

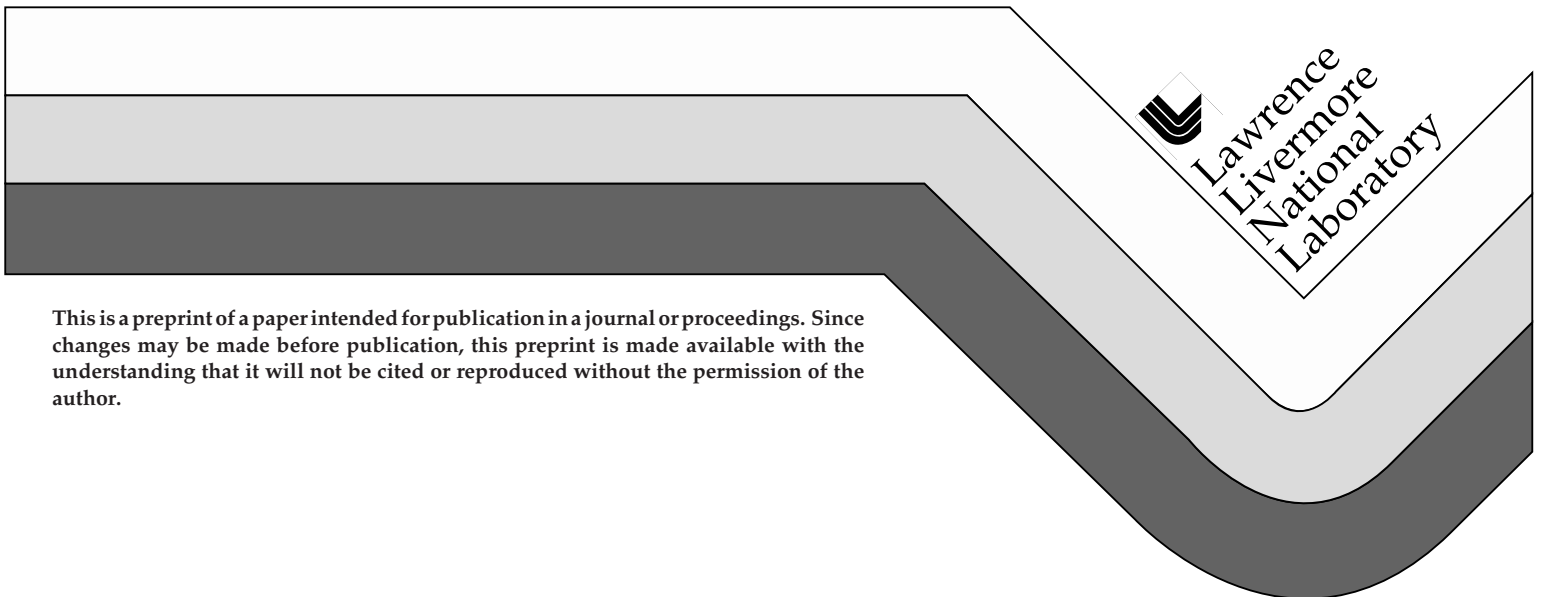
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GAMMA-RAY SCANNER SYSTEMS FOR NONDESTRUCTIVE ASSAY OF HETEROGENEOUS WASTE BARRELS[†]

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ABSTRACT

Traditional gamma measurement errors are related to non-uniform measurement responses associated with unknown radioactive source and matrix material distributions. These errors can be reduced by application of tomographic techniques, that measure these distributions.

LLNL has developed two tomographic-based waste assay systems. They use external radioactive sources and tomography-protocol to map the attenuation within a waste barrel as a function of mono-energetic gamma-ray energy in waste containers. Passive tomography is used to localize and identify specific radioactive waste contents within the same waste containers. Reconstruction of the passive data via the active images allows internal waste radioactivities in a barrel to be corrected for any overlying heterogeneous materials, thus yielding an absolute assay of the waste radioactivities. Calibration of both systems requires only point source measurements and are independent of matrix materials.

The first system housed at LLNL has participated and successfully passed the requirements of a formal DOE-sponsored intercomparison study. The second system is housed within a mobile waste characterization trailer, and has made measurements at three DOE facilities. Both systems have measured 1 to 70 grams of plutonium within a variety of waste matrix materials. Laboratory and field results from these two systems over the past several years show that both systems are capable of precisions of 1 to 3% and accuracies to within 30% of the true values of known standards.

