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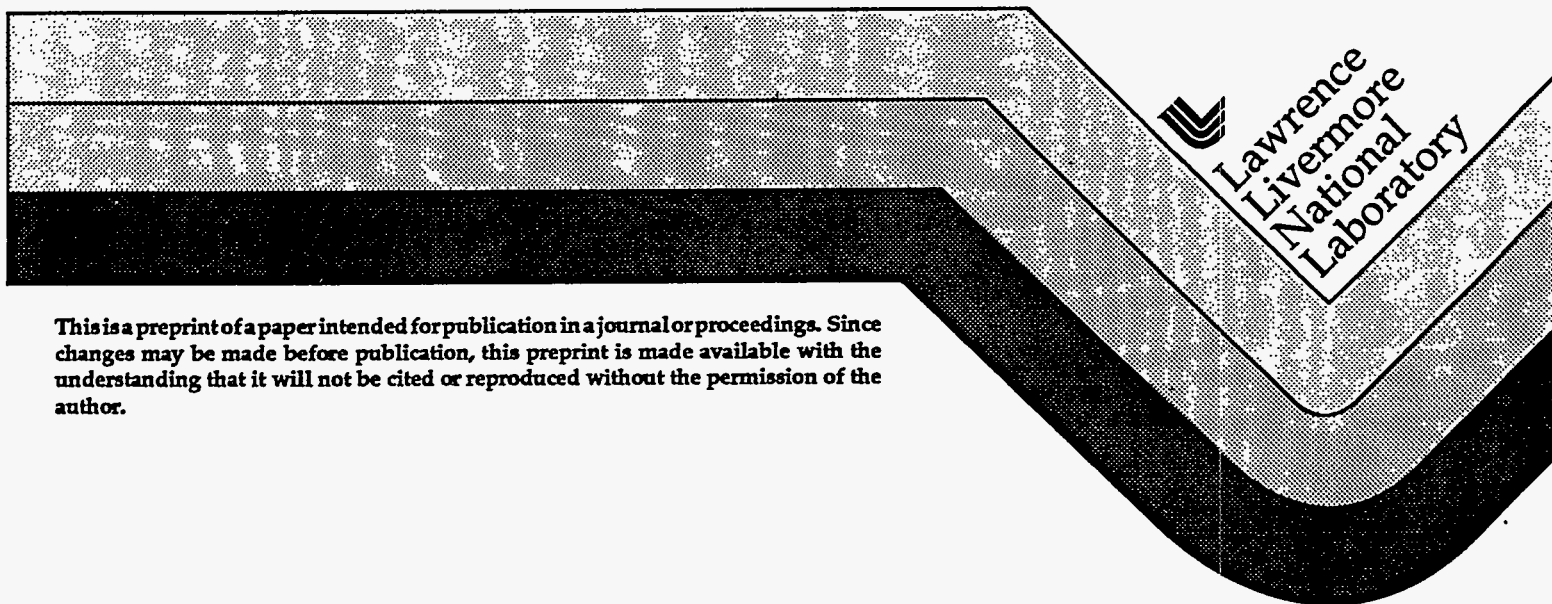# Nondestructive Assay of TRU Waste Using Gamma-Ray Active and Passive Computed Tomography

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Nondestructive Assay of TRU Waste
Using Gamma-ray Active and Passive Computed Tomography

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ABSTRACT

We have developed an active and passive computed tomography (A&PCT) scanner for assaying radioactive waste drums. Here we describe the hardware components of our system and the software used for data acquisition, gamma-ray spectroscopy analysis, and image reconstruction. We have measured the performance of the system using "mock" waste drums and calibrated radioactive sources. We also describe the results of measurements using this system to assay a real TRU waste drum with relatively low Pu content. The results are compared with x-ray NDE studies of the same TRU waste drum as well as assay results from segmented gamma scanner (SGS) measurements.

INTRODUCTION

LLNL has developed a system of active and passive computed tomography, a comprehensive and accurate gamma-ray NDA method, that can identify all detectable radioisotopes present in a container and measure their activity.[MAR92, MAR92a, and ROB94] The A&PCT technology could be used to certify radioactive or mixed wastes when they are below the TRU threshold, determine if they meet regulations for LLW, and quantify TRU wastes for final disposal.

