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ACTIVE AND PASSIVE COMPUTED TOMOGRAPHY FOR NONDESTRUCTIVE ASSAY

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ABSTRACT

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 applying an active and passive tomographic technique (A&PCT) developed at the Lawrence Livermore National Laboratory (LLNL). The technique uses an external radioactive source and active tomography to map the attenuation within a waste barrel as a function of mono-energetic gamma-ray energy. Passive tomography is used to localize and identify specific radioactive waste within the same container. Reconstruction of the passive data using the attenuation maps at specific energies allows internal waste radioactivity to be corrected for any overlying heterogeneous materials, thus yielding an absolute assay of the waste activity.

LLNL and Bio-Imaging Research, Inc. have collaborated in a technology transfer effort to integrate an A&PCT assay system into a mobile waste characterization trailer. This mobile system has participated in and passed several formal DOE-sponsored performance demonstrations, tests and evaluations. The system is currently being upgraded with multiple detectors to improve throughput, automated gamma-ray analysis code to simplify the assay, and a new emission reconstruction code to improve accuracy.

INTRODUCTION

Gamma-ray safeguard measurements are traditionally performed using a segmented gamma scanning (SGS) system. SGS accuracy relies on the assumption that the sample matrix and the activity are both uniformly distributed for each segment that is measured. However, 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 heterogeneity. Imaging techniques that are better at measuring the spatial locations of sources and the matrix attenuation can reduce these errors. A computed tomography (CT) and nondestructive assay technology has been developed that can identify and quantify all detectable radionuclides transuranic isotopes in waste drums independent of the waste drum source and matrix distribution.[DEC96, MAR95, ROB95, ROB98]

The technology uses an external radioactive source to perform active CT (ACT) to simultaneously map on a volume-element by volume-element (voxel-by-voxel) basis any number of selected mono-energetic gamma-ray attenuation values. A second measurement, with the external source shuttered, uses a tomographic method to passively record the gamma-ray energy emitted from the drum on a voxel-by-voxel basis. The passive or PCT data are coupled to the ACT data to reconstruct attenuation-corrected images of specifically desired gamma rays emitted from a drum. From these data radioactive waste content is identified and quantitative TRU gram amounts of detected isotopes are determined. Thus, a Pu-equivalent mass, an alpha-Curie value and total specific power can be determined for low-level, TRU, and mixed-waste drums.

The A&PCT technology currently utilizes a single high purity germanium (HPGe) detector to collect both the ACT and PCT data. This technique is very simply calibrated by recording only one measurement of the gamma-ray emissions from a NIST-traceable source at a known location. Once calibrated, the A&PCT-HPGe measurements can be made of any drum with a known or unknown, homogeneous or heterogeneous waste matrix. No other information is required about drum content and no other calibration steps are necessary. In this technology ACT and PCT measurement times depend on the density of the waste matrix and activity levels of the resident TRU isotopes. In our single-HPGe detector systems the ACT and PCT measurement times varied from tens of hours each for drums containing dense sludge and/or matrices with a low gram TRU loading to several hours each for matrices with low-density dry combustibles and/or matrices with high activity levels. In our future multiple HPGe-detector A&PCT systems, these measurement times will decrease by factors of from 10 to 40 or more.

Two A&PCT systems were developed and tested. The first, Isotopic Measurements by Passive and Active Computed Tomography (IMPACT), is located at the LLNL and culminates a 10-year R&D effort to define the capabilities of this technology. The second A&PCT system, waste inspection technology (WIT), was developed over the past 5-years in cooperation with an industrial partner, Bio-Imaging Research, Inc., of Lincolnshire, Illinois.[BER95, BER97] LLNL is supporting BIR in a Programmatic Research and Development Agreement and a Rapid Commercialization Initiative with the Department of Energy, EM-50. The system was built into a mobile trailer along with additional non-destructive evaluation x-ray measurement modalities. This mobile laboratory provides a complete NDA and NDE capability that is transportable to any DOE waste site. Both systems employ radioactive transmission sources to enable mono-energetic ACT waste-matrix attenuation measurements; both use a HPGe detector for their ACT and PCT measurements; and both use a staging system to manipulate the drum or source-detector pair to enable the A&PCT data to be acquired. These efforts required an objective demonstration of both waste characterization systems.

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 Capability Evaluation Project (CEP), and the Rapid Commercialization Initiative (RCI) studies. 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. 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 WIT system.

The two A&PCT systems have successfully passed all of the DOE performance tests that they have participated in. Initial results of two performance demonstration program (PDP) blind test measurements for the LLNL system show it to have a low bias, but with precision at or below 1% for 15 replicate measurements. Initially, the LLNL system also gave high-biased results for drums with a low TRU loading; however, this appears to be a result of image reconstruction codes that improperly handled statistical zero counts. We have developed a new emission reconstruction code that appears to improve both the negative and positive biases. The WIT system participated in three different blind-test measurement programs, the rapid commercialization initiative (RCI); the capability evaluation project (CEP); and the PDP. The results from these examinations show that the WIT system does not appear to have a low bias for any type of waste matrix. However, the WIT system does appears to have a high bias for low-activity drums, which, like the LLNL system, is related to improperly handling statistical zero counts. This bias would be reduced if our new reconstruction method were used in the assay.

Present work underway for the WIT system includes an upgrade from one HPGe detector to six. These six detectors coupled with an improved PCT scanning modality will enable both the ACT and PCT measurements to be made from 10 times faster (for low activity in high attenuating matrices) to 40 times faster (for high activity in a low density matrix, continuously scanned). We are also developing an improved automated gamma-ray, spectroscopy analysis code that will enable almost all TRU isotopes, as well as other infrequently occurring waste radioactivity, to be identified. Finally, we are implementing a new emission reconstruction algorithm that shows promise in reducing bias due to short integration times or low counting statistics. Performing assays with shorter integration times will increase the system throughput.

A&PCT Technology

In order to assay an unknown waste drum using gamma-ray active and passive CT four processes are required. These processes are: (1) Gamma-ray spectroscopy data acquisition in both the active and passive CT modes; (2) Analysis of the gamma-ray spectra acquired in both the active and passive modes; (3) Reconstruction of both the active and passive images; and (4) Self-attenuation correction. It is useful to describe briefly the principles and issues associated with each of these processes to better understand the challenges of the NDA of waste drums.

A&PCT Data Acquisition

Our active and passive computed tomography technology employs a scanner that uses high purity germanium (HPGe) detectors and their associated electronics.[DEB88, MAR91, ROB91 & ROB94] It differs from conventional transmission CT scanners in that it discriminates between photons of different energies. The quantity that is reconstructed in active CT is the linear attenuation coefficient (μ) value for some volume element, or voxel, at location x, y, and z within a drum. The voxel size and clarity are defined by scan and image reconstruction parameters. For a waste drum, the attenuation due to its contents is accurately measured in three dimensions and displayed as a sequence of two-dimensional images at different z planes (or elevations) of the drum or as a 3-D volume or surface rendered image. Active CT does not identify any isotope or measure the source strengths of any radioactivity within a waste drum.

