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NONDESTRUCTIVE ASSAY SYSTEM

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# FIELD EXPERIENCE WITH A MOBILE TOMOGRAPHIC NONDESTRUCTIVE ASSAY SYSTEM

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## ABSTRACT

A mobile tomographic gamma-ray scanner (TGS) developed by Los Alamos National Laboratory was recently demonstrated at the Rocky Flats Environmental Technology Site and is currently in use at Los Alamos waste storage areas. The scanner was developed to assay radionuclides in low-level, transuranic, and mixed waste in containers ranging in size from 2 ft<sup>3</sup> boxes to 83-gallon overpacks. The tomographic imaging capability provides a complete correction for source distribution and matrix attenuation effects, enabling accurate assays of Pu-239 and other gamma-ray emitting isotopes. In addition, the system can reliably detect self-absorbing material such as plutonium metal shot, and can correct for bias caused by self-absorption. The system can be quickly configured to execute far-field scans, segmented gamma-ray scans, and a host of intermediate scanning protocols, enabling higher throughput (up to 20 drums per 8-hour shift).

In this paper, we will report on the results of field trials of the mobile system at Rocky Flats and Los Alamos. Assay accuracy is confirmed for cases in which TGS assays can be compared with assays (e.g. with calorimetry) of individual packages within the drums. The mobile tomographic technology is expected to considerably reduce characterization costs at DOE production and environmental technology sites.

## INTRODUCTION

On March 24, 1995, the Los Alamos mobile Tomographic Gamma Scanner (TGS) arrived at the Rocky Flats Environmental Technology Site. During the following week, the system was moved inside the protected area and was used to assay 12 drums containing residues. Six different types of residues that were originally measured by a variety of package- and drum-size nondestructive assay systems were represented. The accuracy of TGS was confirmed for cases

in which TGS assays could be compared with declared inventory values that were established by assaying individual items contained within the drum (e.g. by calorimetry, gamma-ray scanning, or neutron counting of small samples). Low-resolution tomographic emission and transmission images of each drum were reconstructed. Real-time radiography and TGS images of one of the drums were compared, showing that individual packages can be resolved by TGS.

A follow-on exercise at Los Alamos was conducted to verify the calibration of the scanner and to continue the TGS demonstration by assaying mock residue drums constructed using available matrix materials and plutonium standards. A variety of plutonium standards and matrix materials were used to explore the performance of the technique for a variety of matrix types and plutonium inventories. Several categories of residue drums were simulated that would be difficult to assay accurately using conventional gamma-ray scanning. Categories included pyrochemical salts, wet combustibles, Raschig rings, and scrap metal.

The purpose of the Rocky Flats demonstration was to provide information needed to evaluate the use of mobile TGS technology in the residue stabilization process. The mobile capability represents a potential cost savings in that drums would no longer have to be transported to a fixed NDA facility. Tomographic gamma scanning enables a wide variety of drums to be assayed with an accuracy exceeding that of conventional gamma-ray assay techniques. In this paper, we summarize the work conducted during the demonstration and report on the performance of TGS for assaying residues.

## SYSTEM DESCRIPTION

The mobile system used in the demonstration contains a gamma scanner that is capable of scanning samples with sizes ranging from 2-ft<sup>3</sup> boxes to 83-

gallon overpacks. Emission computerized tomography, with a complete three-dimensional attenuation correction, is used to determine the distribution of gamma-ray emitting radionuclides within the sample. Tomographic gamma scanning enables accurate gamma-ray assays of samples containing nonuniform matrix and emission distributions. Details of the TGS technique, including experimental validation of the method, are described elsewhere;<sup>1-5</sup> however, a summary of the primary features of the technique is provided here. The system also supports traditional scanning techniques such as segmented gamma scanning. All of the drums assayed at Rocky Flats during the demonstration were assayed using TGS.

### Tomographic Gamma Scanning

TGS was developed by Los Alamos with funding from the Department of Energy's Office of Safeguards and Security (Fig. 1). The technique uses low-resolution transmission and emission tomography to minimize bias due to nonuniformity in the matrix and in the distribution of gamma-ray emitting material (for example, Pu-239 and U-235). High-resolution gamma-ray spectroscopy (HRGS) is used to identify radionuclides within the drum and to provide accurate measurements of gamma-ray intensities. Transmission tomography is used to determine three-dimensional images of attenuating material within the sample (the matrix). Emission computerized tomography, corrected for attenuation by the matrix, is used to determine the distribution of emitting material within the sample. Because nonuniform matrix attenuation and source position effects are accounted for, TGS is accurate over a wide density range.

TGS is capable of high-throughput assays. A scanning protocol, in which the sample is rotated and translated relative to the source and detector without stopping, is used to minimize scan times. With this continuous

scanning protocol, a 208-L drum can be scanned in approximately 1 h with acceptable accuracy.

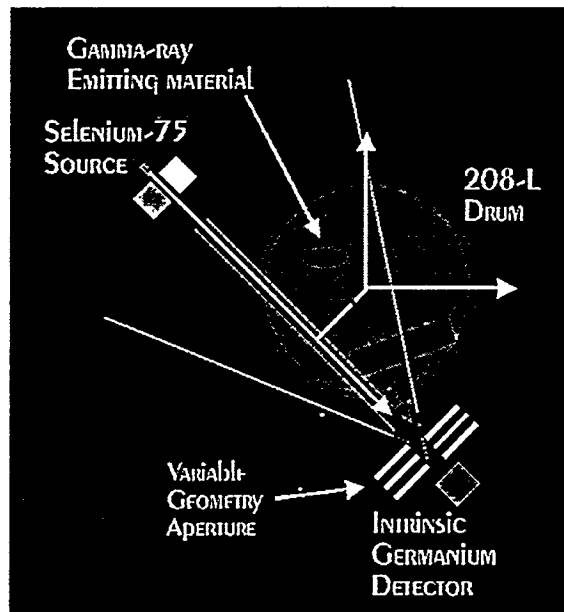


Fig. 1. TGS concept.

Because TGS scan times can be selected by the operator, longer assay times are possible when warranted. In addition, by combining information from segmented gamma scanning, drum-weight, and real-time radiography, overall system throughput can be increased. A preset time of 1 h was used for the demonstration.