The A&PCT system produces two- and three-dimensional images of cross sections of the container contents. The A&PCT technology requires an active CT measurement using gamma-rays at different energies to map the unknown waste matrix in a drum, revealing the attenuating objects. The passive CT measurement locates and identifies any detectable radioisotopes present within the drum. Using the combination of the two measurements, it is possible to correct the measured radioactive intensities for attenuation caused by drum contents. These corrected passive intensities provide an accurate quantitative absolute measure of the source strength of all detected internal radionuclides. This provides the ability to classify detected radioactivity within the drum as transuranic or low-level, according to their measured activities and to repository regulations.

A single detector A&PCT scanner is being used to evaluate both mock and real TRU waste drums. The mock waste drums are used to calibrate system parameters, verify simulation studies and evaluate the scanner's performance. Many parameters within the reconstruction algorithms that are used in this process are determined and verified using both simulated and measured data. The measured data are acquired by scanning mock waste drums containing various waste and emission source distributions.[CAM94] The results of scanning a well-characterized point source are presented in this paper to demonstrate the scanner's ability to perform absolute and accurate assays.

All aspects of the A&PCT technology are being tested by performing scans of real TRU waste drums. At this time, the scanner is being used for research and development and is not considered a production-mode assay system. We have successfully scanned a full TRU waste drum containing approximately 3 to 4-grams of ^{239}Pu. The drum was scanned using various detector integration

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times and reconstructed with both two and three-dimensional emission algorithms. The results from these scans are presented in this paper.

EXPERIMENTAL METHOD

Scanner description

The A&PCT prototype at LLNL has recently been moved and upgraded so that full sized TRU waste drums can be scanned and assayed. The prototype scanner was only capable of scanning two-thirds of the waste drum and was limited to a 200 lb capacity due to the drum handling system. Additionally, it was not possible to scan real waste within the facility where the prototype scanner was located.

The upgraded A&PCT scanner is shown in Figure 1 and is called IMPACT (Isotope Measurements by Passive and Active Computed Tomography). The IMPACT scanner consists of two towers constructed of aluminum interlocking space-frame tubing. One tower supports the active ^{166m}Ho isotope source and the other supports a well-collimated single high-purity Germanium (HPGe) detector system. The towers are designed to be versatile so that additional sources and detectors could be added to the scanner in the future. Between the two supporting towers is a three-axis drum manipulator.

Figure 1: Photo of the IMPACT scanner.

The most significant upgrade to the IMPACT scanner is the three-axis drum manipulator. The manipulator is capable of translating and elevating a 1000 pound waste drum 50 inches. In addition, the manipulator rotates the drum 360 degrees. The drum manipulator was designed to be

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robust with safety features that are necessary for handling TRU waste in a seismic active area. Engineering safety notes were developed for the entire IMPACT scanning system.

A one-ton jib crane is located at one end of the IMPACT scanner and provides easy and safe loading of the heavy waste drums. The crane is interlocked in a home position away from the drum manipulator to assure that the waste drum cannot be driven into the jib crane during system operation.

The IMPACT scanner uses a PC for system control and data acquisition. During both active and passive operations, the PC control computer positions the drum for data acquisition. After positioning, the control computer communicates with a multi-channel analyzer (MCA) that acquires data from the HPGe energy-discriminating detector. The detector integration time is variable, and when completed, the control computer downloads the MCA's data and stores selected energy regions of interest and/or the entire spectrum onto a system disk. After the data is stored, the control computer and manipulator move the drum to the next integration position (also known as a ray sum). The PC again acquires data from the MCA and HPGe detector system. The IMPACT scanner translates the drum in 2-inch increments after each integration point. A 55-gallon (208-L) drum is translated 14 times and then rotated approximately 17 degrees. Each set of 14 ray sums make up a projection. After the rotation, the next projection of 14 integration points is acquired with a 2-inch translation after each ray sum. This process is repeated for 21 rotations (or projections) before the drum is elevated 2 inches and the process is repeated for the next level or slice plane. IMPACT acquires 18 slice planes for a 55-gallon drum.

The data acquired from IMPACT is processed, reconstructed, and analyzed on a UNIX based work station. A UNIX file system is mounted by the PC and data are transferred over an ethernet cable. Both the energy regions of interest (EROI's) and/or spectrum that represents each individual ray sum are transferred. The advantage of this method is that the data can be analyzed on the UNIX system as it is being acquired on the PC.

