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# NONDESTRUCTIVE ASSAY USING ACTIVE AND PASSIVE COMPUTED TOMOGRAPHY[1]

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## ABSTRACT

The United States Department of Energy (DOE) has over 600,000 transuranic (TRU) waste drums temporarily stored at nearly 40 sites within the United States. Contents of these drums must be characterized before they are transported for permanent disposal. Traditional gamma-ray methods used to characterize nuclear waste introduce errors that 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.

The Lawrence Livermore National Laboratory (LLNL) has developed two tomographic-based waste assay systems. They use external radioactive sources and tomography-protocol to map the attenuation within a waste drum 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 drum 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 is housed at LLNL and was developed to study and validate research concepts. The second system is being developed with BioImaging Research, Inc. (BIR) and is housed within a mobile waste characterization trailer. This system has traveled to three DOE facilities to demonstrate the active and passive computed tomography capability. Both systems have participated in and successfully passed the requirements of formal DOE-sponsored intercomparison studies. The systems have measured approximately 1 to 100 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 a precision of 1 to 4% and an accuracy of better than 30% of the true values of known standards for all drums measured.

## 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 drums 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.

Several different tests using various waste matrices and known masses of plutonium (Pu) have been designed by DOE to evaluate the performance of NDA systems. LLNL and BIR are participating in the DOE Performance Demonstration Program (PDP), the Rapid Commercialization Initiative (RCI), and the Capability Evaluation Project (CEP) studies. Understanding these programs and performing well is critical to becoming certified to assay wastes for shipment and disposal.

The PDP consists of a series of tests conducted on a regular frequency to evaluate the capability for nondestructive assay of TRU waste throughout the Department of Energy complex. These evaluation cycles provide an objective measure of the reliability of measurements performed with TRU waste characterization systems. The PDP is designed to help the Carlsbad Area Office evaluate and approve the measurement facilities supplying services for the characterization of Waste Isolation Pilot Plant (WIPP) TRU waste.

The CEP evaluation was designed to establish nondestructive waste assay system technology capability and deficiency determinations and to facilitate resource allocation to areas requiring development. The evaluation was also intended to generate information and data to end-user EM30 Waste Management programs to support appropriate selection and application of a given nondestructive assay technology to the various waste streams.

LLNL is supporting BIR in a Programmatic Research and Development Agreement and a Rapid Commercialization Initiative with the Department of Energy, EM-50. The agreement requires BIR to develop information sufficient to establish compliance with applicable National TRU Program waste characterization requirements and associated quality assurance performance criteria. This effort requires an objective demonstration of the BIR waste characterization system. As with the CEP project, the goal of the RCI test project is to provide a mechanism from which evidence can be derived to substantiate nondestructive assay capability and utility statements for the BIR system.

## 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 drum by an external radioactive source(s) and the second is a passive measurement of the radioactive source(s) within the drum. The results of these two measurements are combined to produce an attenuation corrected gamma-ray assay of the drum. 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 and BIR systems is described.

### 2.1 Isotopic Measurements by Passive and Active CT

Currently there are two working A&PCT systems. One is the Isotopic Measurements by Passive and Active Computed Tomography (IMPACT) located at LLNL, the other is within a mobile Waste Inspection Tomography (WIT) trailer being developed in collaboration with BIR.[2] The former, IMPACT, has been tested on simple cases, e.g., well characterized radioactive sources without attenuators, within uniform attenuators, and within mock heterogeneous-waste drums. These sources do not have lump or clumping attributes. IMPACT has also been used to assay some LLNL real waste drums containing 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 real waste drums. At LLNL, WIT characterized drums that contained smaller containers with solidified chemical wastes; at the Rocky Flats Environmental Technology Site (RFETS) in Colorado, WIT measured drums with low-density combustible matrices. At the Idaho National Engineering and Environmental Laboratory (INEL), WIT characterized graphite-, glass-, and metal-matrix drums, lead-lined drums with combustibles, and very dense sludge drums. The Pu mass within these drums ranged from approximately 1 to 100 g.