Work at the Hungarian Institute has led to the development of modular systems, which can be adapted to several needs. Two system configurations are under construction. One includes additional features to the traditional SGS system, providing a warning when the matrix or waste radioactivity contents may result in serious errors if additional measurements or corrections are not made. A second more versatile and sophisticated SGS-TOMO based system has been developed, which will permit the more accurate safeguards-type measurements to be made on waste barrels.

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1. INTRODUCTION

Traditional gamma safeguards measurements have been performed using a segmented gamma scanning (SGS) system. SGS accuracy relies on the assumption that the sample matrix and the activity are both uniform for a segment. In fact, waste barrels are often highly heterogeneous, and span a wide range of composition and matrix type. Thus SGS system errors are related to non-uniform measurement responses associated with unknown radioactive source spatial distributions and matrix heterogeneities. These errors can be reduced by imaging techniques that better measure the spatial locations of sources and matrix attenuations.

Here we describe the collaboration between the Lawrence Livermore National Laboratory (LLNL) and the Institute of Nuclear Techniques (INT) of the Technical University, Budapest to share results obtained by two different gamma-ray nondestructive assay (NDA) systems used for imaging waste barrels. The basic principles are the same, but the approaches are different. Key factors to judge the adequacy of a method are the accuracy, precision, and detection limit. Test barrels containing a known mass of plutonium (Pu) that represent waste types to be measured are used to determine the basic system parameters and performance. LLNL is participating in a DOE Performance Demonstration Program study. Understanding this program can be useful in a planned European Intercomparison Exercise.

2. GAMMA-RAY NDA MEASUREMENTS AT LLNL

LLNL is developing an emerging gamma-ray NDA technology that will identify and accurately quantify all detectable radioisotopes in closed containers of wastes, regardless of their classification: low level, transuranic or mixed, which contains radioactivity and hazardous organic species. It is called Active and Passive Computed Tomography (A&PCT) [1]. A&PCT uses two separate measurements. The first is an active interrogation of the barrel by an external radioactive source(s) and the second is a passive measurement of the radioactive source(s) within the barrel. The results of these two measurements are combined to produce an attenuation corrected gamma-ray assay of the barrel. The gamma-ray A&PCT method involves: (1) Data acquisition; (2) Image reconstruction and assay; and (3) Gamma-ray spectral analysis. The R&D efforts associated with each of these three components at LLNL and the performance of the LLNL system is described.

2.1 Isotopic Measurements by Passive and Active CT

Currently there are two working A&PCT systems. One is located at LLNL, the other is within a mobile Waste Inspection Tomography (WIT) trailer being developed in collaboration with BioImaging Research, Inc.[2] The former, IMPACT¹, has been tested on simple cases, e.g., well characterized radioactive sources without attenuators, within uniform attenuators, and within mock heterogeneous-waste barrels. These sources do not have lump or clumping attributes. IMPACT has also been used to assay LLNL waste barrels containing up to 4 grams of weapons grade (WG) Pu. The system within the WIT trailer has also been used to characterize well known radioactive sources within a variety of waste matrices in addition to several waste barrels. At LLNL, WIT characterized barrels that contained smaller containers with solidified chemical wastes; at RFETS,² WIT measured barrels with low-density combustible matrices. At INEL,³ WIT characterized graphite-, glass-, and metal-matrix barrels, lead-lined barrels with combustibles, and very dense sludge barrels. The Pu mass within these barrels ranged from 1 to 70 g.

IMPACT uses a single collimated aperture (*width*:5 cm, *height*:5 cm, *length*:25 cm) for a high-purity Ge detector (coaxial, >90% relative efficiency) and a ^{166m}Ho (3 mCi) external radioactive source. The barrel manipulator can handle up to 500 kg.[1] The system can be operated in two different modes: (1) collimated gamma-ray scanner (CGS); or (2) active and passive CT. The CGS mode is similar to SGS except our detector is much more collimated. CGS is used to determine the height and isotopics of source(s) within a barrel.

The active data acquisition for both CGS and A&PCT measures the attenuated gamma-ray spectrum emitted from a ^{166m}Ho source. For CGS the data is integrated while the barrel is continuously rotated at each slice. In ACT the data is obtained by discrete translation and rotation of the barrel for each slice. Typically 14 translations (or ray sums) and 21 angles (or projections) are required per elevation (or slice). An entire barrel requires 19 slices in which one slice is below the barrel. ACT yields a quantitative attenuation map of the waste matrix within the barrel.

