AN INTEGRATED WASTE ASSAY SYSTEM USING TOMOGRAPHIC AND SEGMENTED GAMMA SCANNING FOR NUCLEAR POWER PLANT APPLICATIONS -10375

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ABSTRACT

An integrated system was built and tested for the non-destructive assay of drummed radioactive waste from nuclear power plants. The system is intended for assaying waste items of contact dose rates over a wide dynamic range with a maximum of 200 R/hr. The system is capable of assaying 208 liter drums (55 U.S. Gallons) and 320 liter (85 U.S. Gallons) over-pack drums in two different modes; a Tomographic Gamma Scanning (TGS) mode and a Segmented Gamma Scanning (SGS) mode. The assay geometry for a given item is configured by automatically adjusting a variable aperture collimator, the distance of the detector from the drum, and the usage of lead attenuators of three different thicknesses. The geometry adjustment is chosen based on the measured dose rate of the item.

A 15 milli-Curie Eu-152 transmission source is used in the system to determine matrix attenuation. Waste items with matrix densities between 0 and 1 g/cc were assayed in the TGS mode. TGS combines High Resolution Gamma Spectrometry (HRGS) with low spatial resolution image reconstruction to yield matrix attenuation compensated assay results. In a TGS assay, the item is scanned in 3-degrees of freedom; rotation, translation and elevation. Matrix attenuation and radionuclide concentration are obtained on a ~50 mm voxel by voxel basis. Therefore, the TGS technique is well suited for assaying non-uniform distributions of radionuclides in non-homogeneous matrices. The SGS mode can be used to increase throughput at low densities but also extends the density range to circa 3.0 g/cc by relying on simplified assumptions when the quality of the transmission data deteriorates.

One of the distinguishing features of the current system is that the TGS scan was done in a direction opposite to the scans performed by other Canberra systems in the field. The unique scan direction was necessitated because of site specific requirements for installing the system. The site installation also required data acquisition to be collected via Ethernet through the Canberra Lynx instead of the AccuspecB used on other Canberra systems. In this work, we discuss the performance characteristics of the TGS system and show results of image reconstructions of point sources placed in known voxel locations.

The efficiency calibration in the SGS mode was performed using Canberra's In Situ Object Calibration Software (ISOCS). ISOCS, which uses mathematical ray-tracing techniques to determine gamma ray full energy peak efficiencies. This paper presents also the system performance in the SGS mode.

INTRODUCTION

In this work we describe an integrated waste assay system designed to meet a need identified by VJ Technologies Inc. at the Korea Hydro & Nuclear Power (KHNP) Waste Receipt/Inspection Facility (WRIF). The integrated system uses Segmented Gamma Scanning (SGS) and Tomographic Gamma Scanning (TGS) to nondestructively assay items with matrix densities up to 3.0 g/cc and dose rates as

high as 0.15 Sv/hr. The SGS is designed for waste items whose contents are homogenously distributed. The TGS was developed for waste items containing arbitrary spatial distributions of gamma emitting radionuclides in heterogeneous matrices [1-4]. The intent was to improve the accuracy over traditional non-scanning and axial-scanning (non-imaging) methods. Improving the safety, accuracy and overall cost effectiveness of the processes and methods used to characterize and handle radioactive waste is an on-going mission for the nuclear industry. The integrated waste assay system described in this paper has the capability to assay a wide variety of radionuclides, such as those found in nuclear power plant waste as well as special nuclear material. Low-level radioactive waste is typically stored in drums that can contain a wide variety of materials; from personal protective equipment to contaminated shielding. The radioactive contents can vary; for example, actinides, or long-lived radioisotopes created from fission or activation processes. In order to properly dispose of such waste drums, or identify candidates for potential additional treatment, it is necessary to have an accurate assay of its radionuclide contents. An important contributor to this goal is the development of superior non-destructive assay instruments. The integrated waste assay system employing TGS is a case in point.