### Trailer System Description

A diagram of the mobile segmented/tomographic gamma scanner (S/TGS) is shown in Fig. 2. The scanner is contained in a well-engineered trailer that is roughly 24 ft in length (not including the hitch) and 8 ft wide. The trailer has a goose-neck hitch and is designed to be pulled by a 1-ton pickup truck.

The trailer requires a 36 kVA power source, and can operate using a portable diesel generator or with line power. Currently, the power must be supplied as three-phase 208V, 100 amps. We will shortly upgrade the system to work with 480V. During the demonstration we used a diesel generator to eliminate the need to re-route site power.

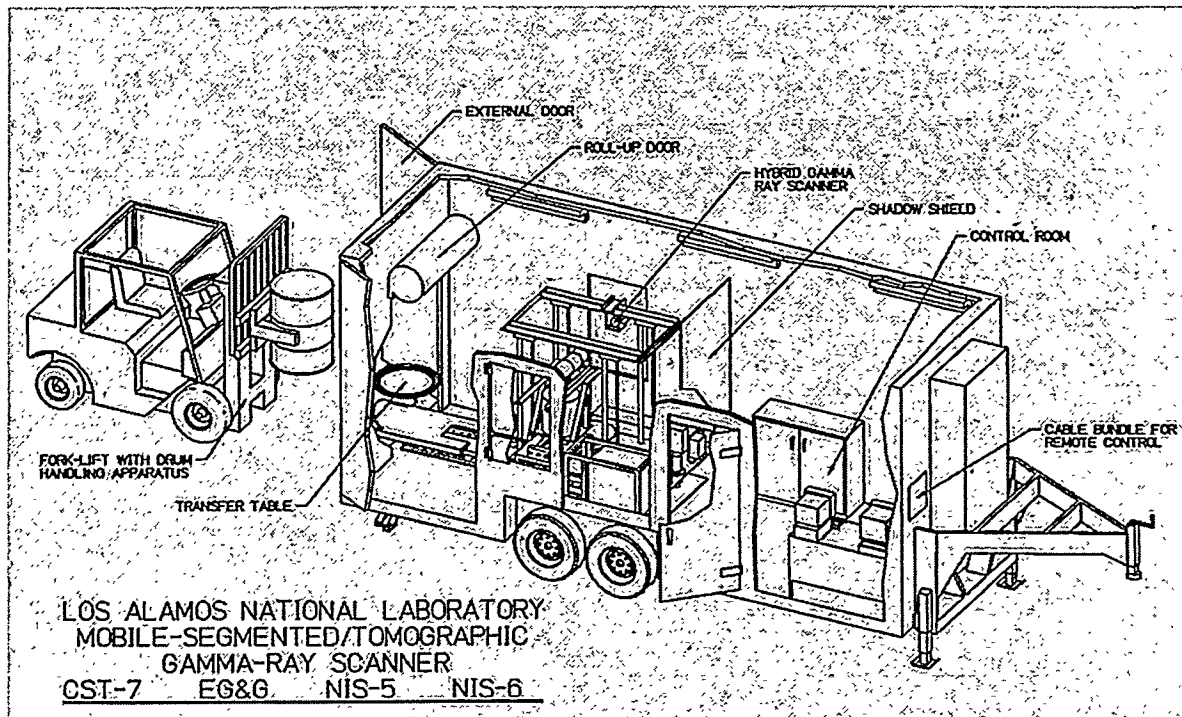


Fig. 2. A diagram of the mobile tomographic gamma scanner.

Power is routed to various subsystems within the trailer, including the scanner, interior lighting, instrumentation and electronics, and the heating, ventilation and air-conditioning (HVAC) system. Exterior lighting and power are provided as well as a "Christmas-tree" to indicate the status of the scanner (for example, scanning versus idle). Critical subsystems, such as the control and acquisition computer and the scanner are connected to an uninterruptible power supply (UPS) to enable safe shutdown of the scanner if external power is lost and to eliminate damage to sensitive electronics when power surges occur.

The interior of the trailer is divided into two parts. A control room is located in the front portion of the trailer and is accessed by a personnel door. The scanner and drum loading system is housed in the aft portion of the trailer. A wall containing a shadow-shield separates the control room from the scanner. The shadow shield consists of 1/4 in. lead and minimizes the dose to operators from low-energy gamma rays (for example, 60-keV line from Am-241). During operations at the Los Alamos transuranic waste site and at Rocky Flats, the dose to operators for contact-handled drums was negligible. Some of the drums at Los Alamos had contact dose rates approaching 100 mR/hour. For drums that must be remotely handled or that may result in an unacceptable dose to the operators, the scanner can be operated by remote control. A cable-bundle that can be attached to an exterior panel provides an interface that allows the operator to control and monitor (via video) the scanner and drum loading system from a remote location.

The gamma-ray instrument included in the trailer is capable of executing a variety of scanning protocols, including segmented gamma-ray scanning and tomographic gamma-scanning. The instrument was developed to assay waste in boxes, 55-gal. drums, and 83-gal. overpacks with a maximum throughput of 18-20 drums per 8-h shift. The instrument consists of a sample positioning system, a transmission source assembly, and a high-resolution gamma-ray spectroscopy system.

The sample positioning system allows reproducible placement of the sample with three degrees of freedom. The sample can be rotated, translated and elevated relative to the detector and source, enabling the system to acquire data for three-dimensional transmission and emission imaging.

The source assembly contains a gamma-ray source such as Se-75 that is collimated with a solenoid-driven shutter that, when opened, allows a narrow



beam of gamma rays to pass through the sample and into the detector. Three-dimensional images of the sample's linear attenuation coefficient are reconstructed from selected gamma-ray transmissions (for example, the 136-, 285-, and 401-keV lines of Se-75) measured using a tomographic scanning protocol. The images are used to correct for the attenuation of gamma rays emitted from the sample. Se-75 has gamma rays that are well-matched in energy to gamma rays emitted by Pu-239. To meet measurement throughput requirements, Se-75 activities ranging from 30 to 200 mCi are required.

The high-resolution gamma-ray spectroscopy system measures gamma rays emitted from and transmitted through the sample. This system consists of a collimated intrinsic germanium detector and associated spectroscopy electronics. The collimator shape depends on the type of scan. For example, segmented gamma scanning requires a rectangular collimator. A diamond-shaped collimator is used for tomographic gamma scanning. Currently, tungsten inserts are used to change the shape of the collimator. A variable geometry detector aperture will be installed this fiscal year to allow automated changes in collimator shape and variable-resolution tomographic scanning. This feature will enable the system to rapidly change scanning protocols to improve accuracy and maximize throughput.