To obtain passive CGS or CT images, the ^{166m}Ho source is shuttered; and the barrel is scanned in a similar fashion used to obtain the active CGS or ACT data, respectively. In the passive mode, the detector collects and records individual energy regions or the entire energy spectrum for gamma-rays emitted from within the barrel. The passive measurements are the integrated radioisotopic activity, modified by one or a multiple of exponential attenuations along the path from the source position within the barrel to the detector. The function that is imaged for PCT is the measured gamma-ray activity at one or more energies of all detectable radioisotopes within a barrel.

The active scans generate attenuation data at specific ^{166m}Ho gamma-ray energies, which when appropriately interpolated or extrapolated, yield attenuation data at each significant gamma-ray energy identified in the passive barrel scan. Thus, the active scans provide energy-specific attenuation data so that attenuation corrections can be made for each internal gamma-

¹ Isotopic Measurements by Passive and Active Computed Tomography.

² Rocky Flats Environmental Technology Site, CO.

³ Idaho National Engineering Laboratory, Idaho Falls, ID.

ray energy identified. To attain an absolute assay measurement, the A&PCT systems are calibrated once on an absolute scale by simple measurements of calibrated radioactive point sources and this method does not need additional calibrations for different Pu gram-loadings or waste matrices.

2.2 Image Reconstruction/Assay Algorithms

Our image reconstruction/assay algorithm uses a 3D MLEM⁴ code developed in collaboration with University of California at San Francisco (UCSF). We incorporated a sum-of-squares and chi-square error estimate to determine the accuracy of the fit between the measured sinogram data and the sinograms generated by the algorithm based on the current image estimate. This allowed us to better track the convergence of the algorithm. We implemented the ability to use a measured collimator response function instead of one calculated from the collimator dimensions. This was required to reconstruct data measured with systems using septa,⁵ which improves the detector-aperture aspect ratio (aperture width/aperture length). The code was upgraded to compensate for a measured background level developed with UCSF and a background estimation scheme developed at Los Alamos National Laboratory (LANL).[3] Unfortunately neither is always successful.

This code was adapted from a code specifically designed for medical imaging geometries, which have much larger collimator-aspect ratios. There are several assumptions in the code, which are valid for the medical imaging case, but not for the barrel-assay problem. Thus, we have entirely re-written the reconstruction software based on the physics and geometry of the barrel NDA problem.

We developed a new maximum likelihood based algorithm that maximizes the correct likelihood function based on the joint probability density function of the peak region count and the background region count for each measurement.[4] This avoids any physically unrealistic “negative counts” that must be set to zero in other estimation approaches to this problem, such as MLEM. The zeroing of negative counts can result in a positive bias in assay estimates (see Section 2.5), and our new method avoids this problem. We are incorporating this algorithm into our new image reconstruction and assay code.

2.3 Isotopics Analyses/Assay

A&PCT allows the gamma-ray spectroscopy analyses portion of the absolute assay problem to be simplified. The passive measurement localizes the activity of interest into slices for CGS or small-sized (5 cm on a side) volume elements (voxels) for PCT. The absolute detector efficiency for each of these elements can be directly related to calibration measurements of known radioactive point sources. The active measurement provides the data for the attenuation correction. The absolute assay can then be obtained by adding the activities in each of the voxels of the reconstructed and attenuation corrected PCT image. Also, most of the nuclides of interest emit gamma-rays of more than one energy, thus allowing additional checks on the image reconstruction and the absolute assay results.

⁴ Maximum Likelihood Expectation Maximization

⁵ This work is mainly required for the WIT trailer A&PCT system.

We developed some procedures for determining the isotopic distribution of the assay. We analyze the overall statistical quality of the data and the presence of interference peaks from other isotopes such as ^{235}U and ^{237}Np . Based on this analysis the procedure can then select appropriate regions of the spectra for more detailed analyses. In order to support these concepts we also developed codes that can sum spectra from several slices of A&PCT data in order to accumulate sufficient statistics for barrels with low radioactive mass.

