TOMOGRAPHIC GAMMA SCANNING OF URANIUM-CONTAMINATED WASTE AT ROCKY FLATS

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ABSTRACT

A tomographic gamma-ray scanning (TGS) instrument was deployed at Rocky Flats Environmental Technology Site (RFETS) to assist with the deactivation of Building 886. Many 208-L drums containing waste contaminated with highly enriched uranium were measured in order to certify these items for shipment and disposal. This project marks a successful cooperation between RFETS and Los Alamos National Laboratory and is the first major field experience using TGS technology to assay uranium.

INTRODUCTION

The assay of 208-L drums of highly enriched uranium (HEU)-contaminated waste poses a technical challenge. Passive neutron techniques are not sufficiently sensitive to HEU, and active neutron techniques may suffer bias when samples with high boron content are encountered. Large size samples and small heat output makes calorimetry impractical. Gamma-ray assay techniques may be more successful than neutron or calorimetric techniques; however, the dominant gamma ray from $^{235}$U has only 185.7 keV of energy, which is attenuated severely in dense waste matrices. Segmented gamma scanning (SGS) may be applicable to some items, but high gamma-ray attenuation coupled with possible nonuniformities in the matrix and arbitrary location of HEU within a drum can lead to significant assay errors.

Tomographic gamma scanning (TGS) is a relatively new technique that combines high-resolution gamma-ray spectroscopy with three-dimensional imaging [1]. Attenuation within the matrix of a sample is mapped using an external gamma-ray source, then this map is used to correct passive gamma-ray emission data to improve assay accuracy. Research and development of this technique took place at Los Alamos and was supported by the Department of Energy (DOE) Office of Safeguards and Security. Blind tests performed in accordance with a DOE-sponsored performance demonstration plan [2] have repeatedly shown that TGS is the one of the most accurate techniques available for $^{239}$Pu samples. The technique is readily adaptable to other gamma-emitting
radionuclides, such as $^{235}\text{U}$, so it is an excellent candidate for this particular assay task at Rocky Flats.

**PROJECT DESCRIPTION**

Building 886 at the Rocky Flats Environmental Technology Site (RFETS) once housed the Critical Mass Laboratory, which ceased operations in 1989. This building and its associated support structures once held approximately 2700 L of highly enriched uranyl nitrate solution. Removal of the solution was required as part of the deactivation mission, and it has been shipped off site for recycling by Nuclear Fuels Service, Incorporated. In addition, hundreds of drums of HEU-contaminated waste have been produced during the deactivation, all of which must be measured before certification for shipment to the Nevada Test Site (NTS) for disposal.

A mobile TGS instrument (mounted in an 8-m long trailer) was constructed in support of the Los Alamos Transuranic (TRU) Remediation Program and has been used at Los Alamos for characterization of waste, residues, and vitrified items. In March 1995, this mobile instrument visited RFETS for a successful demonstration of TGS technology [3]. In June 1997, it was again deployed at Rocky Flats to assist with the deactivation efforts of Building 886.

Most of the material to be assayed consisted of HEU-contaminated borosilicate-glass Raschig rings, packaged in polyethylene bags in 113-L steel drums, then overpacked in standard 208-L steel drums. The remainder of the items consisted of a variety of HEU-contaminated waste materials, including combustibles, plastics, cardboard, soil dirt, light metals, and chemical waste, all packaged in 208-L drums.

**PREPARATIONS**

No particular modifications to the standard TGS protocol are necessary in order to assay $^{235}\text{U}$. The only significant difference from $^{239}\text{Pu}$ assay is the degree of gamma-ray attenuation: the 185.7-keV line used for quantification of $^{235}\text{U}$ suffers more attenuation than the 413.7-keV line used for $^{239}\text{Pu}$. The analysis software, ARC-TGS, automatically compensates for this attenuation. The standard TGS scan protocol consists of a 65-min, two-pass scan at low spatial resolution. For best sensitivity, a large collimator opening (52 cm$^2$) was chosen as appropriate. Lump corrections (which are difficult for $^{235}\text{U}$) were deemed unnecessary because the contamination arises from HEU solutions, and self-attenuating lumps are unlikely.