An automated drum-loading system is provided with the trailer. This system consists of a number of components that are controlled by a computer through a user-friendly Microsoft-Windows-based interface. Samples (boxes, drums, or overpacks) are loaded onto a transfer table from a forklift or crane through a roll-up door. The transfer table is mounted on a linear slide that enables the table to shuttle between a load position outside of the trailer envelope (Fig. 3) and the TGS. The height of the transfer table was selected to prevent the drum from being lifted above a drop-limit specified by the Department of Transportation.



*Fig. 3. Transfer table extended beyond the roll-up door in the load position.*

A unique mechanism to move the drum from the transfer table to the scanner was devised. The drum sits on a removable plate that rides on the transfer table. To deliver the drum to the scanner, the transfer table is positioned over the TGS table and is lifted off the transfer table by elevating the TGS scan table. Once the transfer table (now without the plate and drum) is withdrawn to an idle position, the scan can commence.

## SOFTWARE

The TGS uses a modular approach to software, with separate executable modules for data acquisition, data analysis, and image display. In the Rocky Flats demonstration, data acquisition and scanner control were performed using a Compaq Deskpro 560 PC clone computer (60-MHz Pentium microprocessor), while analysis and image rendering were performed on a Sun workstation (Sparcstation 10). Both the workstation and PC were on a common network.

### Data Acquisition and Control Software

A "beta" test version of the LANL-developed program WIN\_TGS was used for data acquisition and scanner control.<sup>6</sup> WIN\_TGS is a Microsoft Windows 3.1 application written in C/C++. It offers a selection of built-in scan protocols (various TGS and SGS scans) as well as the capability to execute arbitrary user-defined scan protocols. WIN\_TGS currently supports Ortec multichannel analyzers (models 917 through 921, and 92X) and Compumotor stepper motor controllers (X- and 4000-series).

The fundamental data collected during a TGS assay are region-of-interest (ROI) sums extracted from the gamma-ray spectrum. An ROI editor allows three ROIs to be set for each peak of interest (one peak ROI and two background ROIs). The ROIs that are set prior to the scan determine the radionuclides that will be assayed during that scan. The TGS scans in this demonstration acquired 150 "data grabs" for each pass on each of 16 layers of the drum, for a total of 2400 sums for every ROI defined. In addition to the ROI sums, WIN\_TGS offers the option of saving cumulative gamma-ray spectra by layer and for the entire scan. These cumulative spectra can be used for isotopic analysis and for diagnostic purposes.

## Analysis and Rendering Software

Data acquired by the WIN\_TGS software are analyzed using TCNDA. TCNDA is a modular code system that was developed by Los Alamos to model and analyze gamma-ray measurements from general assay systems. Most of the modules included in TCNDA were written in standard ANSI Fortran 77 and have been tested on a variety of platforms, including unix workstations (IBM RS6000, Multiprocessor Sparcstation 10), I860 array processors, and PC's supporting Lahey Fortran. Portions of the code have been adapted to exploit the vector processing capabilities of the I860 and parallel architectures, including workstation clusters.

The central analysis capabilities provided by TCNDA are as follows:

- generalized geometry gamma-ray transport in NDA and CT systems
- special image models for low-resolution tomography
- an accurate model of the spatial response of arbitrary detector and collimator configurations
- variable-resolution transmission reconstructions for high-opacity samples
- emission reconstruction algorithms that are optimized for high-resolution gamma-ray spectroscopy
- precision estimation and error-bounding algorithms for transmission and emission computerized tomography<sup>7</sup>
- algorithms to correct for self-attenuation (for example, by lumps of plutonium metal)
- 2D and 3D image rendering and animation

## Future Software Capabilities

The software for the TGS is still in development, with the first fully integrated version scheduled for completion in December 1995. The completed software package will integrate the data acquisition performed on the PC with the data analysis and image rendering steps performed on a workstation, which for this demonstration were done as separate steps. In addition, isotopic analysis capabilities will be integrated into the analysis stream. Note that the analysis methods in the completed software will be the same as those used for this demonstration; the completed version will be integrated within an operator-friendly interface.

Additional software development work will continue throughout FY 1996, primarily on the analysis and rendering capabilities. A focus of this activity will be on increasing the throughput and accuracy of TGS by integrating data from a variety of sources (for example, digital and real-time radiography, neutron assay, weight measurements, and techniques to bound source location). Improved diagnostic techniques to flag difficult assay conditions will also be developed.

## ASSAY OF RESIDUE DRUMS AT ROCKY FLATS

The residue drums that were assayed at Rocky Flats are described in Table 1. The drums contained a range of materials, including electrowinning salts, Raschig rings, resins, and Leco crucibles. Two of the electrowinning salts (D52934 and D53918) had been characterized using calorimetry and were intended for use as working standards. The drums containing Leco crucibles had recently been repackaged, and it is not known how the plutonium mass were assigned. The last drum assayed (D74309) contained three packages of tantalum filaments that were previously assayed using calorimetry. This drum had been

examined by Los Alamos using real-time radiography. All of the drums were assumed to contain weapons grade plutonium.

All of the drums were assayed using the same scanning protocol (two-pass TGS) with a fixed scan time of 1h. For both emission and transmission scans, a preset count time of 30 minutes was used. Before each drum was scanned, we moved the drum to a predetermined position and acquired a spectrum to verify the presence of Pu-239 and to see if any interfering lines were present (none were identified). The time between the receipt of a drum on the transfer table to the delivery of the drum to the forktruck after the assay was complete did not exceed 1 hour and 15 minutes. Availability of the system was essentially 100%. Throughput was limited by site-related issues (for example, availability of personnel, and the site-wide drill). Despite the limitations, the authors feel that Rocky Flats personnel did an exceptional job coordinating the demonstration and delivering drums to the trailer under conditions that were apparently adverse.

Table 1. List of information provided on the residue drums.