Analysis of the spectroscopic data from IMPACT includes: (1) summing the data from each slice; and (2) summing the data for the entire barrel to determine the isotopic ratios for: $^{238}\text{Pu}/^{239}\text{Pu}$, $^{240}\text{Pu}/^{239}\text{Pu}$, $^{241}\text{Pu}/^{239}\text{Pu}$, and $^{241}\text{Am}/^{239}\text{Pu}$; and (3) inspecting the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio for each slice to evaluate isotopic homogeneity. Except for the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio, the values are determined from at least two different parts of the gamma-ray spectrum. The isotopic ratios are then combined with the A&PCT image reconstruction/assay data to calculate the total alpha curies, thermal power, and fissile gram equivalent for each analysis.

2.4 Performance Measures

One performance objective is to satisfy the Quality Assurance Program Plan (QAPP).[5] The QAPP identifies the quality of data necessary to meet the specific data quality objectives associated with the Department of Energy's (DOE) Waste Isolation Pilot Plant (WIPP) transuranic (TRU) waste characterization program. Two parameters describing the waste must be determined; the total alpha activity and the activity of the individual isotopes present. The quality assurance objectives (QAOs) for precision (% relative standard deviation), accuracy (% recovery), minimum detectable concentration (MDC), completeness, and total bias are stated in the QAPP. The QAPP requires that each NDA system perform 15 replicate measurements for 4 ranges of waste activity (nominal compliance values are 0.1, 1.0, 10, and 160 g WG Pu). Between each measurement the barrel must be completely removed from the NDA system.

The NDA Performance Demonstration Program (PDP) is designed to help ensure compliance with the QAPP QAOs.[6] The PDP consists of a series of tests conducted on a regular frequency and of increasing matrix/source complexity to evaluate the capability for NDA of TRU waste throughout the DOE complex. Each test is termed a PDP cycle. These evaluation cycles are blind tests that provide an objective measure of the reliability and performance of NDA systems. The PDP consists of a set of 208-L barrels and a configuration that includes provisions to install and physically fasten a matrix in place in addition to allowing for the convenient external introduction and precise positioning of PDP standards (radioactive sources) within the barrel volume. The PDP requires only 6 replicate measurements and removal of the barrel between replicates.

Presently there are 4 barrels: air (no matrix), ethafoam, combustibles and glass. Aluminum source insert fixtures are provided for each of three insert tube radii: Center or 0R, 0.5R and 0.8R, where R is the barrel radius. PDP standard(s) are positioned at desired vertical locations in the three insert fixtures. Several versions of standards will be used in the program. The initial standards are weapons-grade (WG) plutonium dioxide (PuO_2) uniformly mixed in diatomaceous earth and then encapsulated in a dual stainless steel cylinder configuration (*o.d.*: 5 cm, *l.*:23 cm). Each assembled PDP barrel for every official cycle includes a tamper

indicating seal. Currently 3 cycles have been completed. LLNL participated in cycle 2 informally and in cycle 3 formally.

2.5 IMPACT Performance

We were not ready to officially participate in the PDP cycle 2, but we were able to gain access to one of the two barrels. This barrel has an ethafoam matrix and was loaded with 4 PDP standards (three 0.3 g and one 0.03 g of WG Pu).[7] The attenuation caused by the barrel and ethafoam matrix was measured by ACT, which required 17 slices with a ray sum integration time of 10 s. We ran several PCT scans (14 ray sums, 21 projections, and 17 slices) with different ray sum integration times as shown in Fig. 1. This data shows that as the integration time decreases the assay result increases. The increasing amounts of additional Pu mass is attributed to increasing amounts of noise added as a real signal to the PCT image. The additional mass is due to the fact that the MLEM algorithm does not accept any negative values (see Section 2.2). Thus, for signals that approach the noise level of our system we end up with a positive assay bias. Methods applied to account for this bias are not always successful; thus, we have developed another method[4]. In the mean time, we need to use integration times that are sufficiently long to provide a signal that is well above the noise. Of course the integration time required is a strong function of the activity of the barrel and its attenuating matrix. For example, we show below (Cycle 3) that for high activity barrels we've not yet seen this bias at short integration times.

