the TGS systems are discussed in detail for both TGS and SGS modes of operation. Quantitative results are presented for point source and rod source nuclide distributions. Transmission and emission images obtained in the TGS assays will be presented.

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1. Introduction

The Tomographic Gamma Scanner [1–3] (TGS) is a High Resolution Gamma Spectrometry (HRGS) based instrument that is being increasingly employed to perform non-destructive assay of radioactive waste. The TGS methodology combines low spatial resolution imaging techniques with HRGS and can yield quantitative results that are more accurate when compared to non-imaging methods in situations where radionuclide source distribution is non-uniform and the waste matrix is heterogenous. In a TGS assay, the waste drum is scanned with three degrees of freedom, i.e., rotation, translation and elevation. The TGS technique can yield accurate quantitative results for low-to-moderate matrix densities (0–1.0 g cm^{-3}) and for contact dose rates less than 6 mSv h^{-1}. Among the limitations of the TGS technique are the statistical fluctuations in the transmission and emission data. On the

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transmission side, the problem can be alleviated by using a transmission source with high activity (3.7E + 09 Bq or more).

An integrated waste assay system was designed and built to extend the dynamic range of matrix densities to 3.0 g cm^{-3} and dose rates up to 0.1 Sv h^{-1}. This was achieved by implementing a design that allowed the system to be configured as a Segmented Gamma Scanner (SGS). The SGS is a well-established NDA instrument that uses analytical techniques developed by Parker [4] to estimate matrix correction factors. In the SGS mode, the waste drum is scanned with two degrees of freedom, rotation and elevation. Matrix homogeneity is assumed for each layer, but matrix material can be different for different layers. To further extend the operational range of dose rates and assay high activity waste, lead absorbers of thickness 9 (A), 17 (B) or 24 mm (C) were introduced in front of the detector to prevent it from saturating.

2. System description

The integrated waste assay system described in this paper is the first one with the capability to assay, in the TGS mode, a wide variety of radionuclides such as those found in nuclear power plant waste as well as special nuclear material. A picture of the system is shown in Fig. 1.

The TGS mechanism is a modular design and consists of the following modules: detector vertical drive module, in-line rotation/translation module, transmission vertical drive module and a transmission source shield and shutter. Automation of vertical, lateral and rotational motions and of the transmission source shutter is controlled by a GE/Fanuc Process Logic Controller (PLC) and directed by Canberra's data acquisition and analysis software platform, NDA 2000. The system consisted of a 5000 mm^2 Broad Energy Germanium (BEGe) detector and a 15 mCi ^{152}Eu transmission source. The detector was housed inside a cylindrical lead shield of 50 mm annular thickness. The detector and the transmission source are accurately aligned with each other and also with the center of the rotating

Fig. 1. Industrial Tomographic Gamma Scanner.

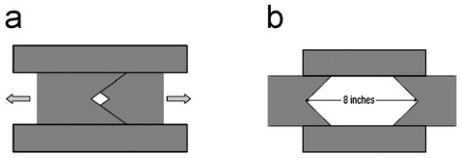

Fig. 2. (a) Narrow TGS (b) Wide SGS.

platform. The pulse processing electronics consisted of an RC Pre-amplifier, a Canberra Model 2060 Digital Signal Processor (DSP), an Accuspec B Board to facilitate high-speed data transfer and a Canberra Model 1654 NIM Reference Pulser. Rate loss corrections for each grab were performed using the reference pulser counts [5].

The collimator used was made of tungsten with a variable diamond-shaped aperture. The aperture is formed by six (three on each side) interleaved layers of sintered tungsten 25.4 mm thick that can be opened and closed to change either the spatial resolution of the TGS measurement or to switch over to an SGS measurement mode. Fig. 2a and b shows examples of TGS and SGS collimators used.