TGS ID	Rocky Flats Drum ID	Item Description Code (IDC) Title	IDC Number	Drum net Mass (kg)	Declared Mass of Pu (g)	Past Assay Technique
RF01	D52934	Electrorefining salt	411	15	312	Calorimetry with gamma-ray isotopics
RF02	D53918	Electrorefining salt	411	10.5	680	Calorimetry with gamma-ray isotopics
RF03	D32160	Unleached resin	430	1	12	Segmented Gamma Scanner
RF04	D51181	Leached resin	431	24.6	21	CNCT (can-counter)
RF05	D50560	Electrorefining salt	411	14.3	199	SGS can-sized counter
RF06	D84824	Leco crucibles	370	111	94.5	Repacked and awaiting calorimetric assay
RF07	D84350	Leco crucibles	370	Unknown	30.5	Repacked and awaiting calorimetric assay
RF08	D84833	Leco crucibles	370	Unknown	20	Repacked and awaiting calorimetric assay
RF09	D83469	Leco crucibles	370	120	77.5	Repacked and awaiting calorimetric assay
RF10	D54164	Raschig rings, solvent contaminated	443	82	182	Segmented Gamma Scanner
RF11	D63251	Leached resin	431	1.8	114	SGS can-sized counter
RF12	D74309	Tantalum filaments	320	Unknown	239	Calorimetry with gamma-ray isotopics

## ASSAY OF MOCK RESIDUE DRUMS AND TGS CALIBRATION AT LOS ALAMOS

A follow-on exercise was carried out at Los Alamos to verify the calibration of the scanner and to continue the TGS demonstration by assaying mock residue drums constructed using available matrix materials and plutonium standards. Ron Harlan, EG&G Rocky Flats, visited Los Alamos to help construct the mock residue drums and participate in the demonstration. We attempted to simulate several categories of residue drums, including Raschig rings, scrap metal, MSE/ER salts, and wet combustibles.

While matrix materials were easy to come by, plutonium standards that are representative of Rocky Flats residues were not available. SGS standards that we used for calibration and a number of self-attenuating standards were used as surrogates.

### Plutonium Standards

Three sets of plutonium standards were used in the construction of the mock residue drums and for calibration: SGS can standards, plutonium oxide vials, and plutonium metal disks.

The SGS standards were cans approximately 12 in. long and 4 in. in diameter that contained plutonium powder mixed with diatomaceous earth. The particle size of the plutonium powder was small, resulting in negligible self-attenuation of Pu-239 gamma rays. The plutonium content of the standards was well-characterized, having been measured during preparation using a mass-balance and after preparation using calorimetry and gamma-ray assay. A table describing the characteristics of the standards follows.



SGS plutonium can standards (96.32 w% Pu-239)

Standard ID	Approx. Pu-239 (grams)	Exact Pu-239 (grams)	Uncertainty (grams)
STDSGA10	10	9.624	0.012
STDSGA30	30	28.874	0.023
STDSGMC-1	50	48.142	0.035
STDSGA100	100	96.269	0.068

A set of "2-gram" standards consisting of six vials containing plutonium oxide was also available to simulate the effect of plutonium self-absorption. Each vial was approximately 2.75 in. long by 0.5 in. in diameter. The total Pu-239 content in the vials was 9.76 g. The average escape fraction for 414-keV gamma rays for the "2-gram" standards was approximately 0.7.<sup>3</sup>

In addition, two disks of weapons grade plutonium metal were available to simulate self-absorption. The characteristics of the disks are listed in the following table.

Plutonium metal disks (Weapons grade)

	16-gram disk	41-gram disk
Mass of Pu-239 (grams)	16	41
Diameter	2 inches	2 inches
Thickness	~.05 mm	~1 mm
Escape fraction at 414 keV	~0.9	~0.7
Effective mass at 414 keV	14 grams	28 grams

Mock Waste Drums

Calibration. 208-L drums used for calibration were constructed using SGS can standards and low-density foam. The Pu-239 mass was varied from 10 g to 190 g.

Raschig rings. A drum was loaded with 141 kg net weight of Raschig rings, which occupied about 75% of the drum volume. The rings provided a low-Z matrix with an average density of about 1 g/cm<sup>3</sup>, which is a challenging matrix for the gamma-ray assay of drums. SGS standards were positioned within the matrix along the drum center-line.

Scrap metal. A 208-L drum was filled with aluminum scrap metal, consisting of small scrap on and near the bottom and various bars, slabs, structural channel and one notable cube, about 13 cm on edge. In order to simulate RFETS metal scrap, which frequently contains iron pipe, and copper pipe, and wires, a box of iron pipe was added to the drum. The 10-g and 50-g standard were surrounded by the pipe segments and were placed along the center-line of the drum within the aluminum scrap matrix. The experiments with this drum were considered a stringent test of the TGS method not only because of the drum density, but because of the drum's heterogeneous nature and the self-absorbing disks that were added. A second set of experiments was conducted without the steel pipe present.

Salts. Paint-cans filled with sodium chloride (NaCl) were prepared with commercial rock salt in an attempt to model the various salts that are found in waste at RFETS and at other DOE facilities. The electrowinning (ER) salts and molten salt extraction (MSE) salts produced in plutonium processing facilities can contain significant amounts of potassium and magnesium chlorides (KCl and MgCl<sub>2</sub>). In addition, the ratio of americium to plutonium found in these samples is often large, resulting in interferences between some Am-241 and Pu-239 lines. Because salt-bearing residues often contain plutonium metal shot, gamma-ray assays of ER and MSE salts are often biased.

To model the effect of plutonium metal shot, self-absorbing standards were included in some of the cans containing rock-salt. The cans were placed in the bottom of the drum surrounding SGS standards. The SGS standards were

included to model the effect of low-concentration plutonium on the energy dependence of the assay results.

**Wet combustibles.** The last matrix prepared was newspaper soaked in water. The net weight of the drum was 60 kg which is within in the range of wet combustible loadings at RFETS. Standards were placed in various locations within the matrix.

## RESULTS AND DISCUSSION

TGS assay results for the 12 residue drums are shown in Table 2. The mass of Pu-239 in grams determined using the 414-keV gamma ray is reported. The mass of Pu-239 was also calculated using the 129-, 203-, and 345-keV gamma rays. Ordinarily, the mass of Pu-239 is constant as a function of gamma-ray energy. However, when self-attenuating material, such as plutonium metal shot, is present, a distinct trend can be observed in the assay results and the reported mass value is usually biased low. In this case, Pu-239 mass increases with gamma-ray energy. Because lumps produce an energy variation that can be detected, the TGS analysis code can indicate when lumps are present. In addition, it is usually possible to correct the 414-keV mass for self-attenuation effects.