A 15 replicate study was completed for one of the four QAPP activity ranges. Three PDP standards (3, 0.3 and 0.3 g of WG Pu) were loaded into the combustible matrix barrel. For all scans 9 slices were obtained. The ACT ray sum integration time was 15 s. Results for a complete sampled (14 ray sums, 21 projections, and 9 slices with a ray sum integration time of 25 s) PCT data set and an under sampled PCT data set are shown in Fig. 2. The complete data set results in an accuracy of 80.4% with a precision of 2.8%, which meets QAPP requirements. The under sampled data was obtained by computationally removing every other ray and projection from the complete sample data set. This results in a 4X reduction of the complete data set or 7 ray sums, 9 projections and 9 slices. The under-sampled data precision is 3.9% with an accuracy of 79.0%. Since the difference in these two data sets is <2% and both meet QAPP requirements, we conclude that we can use the under sampling protocol to assay a barrel with one quarter the passive data acquisition time. The under sampled results were verified experimentally.

LLNL formally participated in and passed PDP cycle 3. This test consisted of the combustible-matrix barrel with 7 PDP standards (50, 15, two 3, two 0.3 and one 0.03 g) resulting in a total of 71.36 g WG Pu; and the glass-matrix barrel with 4 PDP standards having a total of 98.3 g WG Pu.[8] ACT scans consisted of 17 slices with a ray sum integration time of 6 s.

The PCT data consisted of 7 ray sums, 9 projections and 17 slices with a 20 s ray sum integration time. Our assay results are summarized in Fig. 3. For the barrel with the combustible matrix and 71.36 g WG Pu, the measured mean is 47.1 g. The accuracy is 66.0% with a precision of 0.58%. For the barrel with the glass matrix and 98.3 g WG Pu, the measured mean is 69.9 g, an accuracy of 71.1% with a precision of 0.84%. The average of the

two accuracies and precisions is 68.5% and 0.7%, respectively. Therefore, the bias of our IMPACT system is ~30% low. Representative three-dimensionally rendered ACT and PCT images of the glass matrix barrel are shown in Fig. 4.

Two additional measurements were made on the glass matrix barrel to better understand the system bias at shorter integration times. PCT measurements at 10 and 5 s ray-sum integration times resulted in 69.4 and 69.1 g, respectively. These results agree with the 20 s PCT assay to within 1%; Thus, for signals well above the system noise our assay results are independent of ray-sum integration times.

2.6 Summary of LLNL Developed A&PCT systems

There are two A&PCT scanners. One is at LLNL; the other is within a mobile WIT trailer. These systems have been used to assay a wide range of radioactive waste from 1-70 g of Pu within matrices from combustibles to sludge. The preliminary performance of LLNL's IMPACT scanner was determined to have an accuracy of ~70% with a precision to within a few percent for combustible and glass matrices. To increase system throughput LLNL completed a preliminary design for the multiple HPGe detector A&PCT system.[9] Additional R&D is required to better understand the negative 30% bias for good statistical data sets and to reduce the high positive bias for poor statistical data. New image reconstruction and assay codes are near completion and hopefully will reduce these biases. LLNL is also working on automating the isotopic analysis and determining our systematic uncertainties, as well as determining IMPACT's MDC and its performance at the other 3 QAPP activity ranges.

3. COMBINED SGS-LOW RESOLUTION TOMO SCANNER AT INT

INT's effort is aimed at modifying an existing SGS to employ tomographic imaging principles to assay heterogeneous waste. An SGS system was put into operation at a Hungarian Nuclear power plant to characterize solid waste containers of low- and medium-density matrices compacted into 200-L barrels before shipment to the waste disposal site. Results of measurements of 144 compacted barrels, 7 segments for each, demonstrated [10] that transmissions measured at 1275 keV (^{22}Na) are mostly in the range 1-15% (average density 0.65 g/cm^3). Calculations and measurements also demonstrated that a very large error can be introduced if the transmission is lower than 20% or the average density is above 0.2 g/cm^3 . [11,12] These high-attenuating cases become difficult when no a priori information is available because the systematic error is not known. There are some techniques to obtain some indication of heterogeneity, but for these cases significant improvement can only be obtained when some imaging principles are used for measurements of unknown barrels. Even low spatial-resolution imaging can lead to significantly smaller measurement errors. This is demonstrated by measurement using the modular tomographic arrangement developed by INT.[10,11,12] A cost-benefit analysis can provide the compromise between the necessary accuracy level and cost. Lower accuracy is needed for waste management and higher accuracy is needed for safeguards purposes. In both cases, however, monitoring of the measurement is needed to prevent unknown errors, sometimes 2-3 orders of magnitude. These large errors are possible under unfavorable conditions.[10]

3.1 Modular Tomographic Instrumentation

The main objectives of the work at INT include: (1) installing a system that will work together with an SGS; that will warn an operator when conditions exceed the capability of appropriate SGS measurements; and that may switch to a Tomo measurement able to measure medium- to high-density waste barrels with an accuracy level specified; and (2) developing a system capable of safeguards oriented measurements. The latter may include higher accuracy, localization and identification of hot spots of safeguards significance.