The TGS methodology combines low spatial resolution imaging techniques with High Resolution Gamma Spectroscopy (HRGS), which can yield quantitative results that are more accurate compared to nonimaging methods in situations where the radionuclide source distribution is non-uniform and the waste matrix is heterogeneous. 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-tomoderate matrix densities (0–1.0 g cm/cc) and for contact dose rates less than 45 mSv/hr. Among the limitations of the TGS technique are the statistical fluctuations in the transmission and emission data. On the 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 create one system capable of assaying items with matrix densities up to 3.0 g/cc and dose rates up to ~ 0.15 Sv/hr by incorporating the SGS measurement technique.

The SGS is a well-established NDA instrument that uses analytical techniques developed by Parker [5] 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 24mm (C) were introduced in front of the detector to prevent it from saturating.

The TGS, for the measurement of radioactive waste packages, may be thought of as an evolution from the familiar segmented gamma scanner (SGS) device with an infusion of ideas borrowed from the medical imaging community. In essence, the SGS performs a simple axial scan on a rotating item. The rotation is intended to even out the response to radial heterogeneities in the matrix and non-uniformity in the activity distribution. A collimator is used to gather emission data from each segment, or layer, of the scan, and an associated transmission measurement through the center of the rotating item is used to establish an average matrix correction factor for each layer. In the TGS, the rotator is replaced by a stepper motor table; an additional translational degree of motion is incorporated using a stepper motor-controlled slider mechanism. The collimator is replaced by a diamond-shaped collimator with a much narrower field of view. The simple axial scan is replaced by an axial scan which gathers spectral data grabs not only as rotational averages but also as a function of rotational and translational position. In this way, the pencil transmission beam is used to map out the linear attenuation profile of each layer and, together with the emission data image reconstruction techniques, are used to form an attenuation-corrected activity map from which the total activity is estimated.

In addition to the low spatial resolution imaging capability of the TGS, which is intended primarily to improve the overall quantitative assay result by improving the finesse of the matrix corrections, an

important practical benefit of the TGS approach is that the major mechanical movements have been placed under computer control. For general purpose machines this philosophy has been extended not only to include the item rotation and translation, but also the collimator geometry, filter configuration, transmission source intensity modulation and detector to item separation. In this way, a powerful all-purpose data acquisition platform has emerged.

DESCRIPTION OF THE SYSTEM

Mechanical Design

The integrated waste assay system described in this paper is distinguished by its capability to assay waste items using either TGS or SGS measurement techniques. A photograph of the system is shown in Fig. 1.



Figure 1. Photograph of the industrial TGS system during factory assembly. The transmission assembly is viewable on the left. To the right is the germanium detector and surrounding aperture box. The drum is shown on the in-feed conveyor.

The TGS mechanism is a modular design and consists of the following modules: detector vertical drive module, inline rotation/translation module, transmission vertical drive module and a transmission source shield with shutter. Automation of vertical, lateral and rotational motions, as well as the transmission source shutter and variable collimator, is controlled by a GE/Fanuc Process Logic Controller (PLC) and directed by Canberra's data acquisition and analysis software, NDA 2000. The system consisted of a 5000mm² Broad Energy Germanium (BEGe) detector and a 15mCi Eu-152 transmission source. The detector was housed inside a cylindrical lead shield of 50mm annular thickness. The detector and the

transmission source are accurately aligned with each other and with the center of the rotating platform. The pulse processing electronics consisted of a TRP (Transistor Reset Preamplifier), Canberra's third generation digital signal processor LYNX (which allowed data to be retrieved using Ethernet connection) and a Canberra Model 1654 Reference Pulser. Rate loss corrections for each grab were performed using the reference pulser counts. This system also includes a software-integrated weigh scale and a solid state dosimeter.

The collimator 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, 25.4mm thick, that can be opened and closed to change either the spatial resolution of the TGS measurement or to switch to an SGS measurement mode. Fig. 2a and b show examples of TGS and SGS collimators. In the TGS mode, two different collimator and source-to-detector distance combinations were possible; a "Near" geometry with collimator aperture of 50.8mm (Wide or W) and a "Far" geometry with collimator aperture of 40.6mm (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 is selected automatically based on drum weight and dose rate criteria.



Figure 2. Variable Aperture Collimator in the (a) Wide SGS and (b) Wide TGS positions.