Note that several drums have been identified as having lumps. Two of the drums containing electrorefining salts (RF01, RF02) were found to have noticeable self-attenuation, probably due to the presence of plutonium metal shot, and a correction for

Table 2. Comparison between lump-corrected TGS and declared Pu-239 mass values for the residue drums. RF01, RF02, and RF12 are the *only* drums with credible declared mass values (see text).

TGS ID	Rocky Flats Drum ID	Declared Pu-239 mass (grams)	TGS 414 keV assay, Pu-239 mass (grams)	Were lumps detected?	Correction factor for lumps	Lump- corrected Pu-239 mass (grams)	Relative difference [%]	Effective lump diameter (cm)	Percentage of mass in lumps
RF01	D52934	291.6	204	Yes	1.47	300	3	0.47	60
RF02	D53918	647.6	492	Yes	1.31	645	-0.5	0.25	65
RF03	D32160	11.2	12.4	No	1	12.4	11	0	0
RF04	D51181	19.6	11.3	No	1	11.3	-42	0	0
RF05	D50560	186.0	154	Suspect	1	153.5	-17	0.	0
RF06	D84824	87.8	81.7	Yes	1.17	95.6	8.9	0.23	42
RF07	D84350	28.0	21.4	No	1	21.4	-24	0	0
RF08	D84833	18.7	14.0	No	1	14.0	-25	0	0
RF09	D83469	72.4	43.2	No	1	43.2	-40	0	0
RF10	D54164	170.1	153	No	1	154	-9.5	0	0
RF11	D63251	106.5	111	No	1	112.0	5	0	0
RF12	D74309	223.3	96.9	Yes	1.97	191	-15	0.59	82

self-attenuation was made. Correction factors that were multiplied by the 414-keV result to obtain the lump-corrected value are listed. The other electrowinning salt drum (RF05) is suspected to contain lumps, but no correction was made due to incomplete or inconsistent data. Only one of the Leco crucible drums was found to contain lumps (RF06). The drum containing tantalum filaments had the greatest energy variation, resulting in a large correction.

The lump-correction algorithm used with TGS divides the plutonium mass into two categories: a lumped mass that causes the observed variation with assay energy; and a dilute mass that produces no energy variation. The algorithm is also able to determine the characteristic size of the lumped plutonium. These values are reported in Table 2 when a lump correction was made. Notice that drum RF12 contains the largest lumps and has the greatest portion of mass in lumped material. Because the resolution of the TGS is only 2 in., self-absorption artifacts can be caused by a number of sources other than metallic plutonium. Partial volume effects resulting from the imperfect representation of the sample by discrete volumes can produce an observable energy variation for drums containing dense objects. The energy-variation introduced by these effects is usually small, but can in some cases obscure the energy variation caused by lumps. The lump-correction algorithm assumes that self-absorption is caused by metallic plutonium.

Lump-corrected TGS assays were in agreement with declared inventory values of drums containing packages that were assayed using calorimetry. TGS assays of the two drums identified as working standards (RF01 and RF02) matched the calorimetry values to within a few percent. The TGS result for the tantalum-filament drum differed from the calorimetry value by -15%. The calorimetry values are presumed to be quite accurate, perhaps to within 2%.

Results of TGS assays of drums assayed by Rocky Flats using package-sized segmented gamma scanning were comparable with declared inventory values (RF03, RF05, RF10, RF11). The greatest difference observed in these cases was for RF05, which contained electrefining salt and was suspected to have lumps. Larger differences were observed between TGS assays of drums containing Leco crucibles and their inventory values. It is not known how the present inventory values were assigned for these drums.

The results of TGS assays of mock drums assembled at Los Alamos are shown in Table 3. A number of drums contained low-density matrix material and were used to establish calibration constants for each gamma-ray energy. Note that over the entire range of Pu-239 mass loadings (10 g to 190 g), the TGS assay values for these drums differed by less than 3%. Matrix drums included Raschig rings, metal scrap, wet combustibles, and salt as discussed previously. The Raschig rings and metal scrap represent particularly challenging matrices for gamma-ray assay. Nevertheless, the maximum bias observed for TGS assays of these drums, when lumps were not present, was less than 20% and was less than 30% when lumps were present.

We analyzed the TGS data obtained for the scrap metal drum to determine how segmented gamma-ray scanning would have performed. In the cases we examined, SGS was biased low by at least a factor of five. The analysis did not include factors that would in reality have made the SGS assays far worse (for example, SGS transmissions are always through the center of the drum).

For the combustibles and salt matrices, TGS assays were found to be quite accurate, better than 10% when lumps were not present. The maximum bias observed for the 414-keV assay result when lumps were present was 25%. In these cases, lumps were detected and a correction was applied. Lump-corrected results were higher than the known mass, in the three cases examined. With the

short count-times used in the demonstration, self-attenuation effects were obscured for the difficult matrices (Raschig rings and scrap metal). In these cases, longer counting times are required to resolve the lump-effects. The mass of Pu-239 determined by TGS is compared to the declared or known mass for both the Rocky Flats and Los Alamos demonstrations in Figs. 4 and 5.

Images of selected residue drums are shown in Figs. 6, and 7. In each case, side-view radiographs of the contents of the drum derived from TGS images are shown (both for the distribution of Pu-239 and the attenuating matrix). Selected layers of the drums are also displayed. The volume elements in the reconstructed images are approximately 2-in. wide.

The images show that the contents of residue drums are far from uniform and homogeneous. The plutonium is contained within packages, which in many cases have been carefully loaded into the drums. Inside one of the drums was a tube containing contaminated resin. The tube and its radioactive contents are clearly resolved (Figs. 6). Another drum was filled with two layers of packages (Fig. 7). Individual packages can be resolved in the attenuation image.

Table 3. Comparison between TGS and known Pu-239 mass for the mock-residue drums.