To fulfill these requirements, systems are being developed with modular units. The modularity of these units will allow them to configure several instruments, each of them to be used for different purposes. Two systems are under construction. One is a combined SGS and tomography (SGS-TOMO) system. This is an upgrade to the existing SGS system. This will be used to monitor the heterogeneity of a 200-L barrel for SGS measurements, but also provides an additional tomographic measuring capability for barrels. This is a low spatial resolution, low cost system. The other is a mobile tomographic system with variable resolution primarily designed for safeguards purposes.

The mechanical part of the mobile tomographic system consists of several modular units. A manipulator/stage is used to rotate and elevate (75 cm) up to a 150 kg, 200-L barrel. The detectors, collimators, and shielded transmission radiation source are supported by a space frame with adjustable positioning capability for alignment. For the SGS upgrade to the SGS-TOMO system only the detectors, detector collimators and detector supporting frame is required.

The electronics consist of a room temperature (low energy resolution) 16-channel detector (including preamplifiers) with signal processing units; a single detector high-energy resolution gamma-ray spectrometry instrumentation; a stepping motor card and driver for a maximum of 3 motors; sensors for rotation angle and/or translation or elevation (up to 3 channels); computer (laptop for the mobile unit); and control units for elevation of the barrel, radioactive source shutter operation, etc. All of these are required for the mobile tomography system. The SGS-TOMO system only requires 10 channels of the 16-channel detector, the signal processing components, the control units and a computer.

The software consists of four separate components. One is the control program. It includes tracking the stepping motor for the mobile Tomo system or processing signals from motion sensors for the SGS-TOMO system and it stores each detector signal separately. Data acquisition for several geometrical modes can be implemented by a simple menu interface. Two image reconstruction programs are based on the algebraic and simultaneous iterative least square algorithms. These programs are 2D, i.e., they work on a slice by slice basis. Signal and image processing programs are used to extract information and for presentation of results. Simulation programs are able to model arbitrary geometrical configuration, including SGS.

3.2 Modes of Operation

For the SGS-TOMO scanner, the tomography components do not disturb a normal SGS scan. Sensors are attached to the rotation and elevation stages to detect the position controlled by the SGS system. The tomography system has two basic modes of operation. First, there is a monitoring mode. In this mode, for a normal SGS sequence, the tomography

system monitors density as well as activity distribution in each segment. The system gives a warning in cases of conditions exceeding the capability of the SGS system, otherwise the expected error will be higher than the threshold set in advance.

The second mode is a computed tomography imaging measurement. In the case of a warning following an SGS measurement, another measurement in the tomo operational mode should be made to provide the accuracy level needed. This mode can be invoked manually by the operator or automatically. Depending on the accuracy required, the tomography measurement mode usually needs a longer measurement time than the SGS mode. For waste management purposes a factor of 1.5-2 is typical. For safeguards purposes it can be longer. Generally, the total amount of radioactivity is needed and not the image. Sometimes localization of hot spots is required in those case where removal of some activity would characterize the barrel as a lower category of waste.

3.3 Summary of INT Efforts

Waste management and safeguards have different needs for the assay of 200-L barrels. Very large errors can be introduced in measurements made by a standard SGS system. A modular system developed by INT seems to be capable of making measurements that fulfill the different waste assay and safeguards requirements. The cost factor should also be considered when building a low-spatial resolution, low cost monitoring system attached to a standard SGS system.