Passive CT is used to measure and determine both the identity and the strength of radioisotope sources within a drum. The quantity that is reconstructed in passive or single-photon-emitted CT (sometimes called SPECT) imaging is the counts measured in disintegration (d) per unit volume and time of a source within a waste drum. Therefore, a single-photon-emitted measurement for each detector position (ray sum) is the integrated radioisotope activity, modified by one or a multiple of exponential attenuation values, along the path from a source position within a drum to the detector. The function that is imaged for passive CT is the counts corrected for matrix attenuation at one or more energies for all detectable radioisotopes within a drum. The gamma-ray spectrometry detection equipment collects the entire energy spectrum at each integration point and characteristic peaks within the energy spectrum are used to identify radioisotopes.

Data Acquisition Hardware

The hardware required to perform both active and passive modes consist of four principle components. These are:

(1) *Active source*: used to acquire the active attenuation map,

(2) *Energy discriminating detector*: used in both the active and passive modes,

(3) *Staging system*: for manipulating the drum or source and,

(4) *Computer system*: used to acquire data and control staging.

The active source is a radioactive gamma-ray source that typically provides multiple mono-energetic peaks from 180-keV to about 1.3 MeV. These gamma-ray peaks do not need to be the same energy as the gamma rays emitted from within a waste container because interpolation between any two nearest neighboring active peaks or extrapolation can be performed. A radioactive source of low activity (10 mCi or less) is typically used because the HPGe detector has a relatively large-collimated aperture and is very efficient. The A&PCT method requires a high-energy resolution detector for identification and quantification of all detectable radionuclides. Therefore, we use a high-energy resolution and high efficient germanium detector with associated spectroscopy electronics.

Two other important components of the A&PCT scanner are the collimators for both the active source and detector. The apertures in these collimators are square. The detector-collimator aperture defines the spatial resolution (voxel size) of the active-attenuation CT image and the corrected-passive CT image. The source collimator is used to minimize excess radiation. This ensures better data by reducing scatter, provides a safer operating environment, and eliminates any cross transmissions when multiple transmission sources are used with multiple detectors.

Through simulation we studied the trade-off between spatial resolution and signal-to-noise.[KET95] Further, an optimum system design is dependent on expected emission source distribution and activity. Results of our simulation work revealed that A&PCT systems with collimated square apertures from 2.5 to 7.5 cm on a side will perform best. In order to ensure that the detector fills the entire aperture area we need a large diameter detector. The largest diameter detectors with high counting efficiency are about 8.2 cm in diameter. A large detector aperture reduces the assay time in two ways. First, a larger aperture utilizes more detector surface area providing a larger solid angle; second, it yields the advantage of fewer measurement positions (less sampling). Finally, we found that the aspect ratio (aperture length divided by aperture width) performs best if it is in the range of 5:1 to 10:1.[KET95] The smaller the aperture aspect ratio, the closer the detector will be to the waste drum; hence, the higher the counting rate because of the larger solid angle subtended. It is important to note that this has to be traded off with the spatial resolution required to get accurate assay results. For example, an improvement of a factor of two in spatial resolution in all dimensions requires a factor of eight increases in measurement time.

Data are acquired by either manipulating the drum, or manipulating the drum and source/detector pair. If only the drum is manipulated, the staging system must be capable of translating, elevating, and rotating the drum. For other cases, the source/detector pair may translate and/or elevate instead of the drum. Typically, for a single detector A&PCT system, either the waste drum or the source/detector pair is discretely translated by a distance equal to the detector collimator's horizontal aperture dimension. For a single-detector system, the translation is performed for all ray sums required. If there were a sufficient number of detectors to cover a full transverse section of the drum, there would be no need to translate the drum. Thus, only drum rotation and elevation would be required for an assay.

The computer systems are used to control the staging system, acquire data from the spectroscopy electronics, perform pre-processing functions, reconstruct data, and determine the final assay. Data acquisition and digitizing subroutines depend on the detector system being used in the CT scanner. We acquire data from a single high-purity germanium detector where the sum of all interactions within the HPGe detector is digitized. The magnitude of the digitized signal is related to the energy of the photon interactions detected.

Each individual CT scanner dictates specific preprocessing methods and the number of preprocessing steps required as a result of the differing physical characteristics (i.e., source and detector systems used). The main preprocessing procedure consists of calculating the ray sums from the raw gamma-ray counts. Incident and transmitted counts are used to calculate ACT ray sums. These ray sums are used as input to the ACT image reconstruction codes. For PCT the emitted counts for all ray sums are combined with the attenuation images to produce attenuation corrected PCT images of photons per unit time per voxel. These images are used to obtain a specific isotopic activity and mass.

There are three important properties in acquiring both active and passive ray sums. First, the geometry of the ray paths (i.e., the source and detector positions in the object coordinate system) must be completely known. Second, at any given geometric position, the incident and transmitted counts must be accurately measured and recorded during ACT, and the emitted counts for a specific energy gamma ray or the entire spectrum must be accurately measured and recorded during passive CT. And third, for multiple-HPGe detector scanners, all preprocessed detector responses to a given energy and intensity must be identical; therefore, the responses from multiple HPGe detectors must be normalized to one value for all of the detectors.

Data Acquisition Modes and Protocols

We have two main protocols or modes of operation. These are collimated, gamma-ray scanning (CGS) and A&PCT. The CGS protocol is used to quickly determine the location of radioactivity with respect to height within a drum and to determine the data acquisition scan parameters (e.g., number of slices and scan time) required. The A&PCT protocol is used to accurately assay the radioactivity within a waste drum. Both the CGS and the A&PCT protocols employ the active and passive modes of drum scanning.

In the CGS protocol, gamma-ray data is integrated while the drum is continuously rotated for each slice in both the active and passive scan modes. A full drum requires 18 slices. In the active mode the transmission source is opened and data for specific energy regions of interest (EROI) are obtained. In the passive mode, the transmission source is shuttered and selected EROIs or the entire gamma-ray spectrum is recorded.

In the A&PCT protocol a set of transverse ray sums at one angle (a projection) are acquired at all translational positions until the drum diameter has been traversed. After each projection is acquired, the drum is rotated slightly for the next projection acquisition. In the active mode, projections are acquired over 180°, and in the passive mode they are acquired over 360°. Once a full set of projections is acquired, the drum or the source-detector pair is elevated by a distance equal to the vertical dimension of the detector's collimator aperture. This vertical dimension is the slice thickness. For a full drum, 18 slices are acquired for both the active and passive modes. All of the slice projections are used to reconstruct an image of the waste drum's attenuation (ACT) and emissions (PCT).

During an ACT data acquisition drum scan, EROIs are set for each of the major peaks of the transmission source since these are known. Data are collected from each EROI simultaneously for each ray sum acquired. The active data that are saved and used in the reconstruction process are the net counts, which are the gross or total counts in the energy peak for each EROI selected minus background radiation.

In passive drum scans, the entire energy spectrum is acquired for each ray sum. This differs from the active mode because the energy peaks emitted from within a waste drum are unknown. All of the emission spectra are used to evaluate the isotopic makeup of the waste drum's content.

Gamma-ray Spectroscopy Analysis

The A&PCT data sets consist of hundreds of high-energy resolution gamma-ray spectra, one for each ray sum acquired. Generally, the statistics in the spectra for each individual ray sum are poor, but they can be summed to produce better quality spectra that can be analyzed by traditional gamma-ray spectroscopic analysis codes. Such an analysis can determine most of the radioactive isotopes present. This includes isotopes important to transuranic waste characterization, and isotopes that may be of interest to neutron-based NDA technologies or gamma-rays that might interfere with the analysis.