In the TGS mode two different collimator cum source-to-detector distance combinations were possible; a "near" geometry with a collimator aperture of 50.8 mm (Wide or W) and a "Far" geometry with a collimator aperture of 40.6 mm (Narrow or N). In the SGS mode, five different assay geometries could be configured using different collimator, source-to-detector distance and lead absorber combinations. These are as follows; "Near, Open, Wide" or NOW, "Far, Open, Narrow" or FON, "Far, absorber A, Narrow" or FAN, "Far, absorber B, Narrow" or FBN and "Far, absorber C, Narrow" or FCN. The "Wide" SGS collimator aperture was 203.2 mm, and the "Narrow" SGS collimator aperture was 101.6 mm. During operation, the appropriate assay geometry was selected automatically based on the drum weight (density) and dose rate criteria.

3. Operation in TGS mode

A drum containing radioactive sources is loaded on to the rotating/translating platform. In the TGS mode of operation, the source-to-detector distance and the collimator aperture are automatically set to either the "Near_Open_Wide" (NOW) or the "Far_Open_Narrow" (FON) settings based on the measured dose rates. "Open" indicates that no lead absorbers are introduced in front of the detector. In the beginning of the assay, the drum moves clear off the detector–transmission source line of sight. The transmission source is exposed to the detector and an unattenuated spectrum is collected. The unattenuated count rate in each peak is used to determine the transmission fraction at that gamma ray energy. The item (or waste drum) is scanned from its edge to center and back to the edge. The scans are performed in two passes for each vertical layer of the drum; one with the transmission source exposed, and another with the transmission source closed

and only acquiring emission spectra. As the drum rotates and translates across the field of view of the detector and the transmission source, full spectral data grabs or view grabs are taken by the detector and electronics. The detector and transmission source move in unison to the next vertical layer.

The drum was divided into 16 vertical layers for scanning purposes. The assay time was 3600 s which means each scan was performed for 112.5 s. In each scan 150 spectral grabs were performed, each lasting for 0.75 s. Counts in the transmission and emission gamma ray spectra are binned into pre-determined Regions of Interest (ROI), with an ROI around each peak of interest, and background ROIs on either side of a given peak. The data collected in the transmission scans is used to solve for the linear attenuation coefficients over a 10×10 voxel grid per drum layer. And the emission data grabs are used to solve for radionuclide distribution over a 10×10 voxel grid per layer. The TGS method uses a ray tracing code to determine attenuation path lengths through voxels for each view, and to determine the geometric efficiency. Image reconstruction was performed using well-known algorithms such as Non-negative Least Squares (NNLS), Algebraic Reconstruction Technique [5] (ART) and Expectation Maximization [6] (EM). The ART algorithm was used to fit the transmission image, and the EM algorithm was used to fit the emission image. The NNLS fit was used to provide the initial guess for the EM algorithm. The TGS technique uses the Material Basis Set (MBS) formalism [1] to solve for linear attenuation coefficient map for a given matrix. In the MBS formalism, the attenuation map is obtained in terms of a low Z and a high Z component.

The TGS assay result can be expressed as follows:

$$A = K \sum_{j=1}^{n} S_j. \quad (1)$$

In Eq. (1) A is the activity of a given nuclide, S_j is the geometry and attenuation compensated emission image at a gamma ray energy emitted the nuclide in voxel j. The sum of the elements of the emission matrix S is known as the "TGS number", and is a function of gamma ray energy. The quantity K is a calibration parameter obtained by assaying a source of known activity located in a drum with a representative matrix and is expressed in units of activity per TGS number. The variation of TGS number versus energy is similar to the shape of the intrinsic efficiency curve of the detector. This similarity can be exploited to extend the TGS calibration beyond the limits of measured data points, by pegging the calibration parameter using relative efficiencies derived from the intrinsic efficiency curve.

3.1. TGS calibration and verification results

The TGS system was calibrated by inserting six mixed gamma rod source standards containing the nuclides ^{133}Ba (185.87 μCi), ^{137}Cs (32.20 μCi) and ^{60}Co (30.91 μCi), in a 208 L drum containing softboard matrix (0.43 g cm^{-3}). The statistical uncertainty in the TGS results is estimated using a method known as Monte Carlo Randomization [7] (MCR). The MCR method involves randomization of the TGS view data using Poisson distribution and estimating the results for several replicate analyses. The calibration results for the "NOW" geometry are given in Table 1.

The calibration was verified by assaying drums with different matrix types and sources. Table 2 shows the TGS results for rod sources inserted into empty (no matrix) drum and a particle board matrix drum (0.72 g cm^{-3}).