TGS ID	Matrix Description	Known Pu-239 mass (grams)	TGS 414 keV assay, Pu-239 mass (grams)	Relative difference [%]	Were self-shielded standards included?	Were lumps detected?	Lump-corrected Pu-239 mass (grams)
RF13	Empty drum	48.1	47.8	-1	No	No	NA
RF14	Empty drum	48.1	48.8	1	No	No	NA
RF15	Empty drum	57.8	58.0	0	No	No	NA
RF16	Empty drum	57.8	59.4	3	No	No	NA
RF17	Empty drum	9.6	9.5	-2	No	No	NA
RF18	Empty drum	48.1	48.5	1	No	No	NA

RF19	Empty drum	9.6	9.9	3	No	No	NA
RF20	Raschig rings	48.1	52.3	8.6	No	No	NA
RF21	Empty drum	96.3	96.5	0	No	No	NA
RF22	Raschig rings	86.6	101	17	No	No	NA
RF23	Empty drum	182.9	179	-2	No	No	NA
RF24	Empty drum	144.4	142	-2	No	No	NA
RF25	Empty drum	28.9	28.2	-2	No	No	NA
RF26	Metal scrap drum (with iron pipes)	57.8	52.3	-10	No	No	NA
RF27	Metal scrap drum (with iron pipes)	117.0	85.4	-27	Yes	No	NA
RF28	Metal scrap drum (without iron pipes)	117.0	93.9	-20	Yes	No	NA
RF29	Metal scrap drum (without iron pipes)	57.8	62.3	8	No	No	NA
RF30	Salt	125.1	135	8	No	No	NA
RF31	Salt	69.0	51.8	-25	Yes	Yes	79
RF32	Salt	259.0	202	-22	Yes	Yes	320
RF33	Salt	182.9	178	-3	No	No	NA
RF34	Wet combustibles	38.5	39.4	2	No	No	NA
RF36	Wet combustibles	19.4	19.0	-2	Yes	Yes	22.8



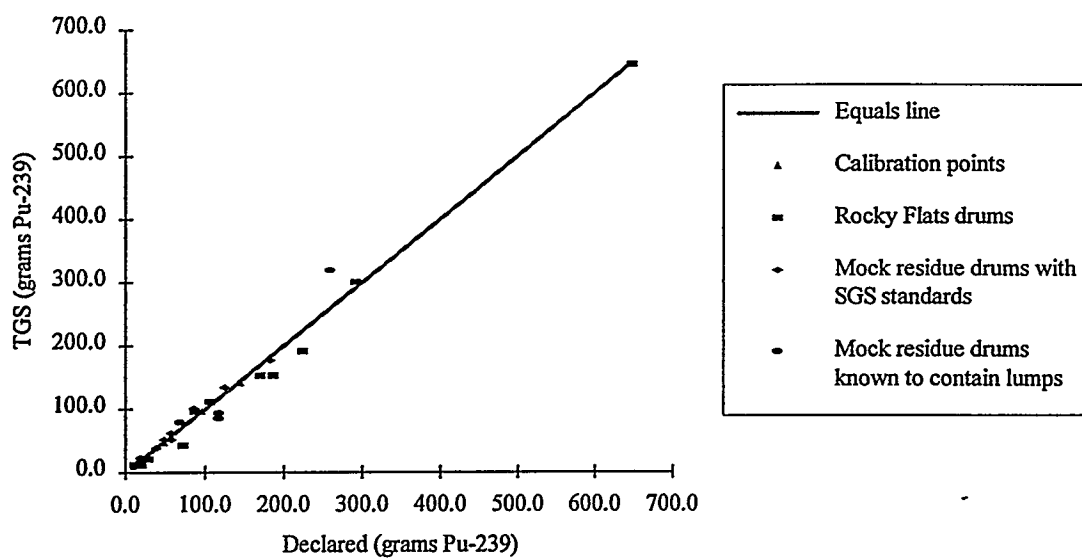


Fig. 4. Comparison between declared and TGS lump-corrected Pu-239 masses for Rocky Flats residues and Los Alamos mock-ups (full-range).

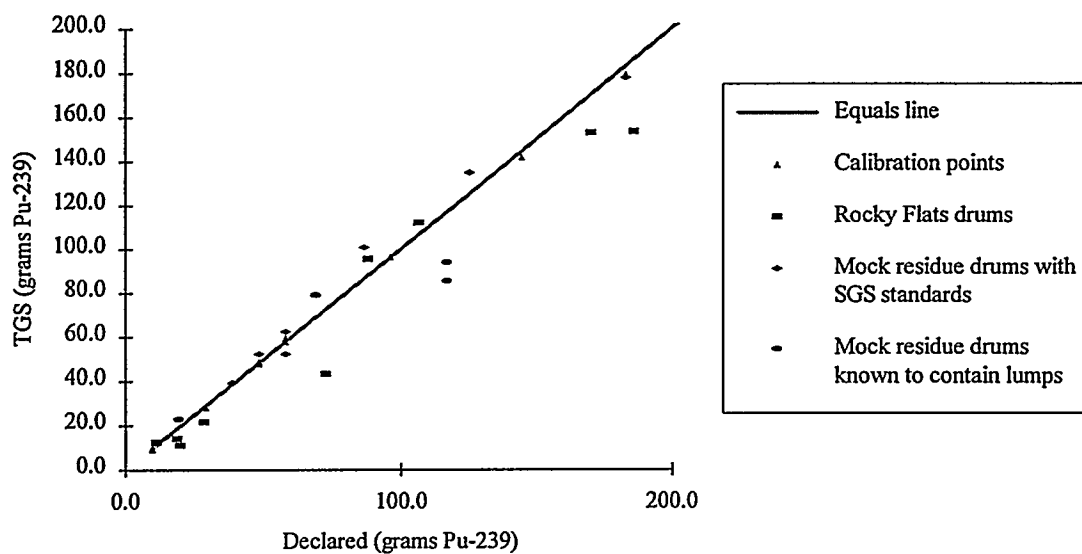


Fig. 5. Comparison between declared and TGS lump-corrected Pu-239 masses for Rocky Flats residues and Los Alamos mock-ups (below 200 grams).