The source of the transmission gamma rays is a highly collimated radioactive source that is located on the side opposing the germanium detector. With such a source, one can assay 208 liter and 320 liter (over-pack) waste drums in the TGS mode with densities up to about 1.0 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; hence the need of the SGS mode in such cases.

Software

VJ Technologies is providing a supervisory software program, which runs on top of Canberra's NDA 2000 software, to automate the selection of measurement technique and geometry based on the results of measurements from the scale and dosimeter components. This enables the integrated waste system to automatically set and perform any of the assays based on previously entered selection rules (based on the dose rate and weight of the drum).

By combining the TGS platform with this supervisory software, a batch of drums can be staged and measured according to type. High dose drums (as determined automatically from external dosimeters) may be assayed in a far geometry with a small collimator and filters in place to control the counting rate. On the other hand, light, fairly uniform, low activity drums might be assayed in a close geometry with a wide collimator to improve detection limits and throughput. Besides setting the geometry based on dose measurement, the weight of the drum combined with dose information will serve to set the mode of the assay TGS or SGS. All of these scenarios, and more, are available because the integrated system allows for all of the major mechanical parameters affecting the scanning protocol to be changed. Scan times and other variables, such as the collimator size, degree of filtration and transmission intensity can also be adjusted to better meet the need of the operator according to the particular waste characteristics.

TGS Assay

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 segment scan was performed for 112.5 s. In each scan, 150 spectral grabs were performed lasting 0.75 s. Counts in the transmission and emission gamma ray spectra are binned into predetermined Regions of Interest (ROI) with an ROI around each peak of interest, and background ROIs on either side of each peak ROI. The data collected in the transmission scans is used to solve for the linear attenuation coefficients over a 10x10 voxel grid per drum layer. And the emission data grabs are used to solve for radionuclide distribution over a 10x10 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 [6] (ART) and Expectation Maximization [7] (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 [8] 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.

TGS MODE CALIBRATION AND VERIFICATION

One of the distinguishing features of the current system is that the direction of the TGS scan protocol was configured in the direction opposite to that performed by other Canberra systems in the field. The unique scan direction was necessitated because of site-specific requirements for installing the system. Looking from the detector to the transmission source (which defines North), the drum is assayed from East to West in a counter-clock wise rotation (CCW). This configuration is often referred to as EAST and CCW scanning. Typically, Canberra TGS systems assay the drum from West to East in a clockwise rotation.

As discussed later in this section, the direction of the scan protocol does not affect the performance of the system.

The TGS assay result can be expressed as follows:

$$A = K \cdot \sum_{j=1}^{\# Voxels} S_j \qquad \qquad \text{Eq(1)}$$

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 by 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 is 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.

The TGS system was calibrated by inserting six mixed gamma rod source standards containing the nuclides Ba-133 (183.18 mCi), Cs-137 (30.85 mCi) and Co-60 (30.60mCi) in empty (No matrix) 208 L and a 320 L drums. The statistical uncertainty in the TGS results is estimated using a method known as Monte Carlo Randomization [9] (MCR). The MCR method involves randomization of the TGS view data using the 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 a "softboard" matrix (0.43 g/cc) drum and a particle board matrix drum (0.72 g/cc). Overall, the verification demonstrates good agreement; within 8% of the declared activity.

Nuclide	Peak Energy	Calibration	Uncertainty	
	[keV]	[µCi/TGS#]	[µCi/TGS#]	
Ba-133	276	140.7	2.6	
Ba-133	303	59.8	1.0	
Ba-133	356	20.5	0.3	
Ba-133	383	150.3	3.2	
Cs-137	662	24.4	0.4	
Co-60	1173	32.4	0.5	
Co-60	1332	35.4	0.7	

Table 1. TGS NOW Calibration Results for the 208 L Drum.

Table 2. Verification of the TGS Calibration Using Mixed-Nuclide Rod Sources.