The analyses of the passive CT gamma-ray spectra also yield the mass ratios of the important plutonium isotopes. The A&PCT method could, in principle, determine a mass for each of these isotopes by reconstructing the ray-sum data sets (sinograms) that represent their characteristic gamma-rays in the spectrum. However, that is rarely possible due to their poor statistics. Instead, the A&PCT assay only needs to determine the mass of the most abundant TRU isotope, which is usually but not always ^{239}Pu. Analysis of the spectra is used to determine other isotopic ratios. This information is used to calculate the thermal power, the total alpha-Curie activity, and the fissile gram equivalent necessary for the characterization of the waste.

The A&PCT data acquisition code creates spectra in a binary format. We have developed a computer code that automatically processes each of the ray-sum spectra. The isotopic analysis code is described in detail by Clark et al. in [CLA98]. Our gamma-ray analysis code reads these spectra and builds a summed binary spectrum as well as an ASCII spectrum data file. The actual spectral analysis is performed on the ASCII file, which is saved for future analysis. In addition to forming the summed spectrum, the code also analyzes each ray sum for all of the energy regions of interest (EROIs) from a user-defined table. The results of this analysis provide gross and background counts from each EROI for each ray-sum. The resulting sinograms are then stored in a binary format used by the image reconstruction and assay code.

Currently, we examine each summed gamma-ray spectrum for the following TRU isotopic mass ratios: ^{238}Pu/^{239}Pu, ^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu, ^{235}U/^{239}Pu, and ^{241}Am/^{239}Pu. These ratio pairs are determined by analyzing closely spaced multiplets of gamma-ray lines emitted from these isotopes in seven regions of the gamma-ray spectrum:

(1) The region between 120 and 135 keV is analyzed to determine the $^{241}Am/^{239}Pu$ ratio. This region contains six peaks, but the analysis is determined by the ratio of the ^{239}Pu peak at 129.3 keV and the peak at 125 keV from ^{241}Am. However, all peaks in this region must be included in the fit to obtain a good analysis. This region gives a good indication of any excess americium in a drum. This has been demonstrated using data from the Stored Waste Experimental Pilot Plant (SWEPP) located at the Idaho National Engineering and Environmental Laboratory (INEEL).

(2) The region from 135 to 155 keV is used to determine the $^{241}Pu/^{239}Pu$ ratio by comparing the 148.5-keV (^{241}Pu) to the 144.2- keV transition of ^{239}Pu. The presence of ^{235}U and its transition at 143 keV can complicate this analysis. Similarly, the 152-keV peak of ^{238}Pu can also be analyzed; however, for weapons grade Pu this is a rather weak transition which greatly limits the accuracy of the derived $^{238}Pu/^{239}Pu$ ratio.

(3) The analysis of the 160-keV region should give the best information about the $^{240}Pu/^{239}Pu$ ratio. The doublet of strong transitions from these two isotopes at 160 keV can be a clear indication of the degree of "burn-up" in the plutonium. Unfortunately, this region is frequently contaminated by high Compton background in typical waste drum spectra, which limits the accuracy achievable.

(4) The 208-keV transition is very prominent in the gamma-ray spectra of the A&PCT systems. This transition is from the decay of ^{237}U to ^{237}Np, the daughter of ^{241}Am and granddaughter of ^{241}Pu. However, because of its higher specific activity, ^{241}Pu will dominate in most cases. This peak can be compared to the 203-keV peak of ^{239}Pu to determine the mass ratios. If ^{235}U is present, its 205-keV peak is compared to the 208-keV peak to obtain its relative abundance.

(5) The regions near 340 keV and at 375 keV give additional measurements of the $^{241}Pu/^{239}Pu$ and $^{241}Am/^{239}Pu$ ratios.

(6) The 414-keV ^{239}Pu transition is used in the PCT analysis. This region is also analyzed to look for possible contributions from ^{237}Np, which may be present in addition to the Pu isotopes.

(7) The region above 600 keV contains peaks from ^{239}Pu, ^{240}Pu, and ^{241}Am and can be very useful in some cases. The high energies of these transitions allow them to escape from highly attenuating waste matrices. However, branching ratios for these peaks are less than those at lower energy regions so the statistical quality of data for a low waste-drum gram loading is a problem.

The results from the analysis of these regions are then combined into a report of the mass ratios. Subsequently, the data are combined with the assay data for ^{239}Pu (or ^{235}U) to calculate the desired waste characterization parameters. This report lists the energies and intensities of the gamma rays found in a spectrum along with their identification when possible. Those peaks not found to agree with any listed in the table are flagged as unknowns. In the future, we intend to implement an option that if no isotopes from these three major elements (U, Pu, & Am) are found, the program will continue to search for other TRU isotopes. These may be isotopes that are identified as important to the neutron-based NDA techniques and to the WIPP WAC (e.g., ^{137}Cs, et al.).

Image Reconstruction and Assay Determination

While it seems simple enough to assay the total radioactivity within a large distributed volume (a nuclear waste drum) by measuring the emitted radiation, the central difficulty is that an accurate absolute assay is impossible unless the emerging measured radiation can be corrected for the matrix attenuation it suffers. This correction requires knowledge of the spatial distribution and density of both emitters and absorbers throughout the volume. An accurate assay will necessarily involve a complete determination of the three-dimensional (3-D) structure of all radioisotopes present even though the original problem posed by the regulations requires only one number, the total radioactivity quoted as Pu-effective grams, contained within a waste drum.

The active CT data is acquired and reconstructed using the robust 2-D filtered backprojection method described elsewhere.[BUD79] The resultant 2-D ACT slice data at a particular energy is merged into a 3-D array of linear attenuation values before it is submitted with the passive count data to the passive CT image reconstruction and assay algorithm. The passive reconstruction algorithms are described in detail by Jackson et al. in [JAC98].

In order to assay the 3-D structure of a waste drum, it is divided into a set of voxels. The number of counts for all detectable radioisotopes is determined for each voxel. The sum of the counts from all of the voxels determines the non-destructive assay of the drum. The passive CT image reconstruction and assay starts by defining a vector **p**, where the elements in the vector represents the number of counts from each voxel. The vector **p** is unknown and is the desired solution. The emitted radiation is measured at a series of detector positions, which constitute the ray sums in a passive CT scan. The, vector $\boldsymbol{g}_\gamma$ is defined, where each element in the vector is the measured radiation at a given detector position. The vector $\boldsymbol{g}_\gamma$ is the sinogram data measured in the passive CT scan.

The relation between the vectors **p** and $\boldsymbol{g}_\gamma$ can be defined mathematically as

$$\boldsymbol{g}_\gamma = \mathbf{Ap} \,. \tag{1}$$

The system matrix, **A**, represents and incorporates the effects of the system's geometry and the attenuation image determined from the active CT scan. The matrix **A** is too large to be effectively inverted to solve for **p**. Therefore, **p** must be determined from $\boldsymbol{g}_\gamma$ and **A** using an iterative optimization technique.

The iterative A&PCT reconstruction codes are divided into an optimization and a model code. Figure 1 shows a conceptual design of the code. The optimizer consists of a cost function and a minimizer algorithm. The cost function calculates a scalar by comparison of the measured, g_γ, to the calculated, $\hat{g}_\gamma$, passive sinogram. The minimizer section searches for the next best solution, $\hat{p}$. The model code determines the calculated passive sinogram from the current solution determined by the minimizer. The optimizer determines when a solution is acceptable, producing the final $\hat{p}$.