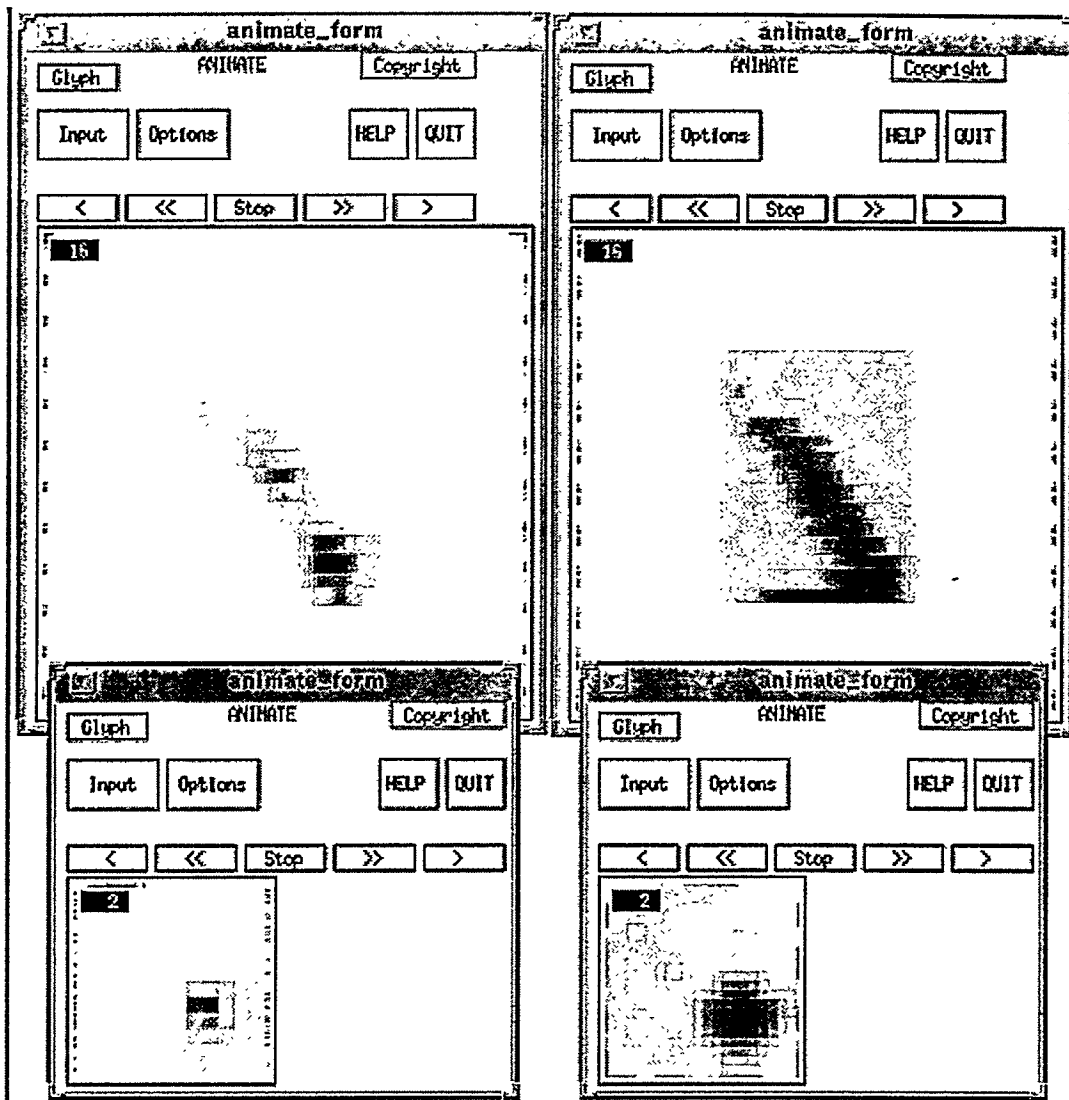


Fig. 6. TGS images of a residue drum (D63251, RF11) that contains a tube filled with leached resin. The left-hand images show the distribution of Pu-239 in the drum. The right-hand images show the distribution of attenuating material in the drum. Upper images are radiographs of the drum contents rendered from TGS tomographs. The lower images are slices through a selected drum layer. Layers are ordered from top to bottom starting from 0 and ending at 14 (15 layers in all). Layer 2 is depicted here.