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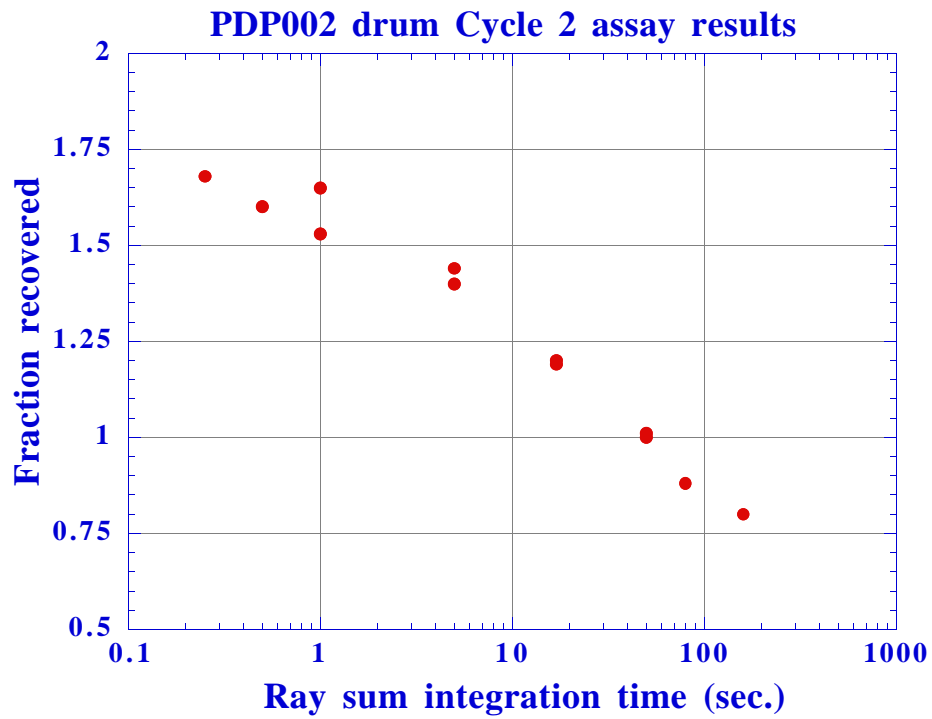


Fig. 1 Summary of the IMPACT assay results as a function of ray-sum integration time for PDP cycle 2 ethafoam-matrix (PDP002) barrel with 0.93 g of WG Pu. Some integration times were repeated two or three times.

Replicate scans of PDP 003 (combustibles) with 3.6 g WG Plutonium

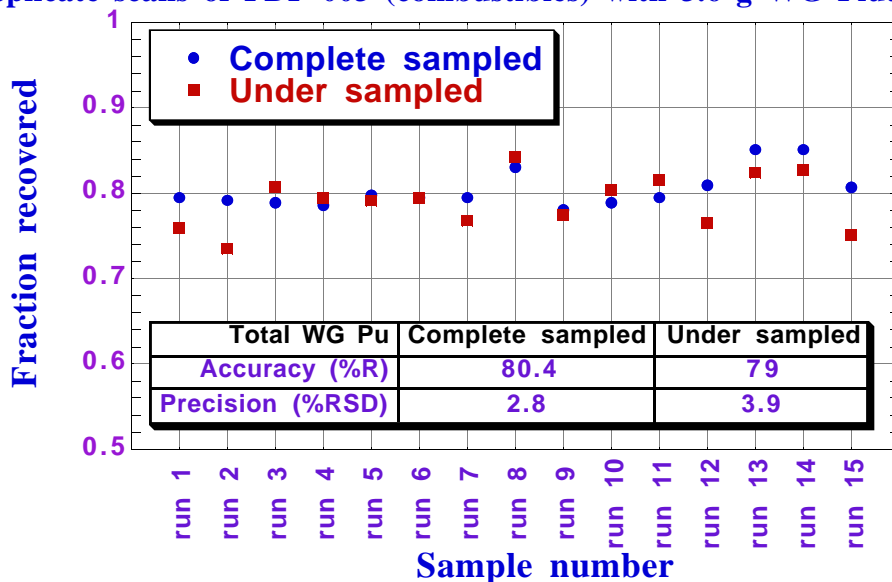


Fig. 2 Summary of 15 replicate measurements for the combustible-matrix (PDP003), 3.6 g WG Pu barrel. Complete- and under-sampled (by a factor of 4) data are shown for comparison.