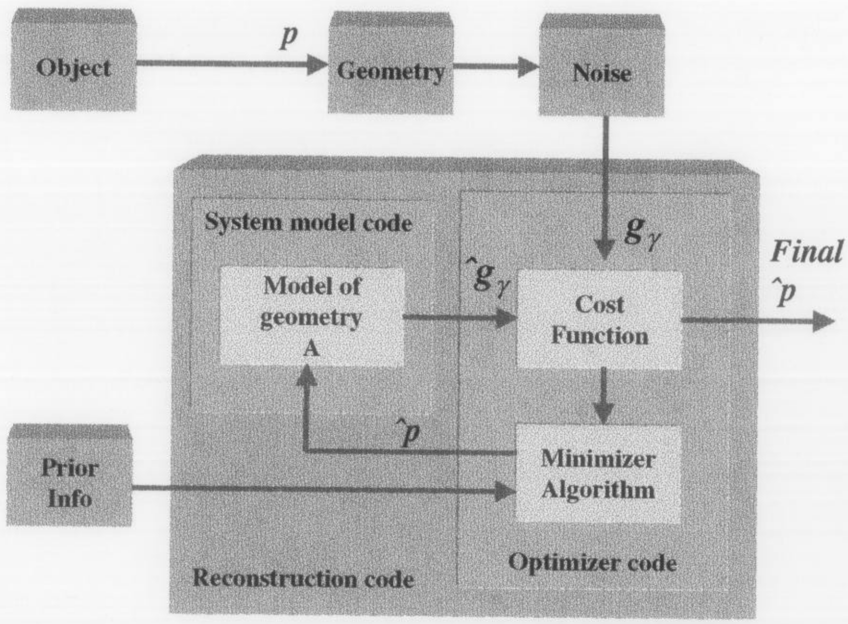

Figure 1 Conceptual design of the passive image reconstruction code.

Model Code

The purpose of the model code is to calculate the values in the system matrix **A.** The values are determined by using the correct CT scanner geometry and physics to relate the contribution of the emission from each voxel to each detector. The effect of the measured attenuation from the active CT scan is also incorporated. In the recent past, A&PCT has used two methods to determine the system matrix, they are known as UCSF and APCT.

The first model code that was used was UCSF. There were several assumptions in the UCSF model code that were valid for the medical imaging case, but not for the drum-imaging problem. A detector collimated to receive radiation from emitting radioactive source measures photon counts decreased as the square of the distance between the emitter and the detector, ($1/R^2$), and decreased exponentially by absorption along the line of sight. In addition, the collimation of high-energy photons is normally done with collimators rather than lenses, and the collimator aperture will have a diverging acceptance angle with a width or area proportional to distance from the source to the detector. Finite spatial resolution for both the active and passive measurements affects the resolving power of the system to measure the waste drum wall, waste items within the drum including either emitters or absorbers with sharp boundaries, and the radioactive assay accuracy of all detectable emission sources.

The non-linear effects in the system, in particular the $1/R^2$ fall-off, the exponential attenuation, and the spatially varying collimator response imply that reconstruction or inversion will not be a linear operation. Although emission tomography is not a new idea, most current applications do not fully account for these non-linear effects and do not provide for accurate quantitative measurements. In particular, in medical imaging, reconstructions are generally done with an inverse Radon transform. This implies the assumptions that the internal attenuation is not too strong, and that the distance through the object, the patient, is not too great so that the divergence of the collimators and the $1/R^2$ fall-off are also small. In medical imaging, it is possible to achieve a reconstruction of sufficient qualitative accuracy to be clinically useful in diagnosis. However, these effects cannot be ignored in quantitative imaging. We have developed a new model code, referred to as APCT, that takes into account these issues.[JAC98] In addition, the attenuation line integral is calculated with finer sampling or spatial resolution than that used in the UCSF model code.

Optimization Code

Iterative methods for image reconstruction generally proceed by successive minimization or maximization of a cost function. For each situation a cost function must be determined and an iterative method for determining the maximum or minimum (optimization) of the function must be selected.

The cost function is usually a likelihood or log-likelihood function. In a method where a model exists for describing accurately the system, including non-linear effects, the likelihood function is often the squared difference between the model's predicted data and observed data. This is referred to as a least-squared likelihood function. Several numerical algorithms exist for least-squares minimization. Most of these algorithms proceed by moving down the gradient of the likelihood function. Examples of these algorithms are steepest descent, conjugate directions, or successive projections. The last is termed the Algebraic Reconstruction Technique.

However, in the waste drum situation, where the system being modeled is the gamma rays produced by the decay of radioactive materials, the process is best represented by a Poisson probability distribution. This is shown where the probability on n counts is *Pr[n]*

$$\Pr[n] = \frac{1}{n!} z^n \exp\{-z\} \ . \tag{2}$$

The parameter z is the mean number of counts. In our waste drum case, it is the measured gross or background counts at each detector ray-sum position.

An iterative reconstruction for determining the maximum likelihood for a Poisson distribution has been developed and used in the medical community.[SHE82] A version of this solution called Maximum Likelihood Expectation Maximization (MLEM) was developed in collaboration with UCSF for waste drum analysis.[BRO95] However, this MLEM method only handles one Poisson distribution signal. In our case there are two

Poisson distribution signals, the gross and background counts. The MLEM algorithm is not flexible enough to incorporate these two Poisson distributed signals correctly.

We have developed a new likelihood function based on the joint probability density functions for the gross peak and the background for each ray sum measurement in a statistically correct fashion.[GOO97] This avoids any physically unrealistic "negative counts" that must be set to zero in other estimation approaches to this problem, such as MLEM. The result of zeroing negative counts can bias assay estimates (i.e., results). This new method avoids this problem. The likelihood function is minimized by a novel constrained conjugate gradient (CCG) algorithm[GOO93] that permits constraints on the estimates (such as non-negativity) at each voxel and uses a bending line-search technique to speed convergence.

Conventional techniques allow only a single variable per iteration to attain a bound, whereas the bending line search allows multiple variables per iteration to attain bounds. This search method is rapid and efficient. Given the dimensionality of our problem, this is a major advantage over the conventional techniques. This algorithm has been applied with great success to a variety of practical problems. These include deconvolution [GOO93], speckle interferometry [GOO93a], limited view x-ray tomography [KOL94, HAD95], and crystallography [SOM95] at LLNL, and pulsed photothermal radiometry [MIL95, MIL96, MIL96a, & MIL96b] at the Beckman Laser Institute and Medical Clinic of the UC Irvine Medical School.

Code Development History

In order to determine the optimal method of image reconstruction and assay, our A&PCT codes have undergone a series of improvements. We have progressed from parallel-beam through fan-beam to cone-beam geometric methods to reconstruct the passive CT data. The parallel-beam geometry method used an iterative, steepest-descent, weighted least-squares technique [HUE77] to reconstruct PCT images with attenuation corrections from the ACT image.[MAR91a] The fan-beam or 2-D work is described by Roberson et al. in [ROB94]. Here, we only describe our development efforts for the 3-D methods. A schematic diagram of the 3-D development is summarized in Figure 2, and each method is described below.

