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David J. Mercer, J. Steven Hansen, John P. Lestone, and Thomas H. Prettyman
Los Alamos National Laboratory
Los Alamos, NM 87545

Larry Kayler
Complex Technical Services, L. L. C.
Loveland , CO 80539

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ABSTRACT

A new skid-mounted tomographic gamma scanner (TGS) was designed to assist in the decommissioning of Rocky Flats Building 371. This instrument was used to assay pyrochemical salts as a prerequisite for disposal at the Waste Isolation Pilot Plant (WIPP). The following paper discusses measurement challenges and results from the first year of operation of the instrument.

INTRODUCTION

Building 371 at the Rocky Flats Environmental Technology Site (RFETS) served from 1981 until 1986 as a plutonium recovery facility. It is now used as an interim storage and repackaging facility for plutonium and is one of the two buildings used for residue processing. After all plutonium is shipped offsite, the building will be decommissioned and demolished, with a planned (and rather ambitious) completion date of 2005.

The destination of the plutonium-bearing waste material from Building 371 is the Waste Isolation Pilot Plant (WIPP), and all of this material must be assayed to quantify the plutonium content prior to shipment. One of the more challenging waste forms is approximately 16,000 kg of pyrochemical salts that were generated in the recovery process. This material poses an interesting challenge for assay because it is impure, heterogeneous, and may contain a significant fraction of metal shot. Impurities including ^{241}Am and low-Z materials make neutron counting methods impractical because of the high (α, n) rate, while heterogeneity and the presence of shot may cause significant bias in traditional gamma counting methods. Calorimetry is highly accurate for this material but requires ~20-hour counts and cannot match the pace of the repackaging line, about 18 packages/day.

RFETS was highly motivated to find an assay solution. Tomographic Gamma Scanning (TGS) with "lump corrections," developed at Los Alamos National Laboratory (LANL), was proposed as a possible method. A 1998 demonstration of TGS technology applied to salts had promising results [1]. A new skid-mounted instrument was designed and constructed for use at RFETS [2] and delivered in 1999. This report addresses the instrument's first year of operation, which was primarily dedicated to measurement of pyrochemical salts.

METHOD

The TGS method combines high-resolution gamma-ray spectroscopy with three-dimensional imaging for improved assay accuracy [3]. It is particularly effective for heterogeneous samples. The assay begins with an active phase, where the attenuation characteristics of the matrix are mapped using an external ^{75}Se source. The sample is rotated and translated during the measurement to cover the necessary phase space for image reconstruction; typically, 150 measurements are made for each of 16 vertical layers. After the active phase, the external source is shuttered off, and

gamma rays from ^{239}Pu in the sample are counted passively from the same 150×16 positions. Data are reduced using a maximum-likelihood estimation maximization (MLEM) algorithm [4] to deduce the amounts of Pu in each of 1520 volume elements, which are then summed to give the total amount of Pu in the sample.

The correction for matrix attenuation is necessary, but not sufficient, to accurately assay pyrochemical salt, which may contain macroscopic particles of Pu metal. A 2-mm diameter “lump” of metal, for example, self-absorbs about 30% of the 414 keV gamma rays that it emits. Assay based on this single energy would therefore give a biased result (underestimate), depending on the typical particle size. Fortunately, the self-absorbed fraction is a predictable function of gamma-ray energy, so an algorithm based on multiple ^{239}Pu energies can compensate for the bias. Such “lump correction” algorithms have been explored [5] and have been further refined for this application. An example of the applied algorithm is shown in Fig. 1.

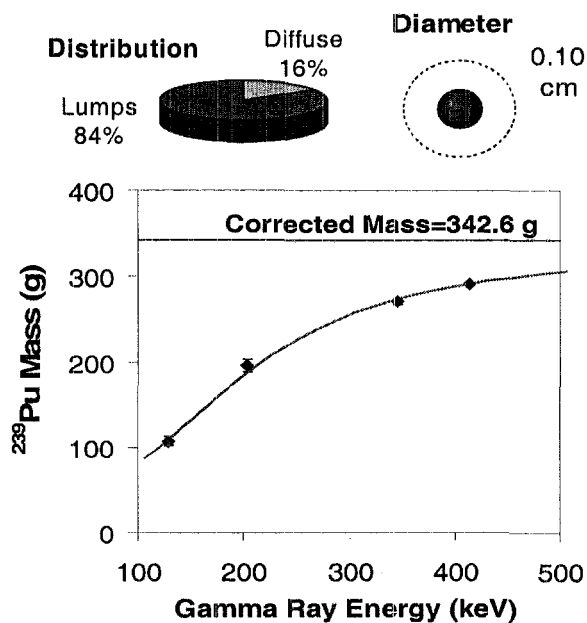


Fig. 1: Lump-correction algorithm as applied to sample RF98062. The uncorrected assay values at 129, 203, 345, and 414 keV are 107.6, 195.5, 271.5, and 290.9 g. The algorithm finds a physics-based fit to the data with three parameters: corrected mass, distribution (fraction of material in lumps), and effective diameter. The corrected mass is within 2% of the actual Pu mass for this sample, 349.0 g, as determined by calorimetry. 84% of the material is found in lumps, with an effective lump diameter of 1 mm.

Several significant upgrades have been made to the TGS application software. The interface has been improved, and the feature of whole-spectrum capture has been added (which allows re-analysis of any region of interest). The most significant improvement, however, is automation of lump corrections.