The EROI data is simply the integrated photon counts within an energy peak of interest where the background is subtracted to produce a net value of activity for some specified integration time. The EROI is set prior to the assay on the known energy peaks of the active or passive sources. The EROI data is processed and reconstructed without any need for further isotopic analysis. The disadvantage of using the EROI data for the A&PCT reconstruction is that the type of emission sources that are being evaluated within the drum must be known prior to the assay. This may not be the case for all waste drums being assayed; therefore, there is an option to save the spectrum acquired for each ray sum. The accumulated gamma-ray spectra are used to determine the isotopics of the waste drum prior to reconstruction. The EROI's required for image reconstruction are extracted from each ray sum's spectrum after the isotopes have been identified.

The A&PCT images are reconstructed from the projection or ray sum data in two steps. The first step produces a map of the attenuation from the active ray sum data. The second step uses passive (or emission) ray sum data to localize the sources of radioactivity. During the second step, the active reconstructed attenuation map is used to correct the radioactivity for attenuation to determine the absolute assay. Filtered backprojection is used for the reconstruction of an attenuation map from the active, or transmission, data. This reconstruction code has been extensively developed and is a standard in industry.[AZE90 & ROB92]

LLNL has collaborated with the University of California at San Francisco (UCSF), School of Medicine, Department of Radiology to develop the emission image-reconstruction software for this project. The emission code uses Maximum Likelihood-Expectation Maximization (MLEM) where we consider the data to be the result of a stochastic process based on a Poisson probability distribution. For any given model of the source, there is a certain probability of actually obtaining the measured data. In the reconstruction, the model of the source is varied to maximize the

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probability of obtaining the measured data.[BRO92 & KETO95] Present procedures reconstruct both 2D and 3D-images. The 2D case assumes that the source structure is uniform along one axis (i.e. line sources) and the 3D image reconstruction program makes no assumptions as to source structure.

Point Source Calibration

A 63.6 µCi ^{133}Ba point source was assayed to verify the operation of the IMPACT scanner and the reconstruction codes. The 356 keV peak was used in the assay. The point source was placed approximately 12 inches off the axis of rotation and was scanned with no attenuation. Several slice planes were acquired above and below the Ba source so that a 3D image could be reconstructed where all the activity is included. Figure 2 shows three images that represent the reconstructed center slices of the ^{133}Ba point source. The location of the point source is well defined in the images and the activity results are shown in Table 1. There is good agreement between the two MLEM codes and the measured results are within 6% of the activity of the calibration source.
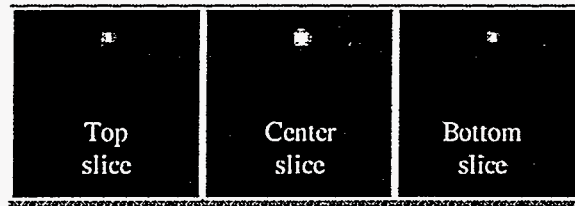

Figure 2: PCT reconstructed images of three slice planes of the point source.

Table 1: Assay of the ^{133}Ba source

63.6 µCi (Ideal)	2D MLEM Recon code	3D MLEM Recon code
Total Counts (measured)	3.82×10^6	3.86×10^6
Activity (measured)	66.6 µCi	67.3 µCi
% Error from Ideal	4.5%	5.5%

RESULTS

The IMPACT system has been used to successfully scan a TRU drum filled with legacy laboratory waste. The TRU waste drum was selected and characterized using both A&PCT and high spatial resolution x-ray radiography/computed tomography. The drum is a standard 55 gallon (208-L) sealed container that has a gross weight of 250 pounds and net weight of 165 pounds. The drum was scanned by SGS and was reported to contain 4.1 grams of weapons grade ^{239}Pu.

Both active and passive tomography were performed on the waste drum using 14 ray sums (integration points), 21 projections and 18 slice planes. Both 10 and 100 second integration times were used for each ray sum acquired and various energy peaks (EROI's) were acquired simultaneously. A spectrum was also saved for each ray sum of the passive data set for an isotopic evaluation. The EROI data are reconstructed using both 2D and 3D MLEM reconstruction codes. The results from the isotopic evaluation and reconstruction characterization and assay are presented below.