UCSF-MLEM

The first 3-D model code used in our A&PCT studies was developed in collaboration with UCSF. It was adapted from a UCSF code specifically designed for medical imaging geometries, and we developed a 3-D model and an MLEM optimization method, which is referred to as UCSF-MLEM[1]. The line integral used to determine the attenuation is calculated on a voxel-by-voxel basis. The system matrix is essentially recalculated each time through the optimizer, in this case MLEM. It can also only use the MLEM optimization technique that it was originally built with. This code however does allow for the use of a measured response function. Details of this code are provided by Brown, et al.[BRO95, TAN98]

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[1] The nomenclature used in the text is to name the passive image reconstruction and assay codes by a hyphenation of the model and optimization code names.

APCT-MLEM

In the APCT model code the system matrix is calculated once and stored. This makes the code more modular and allows other optimization techniques to be used with the same model code. Several combinations of the A&PCT model codes with different optimization codes have been developed. These are APCT-MLEM; APCT-CCG; and APCT-CCG with measured response function. The APCT-MLEM code uses the new model code in conjunction with the MLEM optimization code.

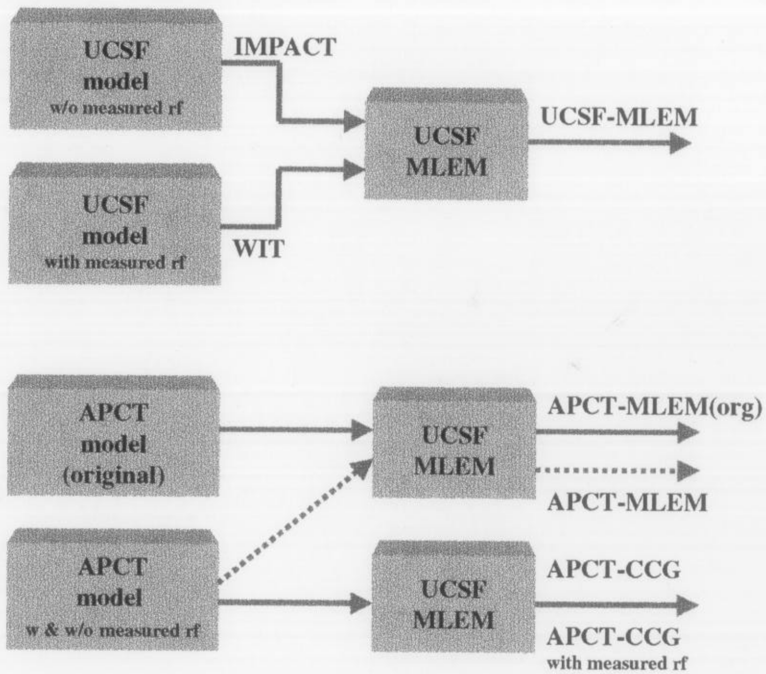

Figure 2 A schematic diagram that summarizes the different 3D-image reconstruction and assay codes developed. "rf" refers to the response function of the detector and its collimator.

APCT-CCG

The APCT-CCG code is a combination of the APCT model code with the CCG optimization algorithm with a two-measurement Poisson-based likelihood function. This code uses only a calculated response function.

APCT-CCG with Measured Response Function

The APCT-CCG with measured response function code is the same as the APCT-CCG code except it allows the user to choose to use a calculated or a measured point source (impulse) response function, or to calculate the response function from the data. This allows us to use one image reconstruction and assay code for both the WIT and IMPACT data.

Self-Attenuation Correction

Transuranic materials that are in a physical form other than difuse distributions, e.g., greater than 500-μm diameter particle size commonly referred to as lumps, are strongly self attenuating for gamma-rays typically used to determine isotopic and radionuclidic information. Because of this, NDA techniques are susceptible to self-attenuation biases when the TRU material is in dense accumulations, agglomerations and/or large clumps. As an example, consider a sphere of Pu metal with a diameter, *D*. The fraction, *F*, of photons with energy, *E*, which can escape from such a sphere, is given by

$$F(D,E) = \frac{3}{X}\left\{1 - \frac{2}{X^2} + \left[\frac{2}{X} + \frac{2}{X^2}\right]e^{X}\right\}, \tag{3}$$

where $X = \mu(E) \cdot D$ and $\mu(E)$ is the linear attenuation coefficient for the material at energy *E*.

As can be seen from this equation the escape fraction is strongly dependent on the absorption coefficient, that is a function of the photon energy. For 129 keV, less than 20% of such photons escape from a 1-mm diameter sphere of Pu, whereas almost 90% of the 414-keV gamma rays would escape. This strong energy dependence can be used to determine if there is a self-attenuation problem with the assay, and it can also be used to estimate the magnitude of this bias. The coarse size of the A&PCT volume elements do not allow a direct observation and/or correction of this form of attenuation.

Our approach for correcting the A&PCT assay data for self-attenuation will be as follows. We will reconstruct emission images for the 203-, 345- and 414-keV gamma rays, and possibly the 129-keV gamma ray. Then we will calculate their corresponding Pu masses. If there is no self-attenuation problem, these assay values will be equal. However, self-attenuation could be inferred if the assay results strongly increase with photon energy. A first order correction for this could be obtained by finding a best fit for the particle size that would reproduce the observed energy dependence. One could then recalculate the assay values using a correction for the escape fractions.

REQUIREMENTS AND PERFORMANCE CRITERIA

An NDA waste assay system's utility is defined in terms of its ability to comply with the requirements and quality assurance objectives for nondestructive assay as delineated in the National TRU Program (NTP) Quality Assurance Program Plan (QAPP).[QAP96] The QAPP identifies the quality of data necessary to meet the specific data quality objectives associated with the Department of Energy's Waste Isolation Pilot Plant (WIPP) transuranic (TRU) waste characterization program. The primary parameter that must be determined is total TRU-alpha activity. The quality assurance objectives (QAOs) for precision (percent relative standard deviation–%RSD), accuracy (percent recovery–%R), minimum detectable concentration (MDC), completeness, and total bias are stated in the QAPP. These parameters must be demonstrated over the spectrum of waste-form configurations that the assay system is intended to characterize.

The performance of IMPACT and WIT/A&PCT are derived from data acquired through a blind testing process. The performance evaluation tests being derivatives of

QAPP QAOs support a direct interpretation of IMPACT and WIT/A&PCT performance results. Thus, the IMPACT and WIT/A&PCT performance measures can be readily interpreted relative to the National TRU Program QAPP requirements. The criteria of these various programs are listed next. LLNL and BIR have participated in and passed three official DOE performance tests for the IMPACT and WIT systems. These tests are the Performance Demonstration Program (PDP), Rapid Commercialization Initiative (RCI) and Capability Evaluation Project (CEP) performance tests.

Performance Demonstration Program

The QAPP requires the facilities intending to use NDA methods to generate data for the National TRU program participate in a PDP. The PDP program is designed as an independent quality assurance test to provide data that supports the overall QAPP compliance assessment process.[PDP97, MAZ97] Data generated during PDP test cycles is also considered in the overall QAPP compliance assessment process. The PDP consists of periodic tests conducted of both increasing matrix and source complexities to evaluate the capability of various technologies to properly characterize 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 the various NDA systems. The PDP test samples are comprised of 208-*L* drums configured with NIST traceable Working Reference Materials (WRMs). The PDP test on any given test sample requires six replicate measurements and removal of the drum between each replicate measurement.