Additional verification measurements were performed by distributing three-point sources of ^{137}Cs inside a softboard matrix drum. The results are given in Table 3.

The transmission and emission images for the three-point source assay are given in Fig. 3. In Fig. 3, the transmission images are shown on the left and the emission images are shown on the right. The images at the top show a cross-sectional view of a given layer, and the images at the bottom show a ray projection at a given angle of

Table 1
TGS (NOW) calibration for 208 L drum

Nuclide	Energy (keV)	Calibration (μCi/TGS♯)	Uncertainty (μCi)/TGS♯
^{133}Ba	276	162.0	6.3
	303	74.1	2.6
	356	24.3	0.8
	383	175.9	6.7
^{137}Cs	662	28.3	1.0
^{60}Co	1173	35.9	1.3
	1332	39.7	1.4

Table 2
Verification of TGS calibration using rod sources

Energy	Empty (no matrix)		0.72 g cm^{-3} matrix	
	Meas/true	Uncertainty	Meas/true	Uncertainty
276	1.0405	0.0425	0.9140	0.0554
303	1.0942	0.0396	0.9623	0.0460
356	1.0469	0.0337	0.9478	0.0358
383	1.0405	0.0420	0.9588	0.0514
662	1.0344	0.0401	0.9478	0.0402
1173	1.0407	0.0412	0.9834	0.0402
1332	1.0416	0.0406	0.9831	0.0406

Table 3
TGS Verification using three-point ^{137}Cs sources

Meas. activity (μCi)		True activity (μCi)		Meas/true	
Activity	Uncertainty	Activity	Uncertainty	Ratio	Uncertainty
187.5	6.56	178.40	5.35	1.051	0.048

rotation. The three regions where there is an activity concentration are seen in the emission image.

In Fig. 3, in the cross-sectional view of the transmission image, the three corner voxels on all four sides lie outside of the drum and are not considered during image re-construction.

4. SGS calibration and verification

The system was calibrated in the SGS mode using measurements and analytical calculations. The measurement-based calibrations were performed in the most sensitive SGS geometry (NOW). In the less sensitive geometries (FON, FAN, FBN, FCN) the calibrations were based on empty drum efficiencies and calculated correction factors for matrix and absorber attenuation.

For the NOW geometry, the nuclide activity can be written as follows:

$$A = \frac{\dot{C}}{\varepsilon_{\text{Empty\_NOW}}\, Y\gamma}\,\text{CF}_{\text{matrix}}. \tag{2}$$

In Eq. (2) A is the nuclide activity, $\varepsilon_{\text{Empty\_NOW}}$ is the empty container efficiency, Y_γ is the gamma ray yield and $\text{CF}_{\text{matrix}}$ is the correction factor for matrix attenuation. For the FON, FAN, FBN, FCN geometries, the relevant equation is given below.

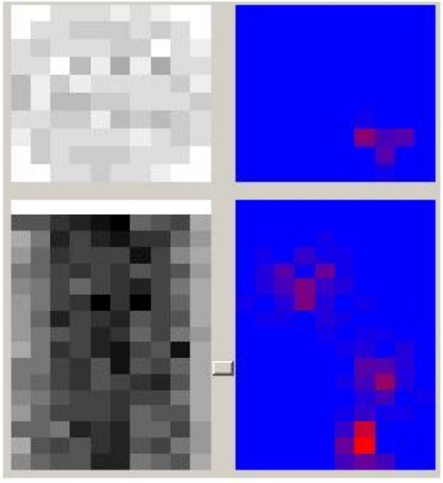

Fig. 3. ^{137}Cs point sources in softboard matrix.

The quantity $\text{CF}_{\text{matrix}}$ is empirically determined using the Parker formula [4]

$$\text{CF}_{\text{matrix}} = \frac{-\ln(T^\kappa)}{1 - T^\kappa}. \tag{3}$$

In Eq. (3), T is the measured (or calculated) gamma ray transmission and κ is a geometry-dependent factor. For cylindrical geometry, κ takes on an approximate value of about 0.83

$$A = \frac{\dot{C}}{\varepsilon_{\text{Empty\_FON}}\, Y_\gamma}\,\text{CF}_{\text{matrix}}\text{CF}_{\text{absorber}}. \tag{4}$$

For the FON geometry, the quantity $\text{CF}_{\text{absorber}}$ is unity since no lead absorber is present in front of the detector.