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Isotopics

The gamma-ray spectra accumulated in our A&PCT measurements were used to determine the Pu isotopics of the TRU waste drum. Ideally this would be done by reconstructing the ray sum data from gamma-ray transitions in each of the isotopes in question and then comparing the absolute assay values. Unfortunately the low statistics for the gamma ray transitions for ^{240}Pu and ^{238}Pu in the passive CT data preclude this approach. However, if it is assumed that the isotopic composition is constant throughout the drum, summed gamma-ray spectra can be used to determine the isotopic ratios of the TRU components. This is done by studying pairs of closely spaced lines in the complex gamma-ray spectrum of TRU material. The energies and branching ratios of the pertinent gamma-ray transitions are well documented.[GUN76]

The ^{241}Am/^{239}Pu mass ratio is determined by comparing the intensity of the 125.29 keV transition in ^{241}Am to the 129.29 keV intensity in ^{239}Pu and correcting for their branching ratios and half-lives. We assume that the differences in attenuation and efficiency are negligible for the small energy difference of these transitions. These two transitions lie in a rather complicated region of the spectrum that contains two transitions from ^{241}Am and 4 transition from ^{239}Pu. A peak fitting code GRPANL [GUN88] is used to extract the intensities of these peaks. In doing this we use GRPANL's ability to relate peaks in both energy and intensity, for this particular analysis we fix the energy of all the peaks relative to the 129.29 keV line. The intensities of the ^{239}Pu lines are also related to this peak. Finally, the ratio of the intensities for two ^{241}Am peaks are also fixed. This procedure allows us to use all the available spectral information to increase the precision of the extracted intensities. A similar method is used to extract the ^{241}Pu/^{239}Pu and ^{238}Pu/^{239}Pu ratios by analyzing the group of transitions in the 140 - 155 keV region of the gamma-ray spectrum. The ^{239}Pu intensity is determined from the 144.21 keV transition whereas the ^{241}Pu and ^{238}Pu intensities are related to the peaks at 148.57 and 152.68, respectively. In addition, the group includes 5 other related transitions.

Finally, the 157 - 166 keV region is used to analyze the ^{240}Pu/^{239}Pu ratio, the group includes 8 transitions that are used to extract the intensity of the 161.45 keV line (^{239}Pu) and the 160.28 keV transition (^{240}Pu). Unfortunately these lines are rather weak in the summed spectrum limiting us to a precision of 20% for this important ratio. Table 2 gives the mass ratios for the isotopes that have been analyzed along with the expected values for nominal weapons grade Pu. One can see that our data are quite consistent with a weapons grade characterization of this material.

Table 2 : Mass ratios extracted from the summed gamma-ray spectra.

Ratio	Experiment	Error	Nominal
^{238}Pu/^{239}Pu	1.84E-04	5.39E-05	1.71E-04
^{240}Pu/^{239}Pu	6.50E-02	1.40E-02	6.31E-02
^{241}Pu/^{239}Pu	2.69E-03	2.73E-04	4.08E-03
^{241}Am/^{239}Pu	2.32E-03	1.04E-04	2.16E-03

Active Reconstruction

Active data were acquired at both the 410 and 365 keV peaks of the active ^{166m}Ho source. The active reconstructed images for the 410 keV peak are shown in Figure 3. Each slice plane (slice 0 through 17) for the TRU drum is shown. The outline of the waste drum can be seen in each of the active images. The first image (slice 0) shows the attenuation map of the upper ratchet style clamp

Slice 0 (0−2")	Slice 1 (2−4")	Slice 2 (4−6")	Slice 3 (6−8")	Slice 4 (8−10")	Slice 5 (10−12")
Slice 6 (12−14")	Slice 7 (14−16")	Slice 8 (16−18")	Slice 9 (18−20")	Slice 10 (20−22")	Slice 11 (22−24")
Slice 12 (24−26")	Slice 13 (26−28")	Slice 14 (28−30")	Slice 15 (30−32")	Slice 16 (32−34")	Slice 17 (34−36")