Presently there are five drum matrices: air (no matrix), ethafoam[2], combustibles, glass, and inorganic sludge. Aluminum source insert fixtures are provided for each the insert-tube radii. NIST traceable WRMs are positioned at desired vertical locations within the insert fixtures. Several types of WRMs are used in the program, e.g., WG Pu, large particle WG Pu and enriched U. The initial WRMs for the first four cycles were 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). Currently five cycles have been completed. The IMPACT system passed cycle 2 informally and cycle 3 formally and the WIT/A&PCT system passed PDP cycle 3 and 4. The WIT system is currently participating in Cycle 5 of the PDP.

Capability Evaluation Project

As part of the MWFA characterization development strategy, a method to objectively evaluate the utility of waste-assay system technologies was implemented in conjunction with the Characterization Monitoring and Sensor Technology (CMST) crosscut area program. This evaluation was designed to support 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 EM-30 Waste Management programs to support the appropriate selection and application of a given nondestructive assay technology to the various waste streams.

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[2] This drum matrix is no longer used in the PDP.

The Capability Evaluation Project (CEP) was specified in a manner such that evidence is derived to substantiate nondestructive waste assay capability and utility statements as a function of waste type and/or characteristics. The waste types for which the evaluation is conducted are those contaminated with transuranic elements. The evaluation program was conducted at the INEEL Radioactive Waste Management Complex (RWMC) using actual waste forms currently in storage and carefully specified and constructed surrogates. To the extent RWMC waste form attributes approximate other site waste inventories, statements can also be made regarding system utility per the site of interest. The capability evaluation plan addressed the acquisition, compilation and reporting of performance data, thereby allowing a given agency a basis for an objective evaluation of NDA systems that participate. The evaluation was structured such that a statement regarding select INEEL RWMC waste forms can be composed relative to compliance potential for applicable National TRU Program requirements and criteria.

The test is designed to provide objective and unbiased data regarding the performance and associated capability of each participating mobile assay system to the MWFA, to the CMST, and to procurers of waste assay system services and technology holders. The test series consists of a combination of surrogate waste-form and actual waste-form test samples. The surrogate-type test samples allow an evaluation of assay system performance where the matrix and radioactive source constituents and configurations are accurately known. The actual waste-type test samples are the unique aspect of the CEP in that performance is assessed with respect to the actual waste forms and their associated configurations. The criteria used to evaluate assay system capability are founded in the NTP program QAPP, Section 9.0, Interim Change version, the Performance Demonstration Program Plan for Nondestructive Assay for the TRU Waste Characterization Program. The WIT/A&PCT system has participated in and passed the CEP performance test.

Rapid Commercialization Initiative Test

Bio-Imaging Research Inc. was engaged in a Program Research and Development Agreement (PRDA) 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. The performance evaluation parameters and criteria used in the RCI project are as indicated for the CEP. Similar to the CEP test project, the RCI test utilized test samples with configurations representative of a large population of waste types in inventory at the INEEL RWMC. The WIT/A&PCT system has participated in and passed the RCI performance test.

SUMMARY OF A&PCT PERFORMANCE

We have carried out numerous tests of the A&PCT technology on surrogate and real-TRU waste drums. Both the IMPACT and the WIT/A&PCT systems were calibrated, validated and used to measure real wastes at the LLNL. WIT/A&PCT was demonstrated, tested and evaluated at three other sites: (1) The Rocky Flats Environmental Technology Site (RFETS); (2) Two separate occasions at the INEEL; and (3) Most recently at the Nevada Test Site. Several different radioactive isotopes within actual waste drums have been detected while WIT was at LLNL, RFETS, INEEL and NTS. A summary of the isotopes detected is presented in Table 1.

Table 1 Radionuclides identified in waste by BIR WIT/A&PCT.

U & TRU	Other Actinides[a]	Other Radionuclides
^{235}U	^{208}Tl	^{22}Na-F(α,p)
^{238}U	^{219}Rn	^{40}K-bckgd.
^{237}Np[a]	^{223}Ra	^{60}Co-*fp*[c]
^{238}Pu[b]	^{227}Th	^{137}Cs-*fp*
^{239}Pu	^{243}Cm	^{211}Pb-bckgd.
^{240}Pu	^{249}Cf	^{214}Bi-bckgd.
^{241}Pu		
^{241}Am		

Notes: a. Primarily identified in LLNL waste drums at NTS, some were identified in drums at LLNL, RFETS and INEEL.
b. ^{238}Pu can be seen in samples enriched in this isotope, but most waste contains WGPu and its percentage abundance is too small to be detected in most waste drum gamma-ray spectra.
c. *fp* is fission product.

At LLNL, many of the drums contained multiple 4 and 20-liter (1 and 5-gallon) containers of solidified chemical radioactive wastes. The RFETS wastes were all low-density combustible matrices. At INEEL the wastes included both lead-lined and normal drums containing waste matrices of graphite, glass, metals, wet and dry combustibles and sludge. The plutonium loading of these drums ranged from ~1 to 100 grams of ^{239}Pu.

A summary of IMPACT's and WIT/A&PCT's performance results in terms of ^{239}Pu mass for the %R and %RSD parameters are shown in Table 2. All data presented in this section were obtained using the UCSF-MLEM image reconstruction and assay code. Also, we are only reporting the surrogate results here since the ^{239}Pu mass is precisely known and NIST traceable. The non-surrogate CEP and RCI waste drum data is excluded from this table simply because there is uncertainty associated with their ^{239}Pu mass. All reported performance measure values are based on the IMPACT and WIT/A&PCT image reconstruction and assay procedure using the 414-keV peak of ^{239}Pu.

Table 2 IMPACT and WIT blind test results for the assay of surrogate drums.

Drum			Measurement	
Test System[1]	Rep. # (grams ^{239}Pu)[2]	Sample ID (Matrix)	%R for ^{239}Pu mass	%RSD for ^{239}Pu mass
IMPACT PDP-3	6 (66.87)	Drum 003 (Comb.)	66.06	0.58
IMPACT PDP-3	6 (92.11)	Drum 004 (Glass)	71.21	0.84
WIT RCI	1 (2.22)	1-SG (Glass)	139.7	2.8[3]
WIT RCI	1 (0.933)	2-SG (Comb.)	149.9	2.7[4]
WIT RCI	1 (0.747)	3-SG (Metals)	174.0	2.7[4]
WIT PDP-4	6 (6.16)	Drum 003 (Comb.)	109.9	2.92
WIT PDP-4	6 (92.27)	Drum 001 (Zero)	98.73	2.14
WIT CEP	8 (46.0)	SG-6 (MSE Salt)	103.53	1.54
WIT CEP	8 (0.961)	SG-9 (Raschig)	145.96	2.67

Note: 1. The RCI and CEP were both scored by LMITCO INEEL and the PDP was scored by DOE-CAO. [PDP97b, PDP97c] All data include 6 (PDP) to 8 (RCI & CEP) replicates per drum for these tests. All WIT test measurements were performed at INEEL.
2. Number of replicate scans on top and in parentheses actual ^{239}Pu content in grams.
3. Determined from the IMPACT 15 replicate QAO test at 3.6 grams of WG Pu.
4. Acquired from the CEP test sample SG-9 at 0.961 ^{239}Pu.