The empty drum efficiencies and matrix correction factors are determined on a segment-by-segment basis applied to the segment count rates.

In the NOW geometry, the system was also calibrated using the multi-curve method. The multi-curve calibration involved measuring peak efficiencies (using the rod source standards) a function of gamma ray energy over a range of uniform matrix densities. Efficiency calibration data for each matrix density is fit using a ln–ln polynomial function. The multi-curve is generated by merging the efficiency calibrations at multiple densities. Peak efficiencies at any gamma ray energy and any matrix density within the calibration range can be determined by interpolation.

The SGS calibrations were verified by assaying drums with uniform matrix densities and a uniform source distribution. The six rod source standards containing the nuclides ^{133}Ba (185.87 μCi), ^{137}Cs (32.20 μCi) and ^{60}Co (30.91 μCi) were used to create a uniform source distribution. The verification measurements at the factory could only be performed in the most sensitive NOW geometry because of lack of high-activity drums. The results are given in Table 4.

The uncertainties are quoted at 1σ and included counting statistics and the uncertainties in the source certificate. To test the FON, FAN, FBN and FCN geometries a real waste drum (208 L) was measured on site at a nuclear power plant where the system had been installed after factory calibration. The average density of the drum matrix was 0.72 g cm^{-3} and the surface dose rate was measured to be 180 μSv h^{-1}. The results obtained for the same drum from the different assay geometries were intercompared.

Table 4
SGS verification (NOW); measured/true activity ratio

Matrix g cm^{-3}	^{133}Ba result		^{137}Cs result		^{60}Co result	
	Ratio	Uncertainty	Ratio	Uncertainty	Ratio	Uncertainty
0.02	1.027	0.040	1.009	0.035	1.015	0.037
0.43	1.009	0.031	0.986	0.033	0.976	0.033
0.72	1.004	0.031	0.974	0.033	0.962	0.033
1.60	1.048	0.034	1.043	0.040	0.962	0.033
3.01	0.944	0.036	1.020	0.036	1.009	0.035

Table 5
SGS results for higher activity geometries

Geometry	^{137}Cs activity, μCi		^{60}Co activity, μCi	
	Activity	Uncertainty	Activity	Uncertainty
SGS_FON	226.6	4.8	2534	35.5
SGS_FAN	234.0	4.7	2576	30.9
SGS_FBN	252.5	6.1	2695	32.3
SGS_FCN	250.1	7.5	2708	32.5
Mean	240.8		2628	
Std. Dev.	10.9		74.9	

In Table 5, the standard deviation of the results is within 3%–5% of the average. But a count-rate-dependent bias is evident in the results for the FON and FAN geometries. The rate loss corrections for the FON and FAN measurements were 1.32 and 1.09, respectively. An under-correction for rate loss could have been the cause for the bias.

5. Conclusions

The performance characteristics of one of the four commercial grade tomographic scanning systems designed and built by Canberra Industries was discussed. The operation, calibration and verification of the system were described for both TGS and SGS modes. The TGS technique has been applied for the first time to assay radioactive waste generated by nuclear power plants. The TGS technique was applied to assay waste drums that were less than $1.0\,g\,cm^{-3}$ in density. The dynamic range of density and dose rate of the system were increased to $3.0\,g\,cm^{-3}$ and $0.1\ Sv\,h^{-1}$ by adding five SGS modes of operation. The limitation of TGS is primarily due to low counting statistics in the view data when matrix densities become high and the transmission gamma rays are not penetrating the matrix. Alternative approaches to the design such as a dual strength transmission source are being pursued. Alternative scanning patterns such as a triple scan (low-beam and high-beam transmission scans and an emission scan) and a hybrid transmission data structure are being studied. The preliminary results from these new approaches are encouraging in terms of extending the density range for TGS applications. Future work will include assay results quoted with total measurement uncertainty estimates.

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