Peak Energy	"Softboard" Matrix (0.43 g/cc)		Particle Board matrix (0.72 g/cc)		
[keV]	Measured/True	Uncertainty	Measured/True	Uncertainty	
276	1.0209	0.0227	1.0477	0.0457	
303	0.9997	0.0222	1.0243	0.0325	
356	1.0335	0.0146	1.0514	0.0225	
383	0.9689	0.0230	0.9841	0.0440	
662	1.0025	0.0223	0.9281	0.0244	
1173	0.9851	0.0177	1.0068	0.0320	
1332	1.0231	0.0271	1.0365	0.0252	

Table 3. TGS Verification using three-point Cs-137 sources

Additional verification measurements were performed by distributing three point sources of Cs-137 inside a softboard matrix drum. The results, shown in Table 3, present an excellent agreement between the measured and declared activity; within 1%. The transmission and emission images for the three point source assay are presented 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 transversal ray projection at a given angle of rotation. The three regions where the point sources are located are shown as activity concentrations 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.

Measured Activity (µCi)		True Activity (µCi)		Measured/True	
Activity	Uncertainty	Activity	Uncertainty	Ratio	Uncertainty
128.43	2.72	126.90	2.78	1.01	0.16





Figure 3. Cs-137 point sources in softboard matrix. (a) Horizontal section of the transmission image along layer #8. (b) Horizontal section of the emission image along layer #8. (c) Transversal section of the transmission image. (d) Transversal section of the emission image.

These verification results are comparable to other Canberra TGS systems. For example, the results presented by Dr. Venkataraman at the ESARDA Conference in 2005 [10] demonstrated agreement for Cs-137 and Co-60 in rod sources placed in the "Softboard" matrix to be within 3.6% and 3.3%, respectively. The three Cs-137 point source verification measurement agreed within 5% of the true activity.

SGS MODE CALIBRATION AND VERIFICATION

In addition to tomographic scanning, the system is capable of performing segmented gamma scan (SGS) assays. The SGS assay is performed with the transmission measurement to perform a layer-by-layer density correction. The SGS result can be determined following two methods. The first method (or transmission based) makes use of the transmission and measured empty drum efficiencies (in the NOW

and FON geometries) for both the 208 L and 320 L drums. The second method (or efficiency multicurves based) uses the computed multi-energy and density calibration curves (computed for the NOW, FON, FAN, FBN and FCN) to perform the efficiency correction based on the average density of the 208 L and 320 L drums. To this end the SGS mode is calibrated using both measurements and calculations.

In the first method (that uses the transmission results); the empty drum efficiency and transmission are measured in the NOW and FON geometries for both 208 L and 320 L empty (No matrix) drums. The

activity, calculated based on gamma line γ with measured count rate C, is given by Eq(2)

$$A = \frac{\overset{\bullet}{C}}{\underset{Empty_drum}{\overset{\bullet}{}} \cdot Y_{\gamma}} \cdot CF_{matrix} \qquad Eq(2)$$

In Eq(2), A is the nuclide activity, $\varepsilon_{Empty_drum}^{Geometry}$ is the measured empty container efficiency for the geometry (NOW or FON), Y_{γ} is the gamma ray yield and CF_{matrix} is the correction factor for matrix attenuation given by Eq(3) [5]. The empty drum efficiencies and matrix correction factors are determined on a segment-by-segment basis and applied to the segment count rates.

$$CF_{matrix} = \frac{-\ln(T^{\kappa})}{1 - T^{\kappa}}$$
 Eq(3)

Where T is the measured gamma ray transmission and k is a geometry-dependent factor. For cylindrical geometry, κ takes on an approximate value of about 0.83.

For extremely dense drums in which the 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 second method (that uses efficiency multi-curves) is calibrated by measuring the empty, "softboard" (0.43 g/cc), particle board (0.72 g/cc), sand (1.5 g/cc) and concrete (3 g/cc) matrices in the NOW geometry for the 208 L drum and extracting the matrix correction factor CF_{matrix} . The matrix correction factors were computed by assaying the 208 L drum with uniform matrix densities and a uniform source distribution. The six rod source standards containing the nuclides Ba-133 (183.18 mCi), Cs-137 (30.85 mCi) and Co-60 (30.60mCi) were used to create a uniform source distribution. For the energy of interest, the matrix correction factor is the ratio of the matrix drum efficiency to the empty drum efficiency.