HARDWARE

The new TGS instrument incorporates many design improvements over the older laboratory prototype. Most significantly, the new unit is skid-mounted, which allows placement within the same room as the materials scheduled for assay. This offers a tremendous savings in the cost and effort involved in moving samples. The design has been “ruggedized,” with a cantilevered lift design instead of the four-post “bedstead” prototype design, which was prone to wear and breakage. The instrument is shown in Figs. 2 and 3, and further details may be found in Ref. [2].

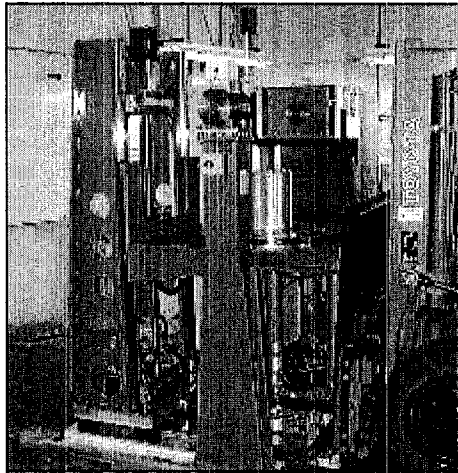


Fig. 2: New skid-mounted TGS, mounted on a forklift prior to installation. The compact cantilevered design was a significant improvement over the older "bedstead" design.

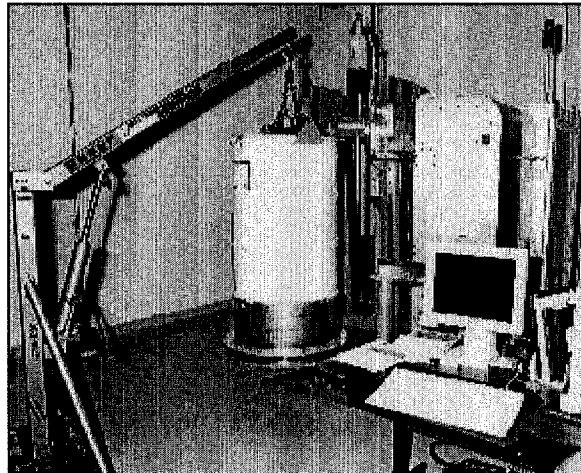


Fig. 3: Instrument installed in RFETS Building 371. A small fraction of the waste items measured in the first year was 55-gallon drums, as shown. The salts were measured in 7-inch diam. \times 10-inch tall cans.

OPERATIONAL EXPERIENCE

To verify TGS accuracy, about 10% of the samples were also measured using calorimetry/isotopics techniques. Calorimetry is more time consuming than TGS, but is sufficiently accurate to serve as a "gold standard." The results from 96 unique samples are presented in Fig. 4. The uncertainty per measurement was found to be 9.4%, and the inventory bias was found to be $(-0.04 \pm 0.99)\%$. Cal/Iso reported a total of 13.467 kg for these samples, and TGS reported 13.461 kg. An earlier analysis is detailed in Ref. [6].

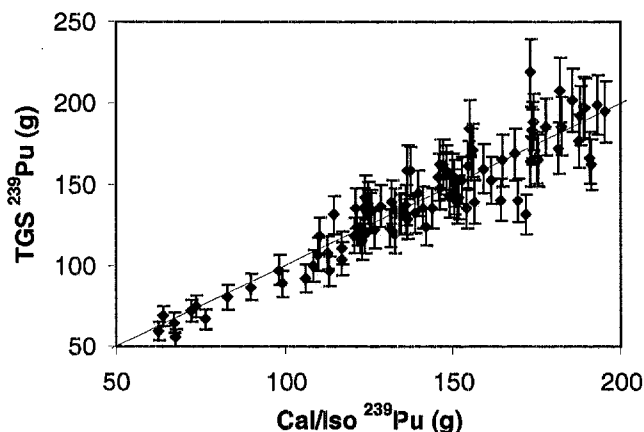


Fig. 4: Comparison between Cal/Iso and TGS assay results, showing no significant bias. This is a major accomplishment for difficult-to-assay pyrochemical salts. The diagonal line indicates unity.

Several unexpected problems arose during the first year of operation. (1) Background from the high-energy gamma-ray continuum was an order of magnitude more intense than anticipated. This reduced the sensitivity of the instrument to about 1 g of Pu/sample. Since most samples were >50 g, the salt measurement campaign was barely affected, although some additional shielding was retrofitted to help mitigate background. (2) An additional Sn liner was added to many of the

samples as personnel shielding, and this liner nearly eliminated the 136 keV gamma rays from ⁷⁵Se, which are crucial to attenuation mapping. The analysis software was robust enough to handle this deficit. Although improved algorithms were explored, these were never implemented. (3) At some undetermined time, a cable in the pileup rejection system was severed. This caused a dramatic increase in background levels below 176 keV, with a loss in sensitivity and precision until the problem was found and corrected. This problem did not affect accuracy. (4) Software quality assurance required considerably more effort than was anticipated, and software configuration management also posed some difficulties. The rushed schedule was a complicating factor.

Throughput as high as 20 samples/day was achieved routinely. By August 2000 the measurements of approximately 16,000 kg of salts were completed. All of the containers are now interred at the WIPP, meeting a major milestone for RFETS. The new skid-mounted TGS has proven to be of tremendous value to the facility, offering the highest throughput and greatest versatility of any nondestructive assay instrument at RFETS.

ACKNOWLEDGMENT

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