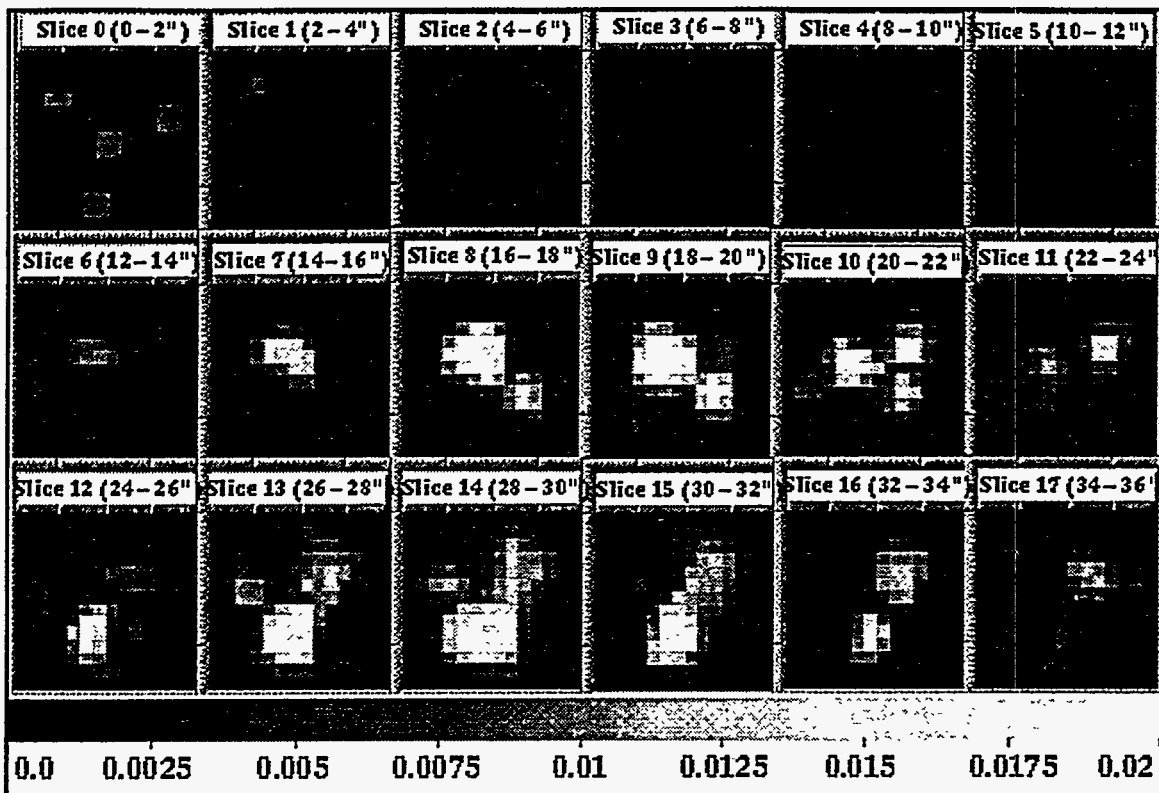| 0.0 | 0.0025 | 0.005 | 0.0075 | 0.01 | 0.0125 | 0.015 | 0.0175 | 0.02 |

Figure 3: Reconstructed active images for each slice plane of the TRU waste drum showing the attenuation map of the waste matrix. The images were reconstructed using the 410 keV peak of the ^{166m}Ho active source.

used to hold the drum in place. The next 5 slices show little or no attenuation within the drum, indicating that no objects exist in this region. The first image where waste objects are present is the 6th slice, located approximately 13 inches below the top of the drum. Highly attenuating objects are present in the last 12 slices that represent the bottom 2/3rds of the drum.

A digital radiograph and high spatial resolution CT image are shown in Figure 4a and b, and can be compared to the A&PCT active image shown in Figure 4c. This figure displays the distribution of the waste matrix inside the drum. The high attenuating waste material appears to be homogeneous. The digital radiograph shows the waste material settled in the bottom 2/3rds of the waste drum.

Figure 5 shows a 3D rendered view of the x-ray high resolution CT data set of the TRU waste drum. The highly attenuating objects within appear to be cylinders of some kind of homogeneous material. There appear to be 3 large cylinders approximately 12 inches in diameter and three smaller cylinders approximately 6 inches in diameter. High resolution film radiography was also performed on the drum and revealed smaller "soup" cans around the larger cylinders. These cans have very low attenuating materials within. The radiograph is not shown in this paper.