Figure 3 shows the system accuracy (%R or recovery) for each of these performance tests as a function of gram loading. The thick line at 100% is the ideal assay value. Both the IMPACT and WIT A&PCT systems passed the performance tests that are shown in the figure. However, the IMPACT system appears to have a negative bias when assaying drums with a high gram loading. The WIT system appears to have a positive bias when evaluating drums with a low gram loading. This is apparently due to unrealistic "negative counts" that must be set to zero in the MLEM estimation code that we currently used in the emission reconstruction process. The result of zeroing negative counts can positively bias the assay estimates. The new CCG method described in the reconstruction section shows improvement over the MLEM code when evaluating drums with a low gram loading. The CCG method has also shown a 10% improvement in the low bias that is observed with the MLEM reconstruction. However, the WIT system appears to have no low bias associated with the evaluation of drums containing a high gram loading.

Figure 4 shows the precision (%RSD or percent relative standard deviation), of the IMPACT and WIT systems as a function of gram loading. The precision of the two A&PCT systems is excellent, especially for a high gram loading.

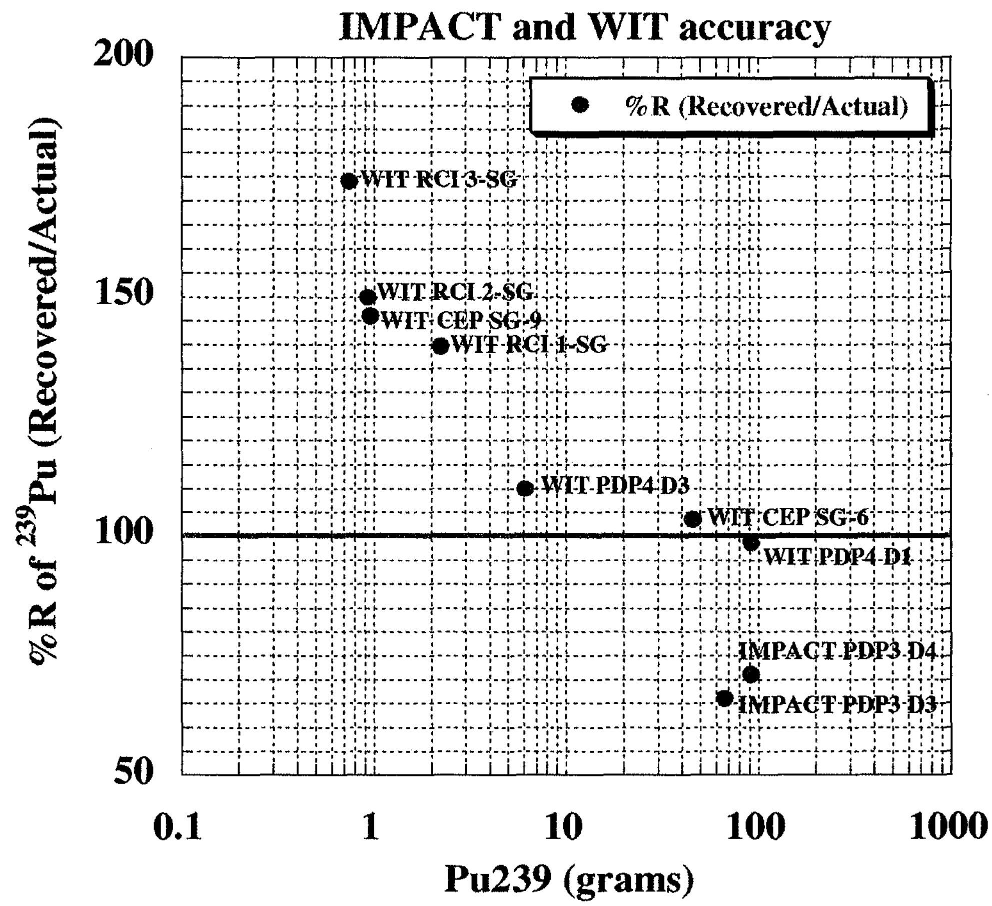

Figure 3: IMPACT and WIT scanner accuracy as a function of gram loading.

SUMMARY

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 grams. LLNL and BIR are participating in the DOE Performance Demonstration Program (PDP), the Capability Evaluation Project (CEP), and the Rapid Commercialization Initiative (RCI) studies. Every official performance measure that the IMPACT and WIT scanners have participated in has been successfully passed. However, the IMPACT system appears to have an approximate 30% low bias with a high gram loading, but with precision at or below 1% for 15 replicate measurements. We have demonstrated a 10% improvement in this bias when using the CCG reconstruction method over MLEM. In addition, the IMPACT system has demonstrated high-biased results for drums with a low TRU loading; however, this appears to be a result of image reconstruction codes that improperly handled statistical zero counts. We have developed a new CCG emission reconstruction code that appears to improve this positive bias. The WIT system participated in three different blind test measurement programs and the results show that it does not appear to have a low bias for any type of waste matrix; however, it does appear to have a high bias for low-activity drums. We can reduce this bias by using our new CCG emission reconstruction code.

Present work underway for the WIT system includes an upgrade from one HPGe detector to six to improve system throughput.[ROB97] We are also implementing a continuos motion scanning modality. These two improvements in throughput will enable both the ACT and PCT measurements to be made from 10 times faster (for low activity in high attenuating matrices) to 40 times faster (for high activity in a low density matrix continuously scanned). We are also developing an improved automated gamma-ray spectroscopy analysis code that will enable almost all TRU isotopes, as well as other infrequently occurring waste radioactivity, to be identified.

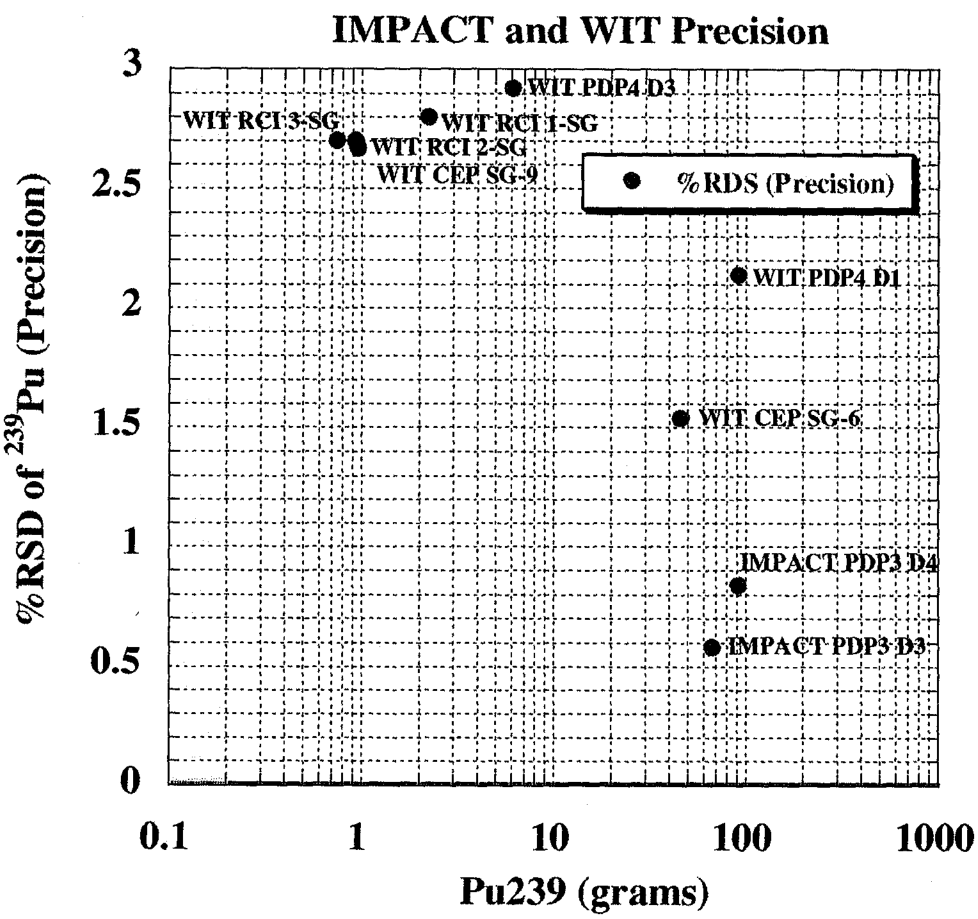

Figure 4: IMPACT and WIT scanner precision as a function of gram loading.