The computed matrix correction factors were used for the other 320 L drum and FON, FAN, FBN and FCN geometries. It should be noted that for all the considered geometries and drum types, an emptydrum efficiency drum is required as indicated in Eq(2). This was estimated using Canberra Industries' Insitu Object Counting System (ISOCS) software [11] for all geometries for both the 208L and 320L drums. With this software, it is possible to quickly compute the SGS efficiency over a wide range of energies, densities, and drum configurations.

Canberra developed in 1997 the In-Situ Object Calibration Software (ISOCS[™], US Patent 6228664) [12, 13]. This mathematical efficiency calibration software uses a combination of Monte Carlo, numerical integration, and ray-tracing methods. First an accurate MCNP (Monte Carlo N-Particle) model of the

radiation detector (Ge, NaI, LaBr, ...) is created based upon our best knowledge of the detector construction. This model is then validated by comparisons to a series of carefully controlled comparisons to NIST-traceable reference point sources. Where necessary the MCNP model parameters are adjusted so that the measured efficiency matches the MCNP modeled efficiency at ~10 energies and multiple spatial locations (close and far, front side and back). This validated MCNP detector model is then used to compute the efficiency at about 800 different locations at multiple distances close (0.01mm) and far (500m), and many locations from front (on-axis) to rear (behind detector). These point-efficiencies are then integrated into a spatial map and supplied as a detector characterization file with that specific detector. The ISOCS software uses this file, along with the information entered by the user to define the shape of his measurement object (21 different basic shapes are available) and the dimensions of the many parameters that can be used to define the shape to best represent the item to be measured. The ISOCS computation software then performs volume integration and matrix attenuation calculations to correct for sample self-attenuation, container attenuation, air attenuation, and collimator attenuation, to determine the energy-vs.-efficiency calibration curve for energies in the 10-7000 keV range. This software has been validated [14] against 109 different multi-energy large volume reference sources and shown to be accurate within 4.5% relative standard deviation (>150 keV) and 7-9% (150 keV).

The SGS calibrations were verified by assaying drums with uniform matrix densities and a uniform source distribution using the six rod sources. The verification measurements at the factory were performed in the most sensitive NOW geometry because the calibration sources could not provide enough statistics for reliable verification in the other geometries. The results are given in Table 4, and, overall, are within 20% of the true activity. The uncertainties are quoted at 1σ and included counting statistics and the uncertainties in the source certificate.

Matrix Density	Ba-133 Results		Cs-137 Results		Co-60 Results	
[g/cc]	Ratio	Uncertainty	Ratio	Uncertainty	Ratio	Uncertainty
0.2	1.04	0.057	0.98	0.072	1.00	0.037
0.43	0.97	0.062	0.93	0.079	0.98	0.030
0.72	0.95	0.071	0.90	0.092	0.94	0.027
1.50	0.98	0.080	0.93	0.111	0.96	0.034
3.01	1.18	0.099	1.11	0.138	1.12	0.046

Table 4. SGS Verification (NOW geometry and 208 L drum) Results Presented as the Ratio of Measured to True Activity.

CONCLUSIONS

A multi-modal gamma-ray 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 performance characteristics of a commercial-grade Tomographic Gamma Scanning system designed and built by Canberra Industries were discussed. The operation, calibration and verification of the system were described for both TGS and SGS modes. In this application, the TGS technique has been applied to assay radioactive waste generated by nuclear power plants. One of the distinguishing features of the current system is that the TGS scan protocol is performed in a direction opposite to that performed by other Canberra systems in the field. The TGS technique was applied to assay waste drums that were less than 1.0 g/cc in density. The dynamic range of density and dose rate of the system were increased to 3.0 g/cc and 0.15 Sv/hr 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.

An alternative approach to the traditional TGS design discussed in this paper is to employ a dual-intensity transmission source as developed by Canberra [15]. With the new extended range TGS system, it is now possible to assay a wider range of waste drums with a single system than was previously available.

An obvious extension of the work presented here is to perform an in-depth study and the quantification of the total measurement uncertainty estimates once the system is deployed onsite.

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