The IMPACT and WIT A&PCT systems both use a single collimated aperture for a high-purity Ge detector (coaxial, >90% relative efficiency) and a $^{166m}$Ho external radioactive source. The active data acquisition measures the attenuated gamma-ray spectrum emitted from a $^{166m}$Ho source. In active CT the data is obtained by discrete translation and rotation of the drum for each slice. Typically 14 translations (or ray sums) and 21 angles (or projections) are required per elevation (or slice). An entire drum requires 18 slices in which one slice is below the drum. Active CT provides quantitative attenuation maps of the waste matrix at any desired energy.

To obtain passive CT images, the $^{166m}$Ho source is shuttered; and the drum is scanned in a similar fashion used to obtain the active data. In the passive mode, the detector collects and records individual energy regions of interest (EROI) or the entire energy spectrum of gamma-rays emitted from within the drum. The passive measurements are the integrated radioisotopic activity, modified by one or a multiple of exponential attenuations along the path from a source position within the drum 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 drum.

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 drum scan. Thus, the active scans provide energy-specific attenuation data so that attenuation corrections can be made for each internal gamma-ray energy identified. To attain an absolute assay measurement, the A&PCT systems are calibrated once on an absolute scale by simple measurements of one or more NIST-traceable, 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 Maximum Likelihood Expectation Maximization (MLEM) 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, which improves the detector-aperture aspect ratio (aperture length/aperture width). Septa are thin but highly attenuating dividing-plates that run the length of the collimator. The detector collimator length can be reduced when using septa, which is a benefit in the limited space of a trailer-based system like WIT.

The 3D MLEM code was adapted from a code specifically designed for medical imaging geometries, which have much larger detector-aperture aspect ratios. There are several assumptions in the code, which are valid for the medical imaging case, but not for the drum-assay problem. Thus, we have entirely re-written the reconstruction software based on the physics and geometry of the drum NDA problem.

In addition, 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.[3] 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 simplifies the gamma-ray spectroscopy analyses that are required in an absolute assay. The passive measurement localizes the activity of interest into small-sized (~5 cm on a side) volume elements (voxels). 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 assay results.

We have also 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 drums with low radioactive content.

Analysis of the spectroscopic data includes: (1) summing the data from each slice; and (2) summing the data for the entire drum to determine the isotopic ratios for: $^{238}Pu/^{239}Pu$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$, and $^{241}Am/^{239}Pu$; and (3) inspecting the $^{241}Pu/^{239}Pu$ ratio for each slice to evaluate isotopic homogeneity. Except for the $^{238}Pu/^{239}Pu$ 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).[4] The QAPP identifies the quality of data necessary to meet the specific data quality objectives associated with the DOE's Waste Isolation Pilot Plant 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).

The PDP, RCI, and CEP performance tests are designed to help ensure compliance with the QAPP QAOs.[5] These performance measures are blind tests that provide an objective measure of the reliability and performance of NDA systems. Each of these tests consist of a set of 208-L surrogate drums and a configuration that includes provisions to install a simulated waste matrix in addition to allowing for the convenient external introduction and precise positioning of standards (radioactive sources) within the drum volume. The RCI and CEP tests also included real waste drums. The PDP requires 6 replicate measurements per drum evaluated, while the RCI and CEP require 8 replicate measurements per drum. Between each measurement the drum must be completely removed from the NDA system.

## 2.5 IMPACT and WIT Performance

The IMPACT scanner has formally participated in and passed the PDP cycle-3 performance measure. The results are shown in Table 1 and Figures 1 and 2. This test consisted of a combustible-matrix drum with PDP standards resulting in a total of 66.76 g of $^{239}Pu$; and a glass-matrix drum with 4 PDP standards having a total of 91.9 g of $^{239}Pu$.[6]

The WIT scanner has formally participated in and passed the RCI, PDP cycle-4, and the CEP performance tests. Here we only report the surrogate results as shown in Table 1 and

Figures 1 and 2.[7,8] The RCI surrogate drums contained glass, combustible, and metal matrices with 2.3, 1.0, and 0.8 g of $^{239}$Pu, respectively. The PDP performance tests used drums containing combustible and zero waste matrices loaded with 6.17 and 91.9 g of $^{239}$Pu, respectively. The CEP performance test included surrogate drums with metals, Molten Salt Extraction (MSE), Raschig rings, and sludge loaded with 3.23, 67.4, 0.91, and 4.39 g of $^{239}$Pu, respectively.