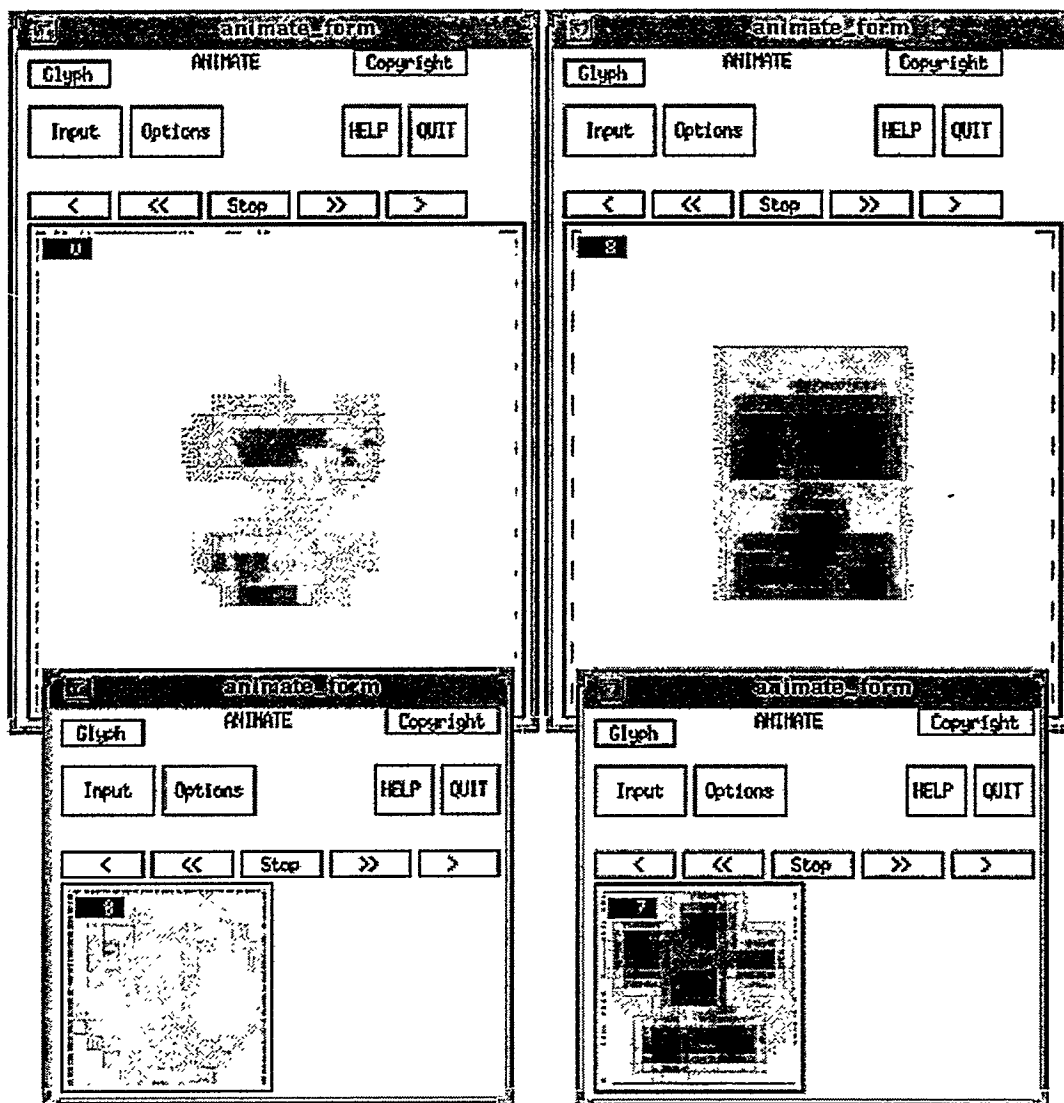


Fig. 7. TGS images of a residue drum (D51181, RF04) that contains packages filled with leached resin. The left-hand images show the distribution of Pu-239 in the drum. The right-hand images show the distribution of attenuating material in the drum. Upper images are radiographs of the drum's contents rendered from TGS tomographs. The lower images are slices through a selected drum layer. Layers are ordered from top to bottom starting from 0 and ending at 14 (15 layers in all). Layer 7 is shown. Six packages can be resolved in this slice.

## SUMMARY AND CONCLUSIONS

Based on the results of the demonstration at Rocky Flats and previous experimental evaluation, tomographic gamma scanning has a number of advantages over currently available techniques for assaying plutonium residues and TRU waste:

1. *Accuracy.* Tomographic gamma scanning represents a significant advance in gamma-ray assay. Tomographic imaging of the attenuating matrix and the distribution of emitting material enables accurate assays of drums. Accuracy is high even for drums that are nearly opaque. From an accuracy standpoint, TGS outperforms all other conventional gamma-ray assay techniques for drum-sized samples with heterogeneous contents by a considerable margin.
2. *Completeness:* TGS systems are capable of executing a variety of scanning protocols, including segmented gamma-ray scanning. TGS systems are capable of identifying and assaying any gamma-ray emitting radionuclide.
3. *Self-attenuation corrections:* TGS is able to reliably detect the presence of self-attenuating material or lumps. Residues containing plutonium metal shot can be identified. Once identified, a correction can be applied that prevents the amount of fissile material from being underestimated. Other assay techniques (for example, those involving thermal neutron interrogation), are biased considerably when lumps are present. Gamma-ray assay techniques other than TGS cannot reliably correct for lumps because the effect of lumps is obscured by other factors.
4. *Complementary:* TGS can provide accurate assays of drums for which other techniques such as passive neutron counting are invalid. Specifically, samples with a high ( $\alpha, n$ ) background cannot be assayed reliably using passive neutron counting. For active neutron assays

- (DDT or shuffler), TGS emission images can be used as a position correction for enhanced accuracy.
5. *Repackaging*: The information about the location of nuclear material provided in TGS images can in principle be used to identify hot-spots in the drum to enable the sorting of packages within drums.
  6. *Throughput*: The ability to accurately and rapidly measure nuclear material within drums without having to unpack the drums for package-counting campaigns saves time, reduces costs, and minimizes radiation exposure to workers. Nominal throughput of TGS is 1 h per drum.
  7. *Transportability*: The mobility of the TGS not only eliminates transportation costs for moving drums to an fixed NDA facility, but it also allows the assay operation to be independent of the building safety envelope. The mobile TGS system has its own criticality safety limit and is powered by a portable diesel generator.
  8. *Maturity*: Over 5 yr of research and development have lead to the current state of TGS technology. A well-engineered prototype system at Los Alamos was the first tomographic scanner to assay transuranic waste. The mobile TGS system is currently being used to assay transuranic waste at Los Alamos waste storage sites. A fully integrated TGS acquisition and analysis software package for use by plant operators will be available before the end of 1995.

The results of the demonstration of TGS technology at Rocky Flats are promising and suggest that TGS may be an effective and timely tool for use in the residue stabilization process. The study included drums with heterogeneous matrices and source distributions that are particularly difficult to assay with SGS or gamma-ray methods in general. Results of TGS assays of metal scrap and Raschig rings indicate that TGS may be competitive with passive neutron methods in metals and for densities that cannot ordinarily be assayed using conventional gamma-ray methods. The TGS method

might even help discover or prevent unacceptable inventory differences that could develop if large biases were discovered by item or by item category prior to treatment.

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Photo of Rocky Flats and Los Alamos personnel standing by the trailer near Building 776.

## REFERENCES

1. T. H. Prettyman, R. A. Cole, R. J. Estep, and G. A. Sheppard, "A maximum-likelihood reconstruction algorithm for tomographic gamma-ray nondestructive assay," *Nucl. Instr. Meth.* A356 470-475 (1995).
2. R. J. Estep, T. H. Prettyman, and G. A. Sheppard, "Tomographic gamma scanning to assay heterogeneous radioactive waste," *Nuclear Science and Engineering* 118 145-152 (1994).
3. R. J. Estep, G. A. Sheppard, T. H. Prettyman, and G. Walton, "Tomographic gamma-ray scanner (TGS) for measuring radioactive waste," *1995 R&D 100 Entry*, Los Alamos National Laboratory, Point of contact: Greg Sheppard (505) 667-6599.
4. T. H. Prettyman, J. K. Sprinkle, Jr., and G. A. Sheppard, "Performance of an advanced lump correction algorithm for gamma-ray assays of plutonium," *Nucl. Mater. Manage.* XXIII (Proc. Issue) 893-896 (1994).
5. T. H. Prettyman, J. K. Sprinkle, Jr., and G. A. Sheppard, "A weighted least-squares lump correction algorithm for transmission-corrected gamma-ray nondestructive assay," *Nucl. Mater. Manage.* XXII (Proc. Issue) 682-690 (1993).
6. R. J. Estep, J. Cavender, "The WIN\_TGS Software Package for Tomographic Gamma Scanner Systems," Los Alamos Report LA-UR-94-2386 (1994).
7. T. H. Prettyman, "Precision Estimates for Tomographic Nondestructive Assay," Los Alamos Report LA-UR-95-3299 (1995).

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