Accuracy of the TGS method has been thoroughly validated for $^{239}\text{Pu}$ assay [2,4]. To demonstrate accuracy for $^{235}\text{U}$ assay in the expected RFETS matrix type, data were acquired for several test cases in which placement and number of standards and loading of Raschig rings were varied. Results are shown in Fig. 1. All measurements agreed with the declared quantity within 14%, and the relative standard deviation for the 20 measurements was 6.5%, consistent with the historical accuracy for $^{239}\text{Pu}$. For the 14 cases including Raschig rings, the average difference from the declared value was less than 2%, suggesting that this matrix does not cause significant bias.
The mobile system was calibrated at Los Alamos using NIST-traceable $^{235}\text{U}$ standards (10–100 g range) placed within a Raschig ring mockup drum. Reconstruction algorithms automatically correct for an arbitrary matrix, so a priori matrix calibrations are not generally performed for the TGS. However, they were required in this case to comply with DOE Order 5633.3B. Calibration was verified using the Raschig ring mockup drum and a 23-g $^{235}\text{U}$ standard and was found to have replicate precision of 3% (one standard deviation) and a bias of less than 2% of the declared value. While at RFETS, routine system checks were performed by assaying a 2.4-g $^{235}\text{U}$ standard centered in a Raschig ring mockup drum. The average value from 78 measurements spread over three months was 2.23 g, agreeing with the declared value to within 7%. Replicate precision from these 78 measurements was 7%. Verification measurements were also performed near the lower quantification limit using a 1.0-g $^{235}\text{U}$ standard centered in a Raschig ring mockup drum. The average value from eight measurements was 1.15 g, and replicate precision was 5%. Since no smaller standards were available to verify the TGS performance for sub gram quantities, 1.0 g was administratively chosen as the lower quantification limit, although it seems clear that quantification of 0.5 g of $^{235}\text{U}$ in this matrix should be possible.

The calibration curve is highly linear, but a bias (offset) is observed near the lower limit of detection. This may be seen more clearly when data from two smaller sources (1.9 g and 4.6 g of $^{235}\text{U}$, not NIST-traceable) are included, as in Fig. 2. In practice, a three-parameter curve was fitted to the calibration data, following the basic routine outlined in Ref. [5]: $M_{^{235}\text{U}} = R_{\text{TGS}} A \left(1 + B \exp\left(-\frac{N_{\text{net}}}{C}\right)\right)$, where $R_{\text{TGS}}$ is the corrected emission rate, $N_{\text{net}}$ is the summed net counts in the energy bin, and $A-C$ are adjustable parameters. More recent experience suggests that the simpler form, $M_{^{235}\text{U}} = R_{\text{TGS}} A \left(1 + B/N_{\text{net}}\right)$, may be accurate, and the possible dependencies of $B$ are still under study.

![Graph 1](https://via.placeholder.com/150)

**Figure 1:** Preliminary verification of TGS accuracy for $^{235}\text{U}$ assay in Raschig rings. Calibration is based on a single measurement of 93 g $^{235}\text{U}$ in a noninterfering matrix.

![Graph 2](https://via.placeholder.com/150)

**Figure 2:** A portion of the calibration curve for the Mobile TGS. All data have been collected using well-known standards in a Raschig ring matrix.
RESULTS

A total of 212 samples from the Building 886 deactivation project were assayed, all of them 208-L size. Of these, 123 contained Raschig rings. Others contained combustibles (44 samples), plastics (29 samples), light metals (12 samples), soil dirt (3 samples), and chemicals (1 sample). Figure 3 shows a tally of the results. Most were found to contain less than 5 g of HEU, and many were found below the lower limit of quantification. Maximum throughput was 6 drums/day in addition to a daily calibration recheck with the 2.4-g $^{235}\text{U}$ standard.

![Figure 3: Drums assayed using the Los Alamos mobile TGS, sorted by mass range and item description code (IDC).](image)

Generation of three-dimensional images of the gamma-ray attenuating material and gamma-ray emitting material are essential steps in the TGS quantification algorithm, performed by ARC-TGS. Figure 4 shows projections of the three-dimensional attenuation and emission maps of example RFETS waste drums. The low spatial resolution (about 10 cm) is adequate to perform attenuation corrections required for quantification of HEU. Higher resolution scanning protocols are available, but require a sacrifice of throughput and sensitivity.

Examination of the TGS images suggests that SGS assay may also have been accurate for most of the Raschig ring samples (the SGS algorithm presumes nearly homogeneous samples with nearly uniform radioisotope distribution). Many of the other samples display such a high degree of heterogeneity and/or nonuniform distribution, that they would be poor SGS candidates, but still ideal for TGS. Two such items, "Light Metals" and "Plastics," are shown in Fig. 4.
CONCLUSION AND DISCUSSION

TGS has proven to be a viable technology for assaying drums of HEU-contaminated waste. The measurements described here were used to meet RFETS safeguards, transportation, and disposal requirements, and all of the samples assayed to contain < 15 g of $^{235}$U have been shipped off site. The remainder may require repackaging to reduce individual activity, and the TGS images will provide useful guidance for the repackaging efforts. Good cooperation and planning between Los Alamos, RFETS, and NTS staff, and negotiations coordinated by Los Alamos Technical Associates, helped to make the project a success.
The mobile aspect of the TGS instrument was found to be extremely useful, since it was possible to deploy the instrument quickly to a location near the drum repository. The system was fully operational within two days of delivery to RFETS, and the first waste drum was assayed within one week. Most of the intervening time was spent verifying the calibration and establishing control charts. There were some throughput delays, primarily due to facility concerns and lack of access to the drums. It is estimated that streamlining the process could dramatically improve throughput in the future. In special cases where a large number of samples scheduled for measurement are stored in a single repository, it may be useful to place a skid-mounted TGS instrument within the repository building itself, eliminating the need to remove safeguarded items from the building.

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REFERENCES