Figure 1 shows the system accuracy for each of these performance tests as a function of percent recovery (%R). The vertical bar in the figure represents the allowed error in the accuracy as defined by the QAPP. The ball on each bar represents the measured recovery for each examination. Figure 2 shows the results of precision for these performance tests. The vertical bar on this graph represents the allowed error in precision and the ball represents the measured precision as a function of percent relative standard deviation (%RSD).

Table 1**:** ***IMPACT and WIT blind test results for the assay of surrogate drums***

| **Drum** | | | **Measurement** | | **QAPP Acceptance Criteria** | | |
|---|---|---|---|---|---|---|---|
| **[1]Test System** | **[2]Rep #/ grams $^{239}$Pu** | **Sample ID (Matrix)** | **%R PASS/ FAIL** | **%RSD PASS/ FAIL** | **Lower Bias %R** | **Upper Bias %R** | **Precision (MAX) %RSD** |
| IMPACT PDP-3 | 6 (66.76g) | Drum 003 (Comb.) | 66.41 ***PASS*** | 1.98 ***PASS*** | 52.08 | 147.92 | 6 |
| IMPACT PDP-3 | 6 (91.9g) | Drum 004 (Glass) | 70.39 ***PASS*** | 1.93 ***PASS*** | 52.03 | 147.97 | 6 |
| WIT RCI | 8 (2.3g) | 1SG (Glass) | 141.4 ***PASS*** | 3.89 ***PASS*** | 32.2 | 197.8 | 14 |
| WIT RCI | 8 (1.0g) | 2-SG (Comb.) | 162.5 ***PASS*** | 4.15 ***PASS*** | 32.5 | 197.5 | 14 |
| WIT RCI | 8 (0.8g) | 3-SG (Metals) | 179.6 ***PASS*** | 4.15 ***PASS*** | 33.5 | 196.5 | 14 |
| WIT PDP-4 | 6 (6.17g) | Drum 003 (Comb.) | 109.83 ***PASS*** | 2.95 ***PASS*** | 33.09 | 196.91 | 12 |
| WIT PDP-4 | 6 (91.9g) | Drum 001 (Zero) | 99.06 ***PASS*** | 1.54 ***PASS*** | 76.62 | 123.38 | 3.5 |
| WIT CEP | 8 (3.23g) | RF-20 (Metals) | 148.75 ***PASS*** | 1.2 ***PASS*** | 31 | 198.9 | 14 |
| WIT CEP | 8 (67.4g) | SG-6 (MSE Salt) | 70.7 ***PASS*** | 1.1 ***PASS*** | 50.9 | 149.1 | 7 |
| WIT CEP | 8 (0.91g) | SG-9 Raschig | 154.9 ***PASS*** | 4.2 ***PASS*** | 33.5 | 196.5 | 14 |
| WIT CEP | 8 (4.39g) | RF-11 Sludge | 61.1 ***PASS*** | 12.8 ***PASS*** | 40.8 | 189.2 | 14 |

Note: 1. The RCI and CEP were both scored by DOE INEEL[8], the PDP was scored by DOE-CAO[6,7]. All data includes 6 (PDP) to 8 (RCI and CEP) replicates per drum. All tests were performed at INEEL.
2. Number of replicated scans on top and actual $^{239}$Pu content on bottom in brackets.