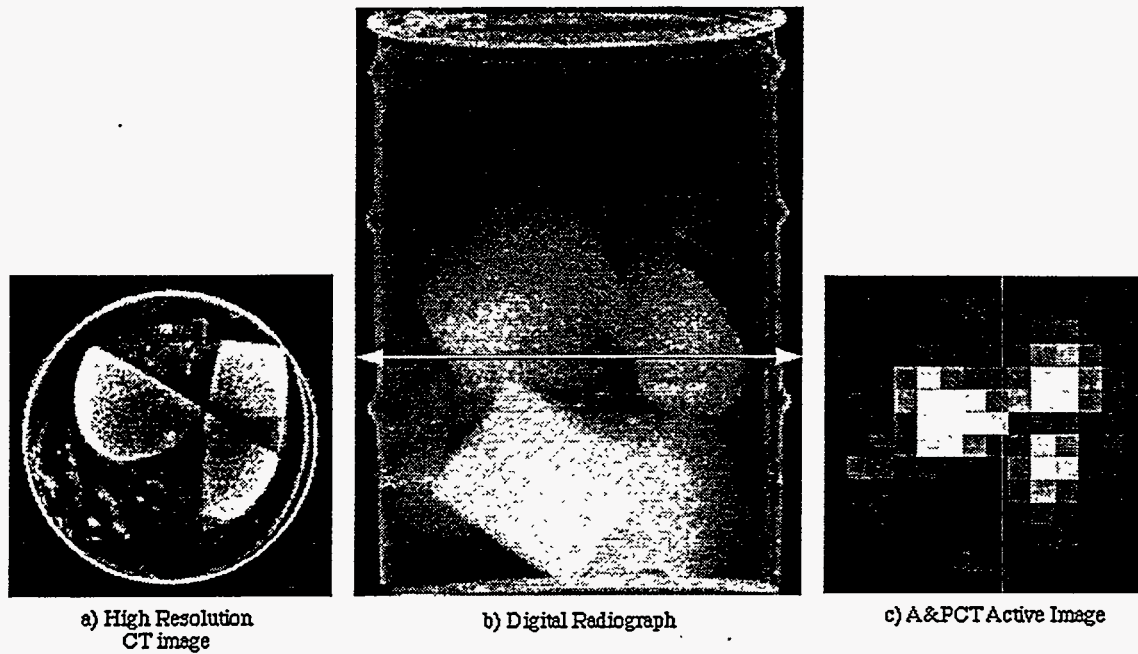a) High Resolution   b) Digital Radiograph   c) A&PCT Active Image
CT image

Figure 4: (a) High spatial resolution x-ray CT image, (b) digital radiograph and (c) active A&PCT image of the TRU waste drum. The slice plane for the CT images is located approximately 21 inches below the top of the drum (arrow in b).

Figure 5: A 3D rendering of the x-ray CT image of the TRU waste drum shows cylinders of homogeneous material that are highly attenuating.

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Passive Reconstruction

Passive CT data were analyzed for both the 414 keV and 375 keV peaks of ^{239}Pu and were corrected for attenuation using the 410 keV and 365 keV ^{166m}Ho peaks, respectively. The passive reconstructed images for the 414 keV peak are shown in Figure 6. All of the slice planes for the drum are shown. The passive images reveal that little or no activity is present in the first 7 slices, or top 15 inches of the drum. There is also little activity in the bottom slices of the drum. Most of the activity in this TRU drum is in the center and is approximately located in the same area as the highly attenuating objects.