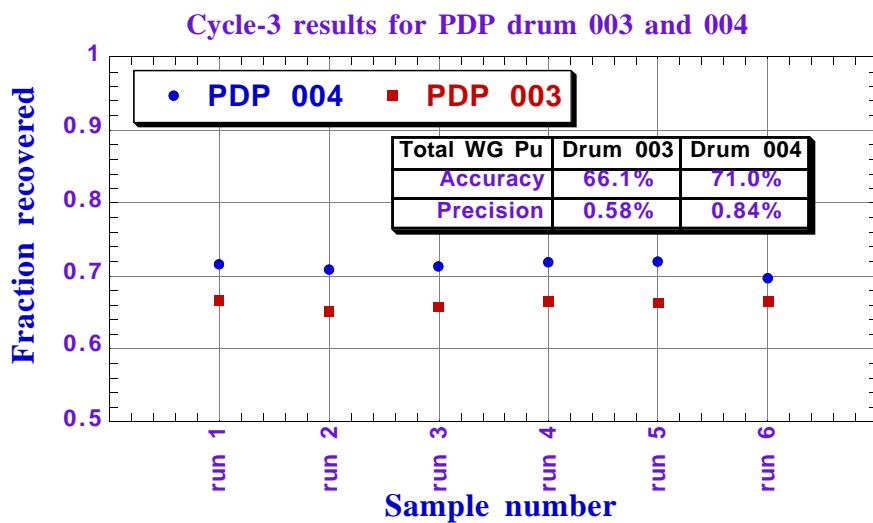


Fig. 3 Summary of the assay results for PDP cycle 3 combustible (PDP003) and glass (PDP004) matrices with 71.36 and 98.3 g of WG Pu, respectively.

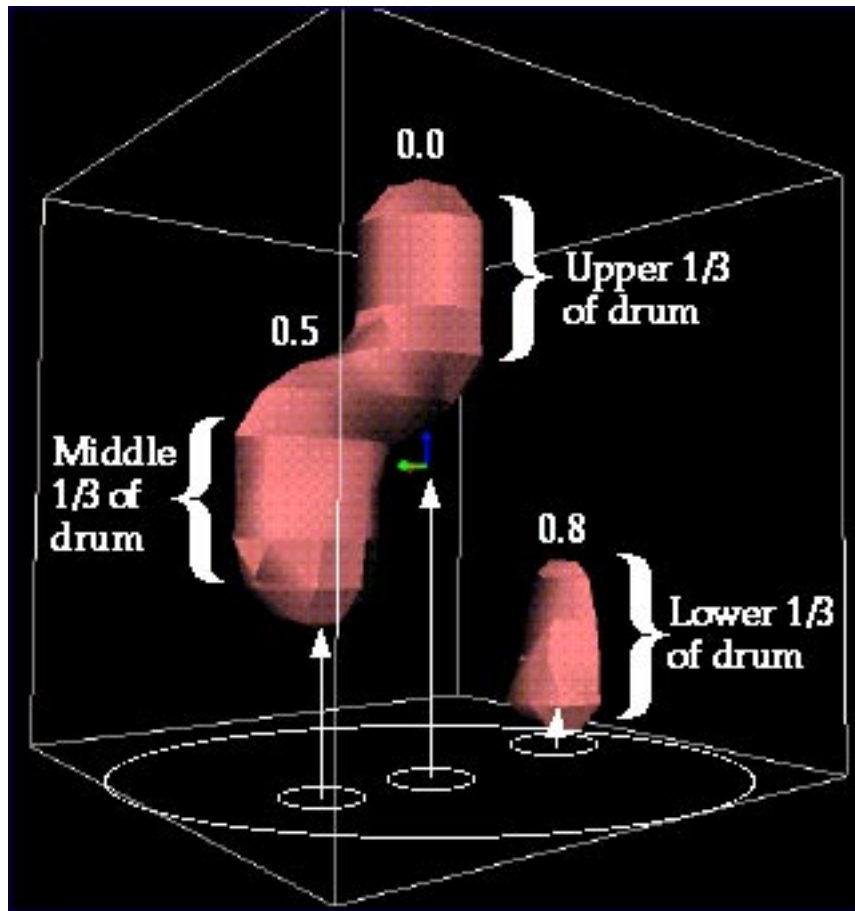


Fig. 4 Representative three-dimensionally rendered PCT image of the PDP cycle 3 glass-matrix barrel. The standards for the glass-matrix barrel consisted of WG Pu: 65 g at 0R, 30 g at 0.5R, 3 g at 0.8R, and 0.3 g at 0R, at a height from the bottom of the barrel of 40.6, 17.8, 0, and 12.7 cm, respectively. Note the smallest mass standard was not recovered.

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