FUTURE DEVELOPMENT ACTIVITIES

There are six areas associated with the A&PCT technology that are currently being developed or should be further developed. They include:

(1) Multiple detectors,

(2) "Smart" scanning techniques to shorten drum scan times,

(3) Further optimization of the automated isotopic analysis program,

(4) Further investigation into the apparent bias as a function of mass,

(5) Continued testing of lump correction techniques,

(6) Fusion of other NDE and NDA modalities that could lead to more accurate assay values and more complete waste characterizations.

Multiple Detector Development

One limiting restriction to the A&PCT technology is the relatively long scan time required to assay a drum. In part, long scan times are due to the use of only a single HPGe detector for recording data and the overhead (time spent) associated with discrete movement of the drum. We are working on methods to reduce data acquisition scan times in the ACT and PCT modes without compromising the accuracy of the assay.[ROB97]

We are working with BIR to upgrade the WIT/A&PCT scanner to incorporate multiple detectors and to include a continuous motion scanning mode. Figure 5 shows the expected speed up of the new scanner (red and green lines), and is compared to the current single-detector A&PCT scanner (blue line). The ACT and PCT ray sum integration times used for a drum assay are dependent on the waste matrix gamma-ray attenuation and the activity level of the transuranic radioisotopes. Depending on the ray sum integration times used, the new A&PCT system design should reduce the counting time by a factor of 10 for the long integration times currently required to assay low waste activity and to perhaps as much as a factor of 40 or more for the highest levels of waste radioactivity.

Smart Scan Techniques

There are methods that will increase the throughput of the A&PCT technology. One solution may be to develop a "smart" scanning scenario for dense (>0.7 g/cm^3) waste drums. If they are found to be relatively homogeneous when evaluated by an NDE radiographic or transmission CT system, then a single ray sum of the drum and matrix is all that is required to computationally construct the active volume image. This smart scanning method would decrease the active counting time by a factor of up to 1000. There may be other smart scanning scenarios that are waste matrix dependent. These could be studied and possibly used to increase waste drum throughput.

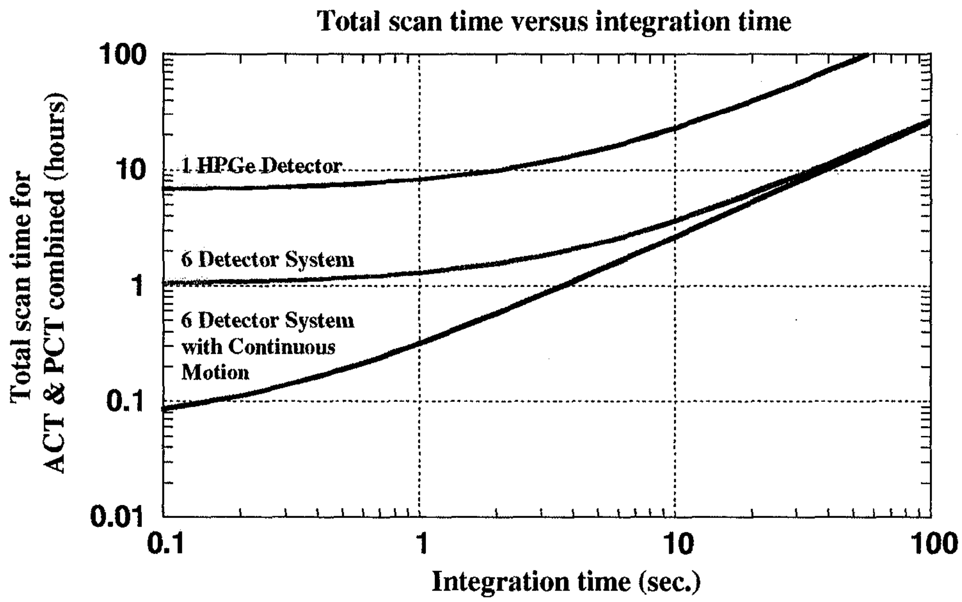

Figure 5 The total active and passive assay scan times required for various A&PCT scanner configurations plotted as a function of ray-sum integration time. The ray-sum integration times shown are the same for both active and passive modes.

Automated Isotopic Analysis

We have developed an automated isotopic analysis program for a select number of TRU waste streams. This program is designed to be flexible enough so that it can be tailored to variable waste streams. Our experience in analyzing TRU waste isotopics will increase over time and reveal changes and additions that could improve the isotopic analysis code. These enhancements may include the addition of isotopes that were not initially expected in the TRU waste streams, and optimization for isotopes that occur more than originally expected. Incorporating these and other future experiences will lead to a more robust automated operation of the code.

Assay Bias Evaluation

Data acquired to date indicates an apparent bias as a function of mass. We have investigated and partially addressed this situation. We are not thoroughly convinced that this bias is completely resolved; therefore, further evaluation is needed.

Lump Corrections

Lump correction is required when the size of the radioactive material or lump being assayed is massive enough to self-attenuate its own gamma-ray emissions. Although we have developed a method to correct for the self-attenuation expected from certain size lumps of Pu, it might be possible to improve this technique by other methods. Just recently, PDP standards that exhibit self-attenuation problems have become available. These PDP sources are well characterized and should help reveal which lump correction methods will perform the best. However, these sources should also be used to evaluate and optimize current methods that show promise for solving the lump problem. Further work may be necessary in this area to develop new methods or improve existing methods for lump corrections.

One possible solution to this problem is to use a high-spatial resolution transmission computed tomography volume image fused with the A&PCT image to identify where TRU lumps are located. If the TRU lumps can be identified within the transmission CT image, a segmentation process can be performed on the volume to extract the objects. Segmentation is a process that is used to remove an object of interest from a CT volume image based on pixel values and connectivity. Once the TRU lump is computationally removed, the volume can be determined from the dimensionally correct pixel number that represents the object. If this method is feasible, it could be implemented on a system like the WIT trailer because it contains a transmission CT scanner that has adequate spatial resolution.

Data Fusion

Data fusion is the process of integrating the results from both NDE and NDA characterization techniques to achieve a more accurate assay or to increase the confidence of an assay. The potential of this integration has yet to be realized. The integration of A&PCT quantitative data and NDE high-spatial resolution data may solve problems related to lump corrections as mentioned above. Also, NDE radiographs or CT images can be used to determine information about the homogeneity of a drum that in turn can be used to determine the optimum A&PCT scan geometry that should be used to obtain maximum throughput. The integration of other NDA assay modes with A&PCT data sets will provide increased accuracy and confidence.

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