The distribution of the activity is more clearly revealed in Figure 7. This figure is a rendering of only the activity of the ^{239}Pu source within the drum. Most of the activity appears to be located in the middle portion of the drum and is somewhat lumped together.
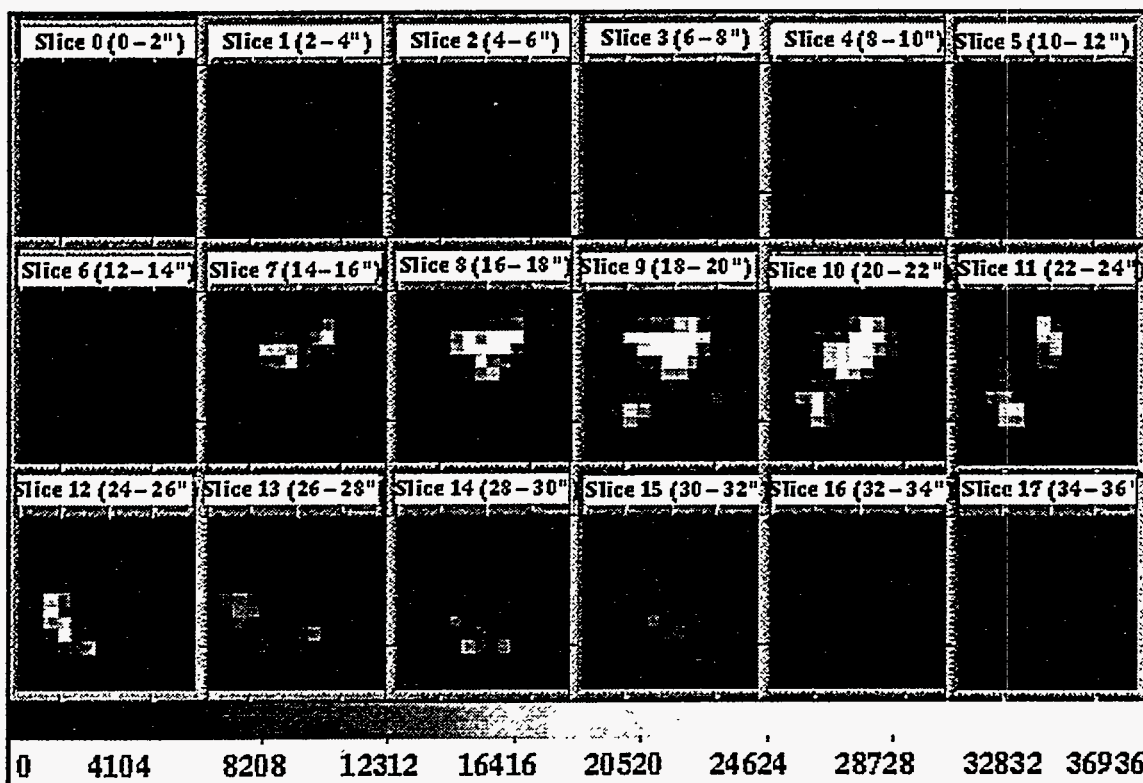

Figure 6: Reconstructed emission images at 414 keV showing the location of ^{239}Pu activity within the TRU drum. The activity is corrected for attenuation loss.

A&PCT Assay

For the assay of this drum, the 410 keV peak of the ^{166m}Ho active source is used to correct for attenuation of the 414 keV peak of ^{239}Pu. Also, the 365 keV peak of ^{166m}Ho is used to correct the 375 keV peak of ^{239}Pu. The results of this assay are shown in Tables 3 and 4. This data does not include corrections that may be required for self absorption of the passive sources. The high spatial resolution radiographs and CT scans indicate that the materials are homogeneous (i.e. apparently no large lumps of ^{239}Pu exist) and may not need these corrections. However, self absorption is a
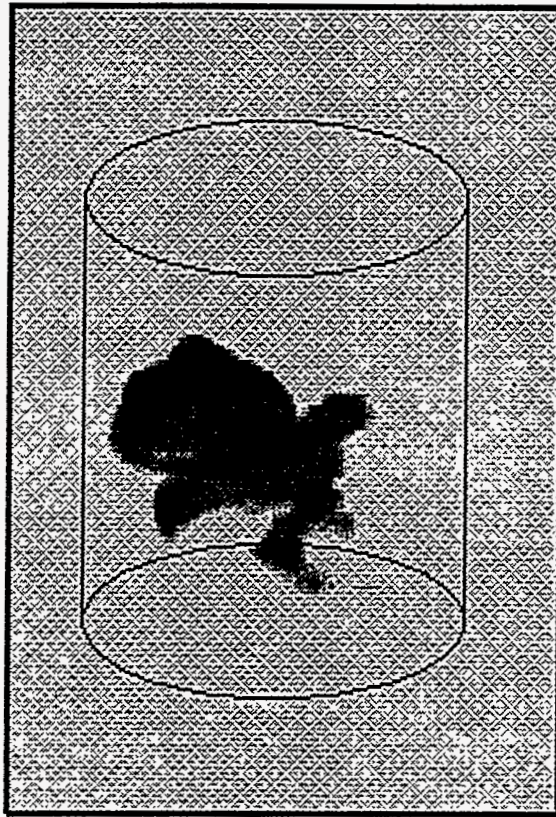
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Figure 7: 3D rendered view of the distribution of the ^{239}Pu emission within the TRU waste drum.

possible source of error in the assay of waste drums and will be examined in future analysis. Data are tabulated for two ray sum integration times of 10 and 100 seconds.