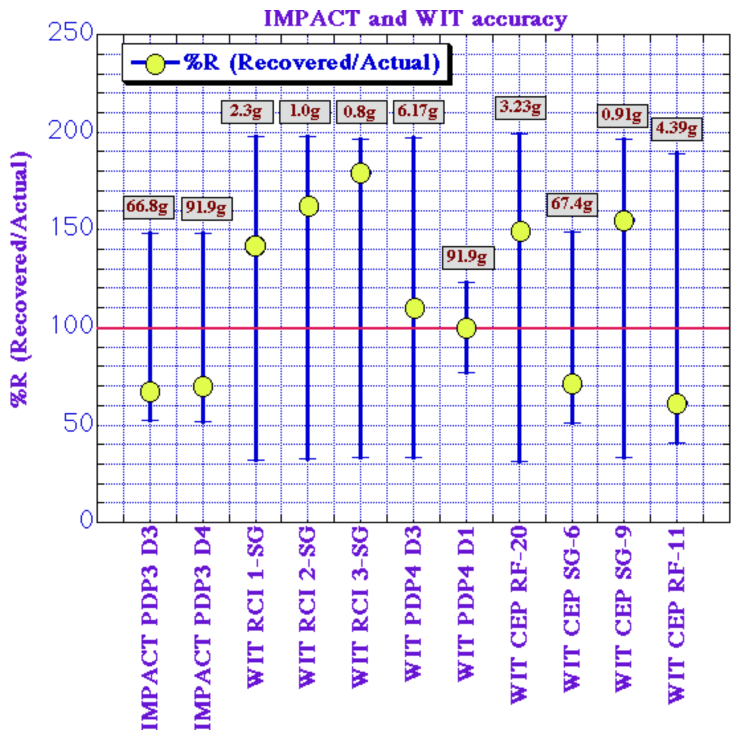


Figure 1: IMPACT and WIT scanner accuracy. The vertical bar represents allowed error in %R (recovery/actual). The ball represents system measured %R.

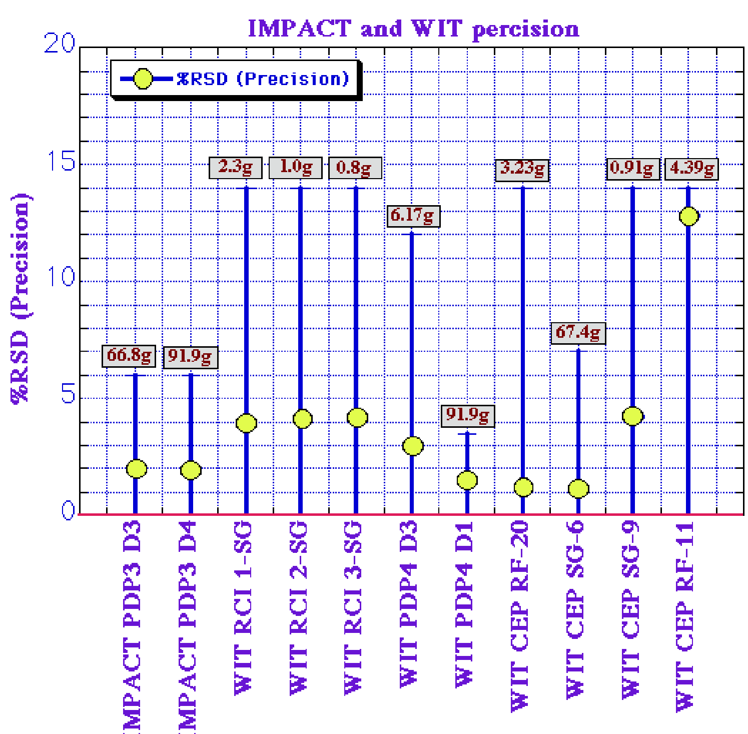


Figure 2: IMPACT and WIT scanner precision. The vertical bar represents allowed error in %RSD (relative standard deviation). The ball represents the system measured %RSD.

## 2.6 Summary of LLNL Developed A&PCT systems

LLNL and BIR have been involved in developing two A&PCT scanners. One is located at LLNL; the other is located within a mobile WIT trailer. These systems have been used to assay a wide range of radioactive waste within matrices ranging from combustibles to sludge with Pu content ranging from approximately 1-100 g. Every official performance measure that the IMPACT and WIT scanners have participated in has been successfully passed.

To increase system throughput, LLNL has completed a preliminary design for a multiple detector A&PCT system.[9] The WIT system is currently being upgraded to multiple detectors. New image reconstruction and assay codes are near completion and show promising results with respect to reducing our system biases. LLNL is also working on automating the isotopic analysis and developing a method for determining the systematic and total uncertainties.

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