The results of the 2D reconstruction are shown in Table 3. Two-dimensional passive reconstruction is performed for each slice plane of the waste drum. The 2D images are then merged to form a 3D image and the sum is calculated to determine the total activity. The activity in the two ^{239}Pu peaks for both the 10 and 100 second integration times are within 3% of each other. However, there is a difference of approximately 20% between the 10 and 100 second data for both peaks. The variation could be simply due to a difference in the counting statistics of the data.

Table 3: TRU drum assay for two energy peaks and two different integration times using the 2D MLEM code.

	2D MLEM Emission Reconstruction Code					
	^{239}Pu 414 keV peak			^{239}Pu 375 keV peak		
	Total counts	Activity	Pu quantity	Total counts	Activity	Pu quantity
100 sec. ray sum	4.66×10^6	176 mCi	2.84 g	$4.99 \ 10^6$	171 mCi	2.76 g
10 sec. ray sum	5.60×10^5	212 mCi	3.42 g	6.26×10^5	216 mCi	3.48 g

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The results of the 3D MLEM reconstructions are shown in Table 4. The 3D image reconstruction program makes no assumptions as to source structure where the 2D case assumes that the source structure is uniform along one axis (i.e. line sources). The 3D code requires the entire volume of both the active and passive data sets for the reconstruction process. The results show that the two ^{239}Pu peak activities are within 10% of each other. However, as with the 2D reconstructed data, the 10 second data is calculated to be 13 to 19% higher in activity then the 100 second data. Again, this is most likely due to the difference in counting statistics between the two data sets.

Table 4: TRU drum assay for two energy peaks and two different integration times using the 3D MLEM code.

	3D MLEM Emission Reconstruction Code					
	^{239}Pu 414 keV peak			^{239}Pu 375 keV peak		
	Total counts	Activity	Pu quantity	Total counts	Activity	Pu quantity
100 sec. ray sum	5.59x10^6	211 mCi	3.40 g	5.51x10^6	189 mCi	3.05 g
10 sec. ray sum	6.42x10^5	242 mCi	3.90 g	6.79x10^5	233 mCi	3.76 g

CONCLUSION

We have significantly modified the prototype A&PCT scanner. The modified scanner (IMPACT) is now located at LLNL and can scan full-sized TRU waste drums that weigh up to 1000 pounds. The system operation and reconstruction algorithms have been evaluated by scanning a ^{133}Ba calibration point source. The measured results are within 6% of the actual source activity. These results indicate that the system and algorithms are apparently operating as expected. A TRU waste drum was also scanned using the new IMPACT system to further evaluate the system and algorithm operation. This evaluation is not meant to be a validation of the system and reconstruction algorithms at this time, but only to verify the operational status of the IMPACT scanner.

We have performed an isotopic evaluation of the TRU waste drum comparing the results with the expected values for nominal weapons grade Pu. The results are quite consistent with a weapons grade characterization of this material.

The active assay of the TRU drum reveals that heavy attenuators are present within the lower 2/3rds of the waste drum. These attenuators are apparently homogeneous cylinders of various sizes. The Passive assay is performed using two different MLEM codes that correct for the attenuation matrix of the waste drum. The results from these codes are consistent. For this assay, no corrections are made for the possible self absorption of the ^{239}Pu emission source. The assay shows that approximately 3 to 4 grams of ^{239}Pu is present in the waste drum. At this time there is no error (systematic or reconstruction) included in the assay.

Future Work

In the upcoming year, we will be concentrating on increasing the speed of the A&PCT system and implementing an estimate of error associated with the assay. Multiple detectors will be added to the IMPACT scanner. System components and parameters will be optimized for speed. Validation procedures will be performed in the next year to characterize the system performance.

ACKNOWLEDGMENTS

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