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# WASTE CHARACTERIZATION ACTIVITIES AT THE LAWRENCE LIVERMORE NATIONAL LABORATORY

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## ABSTRACT

Radioactive and hazardous wastes are generated at many national laboratories, military sites, fuel fabrication and enrichment plants, reactors, and many other facilities. At all of these sites, wastes must be separated, categorized, possibly treated, and packed into containers for shipment to waste-storage or disposal sites. Prior to treatment, storage or, shipment, the containers must be characterized to determine the ultimate disposition of the contained waste. Comprehensive and accurate nondestructive evaluation (NDE) and nondestructive assay (NDA) methods can be used to characterize most waste containers in a safe and cost-effective manner without opening them. The Lawrence Livermore National Laboratory (LLNL) is investigating and developing the application of x-ray and γ-ray methods to nonintrusively characterize waste containers and/or items. X-ray NDE methods are being investigated to determine whether they can be used to identify hazardous and non-conforming materials. A γ-ray NDA method is used to identify the radioactive sources within a container and to accurately quantify their strength. In this paper we describe five waste characterization projects being conducted at LLNL that apply both the NDE and NDA methods and present results.

## INTRODUCTION

The Lawrence Livermore National Laboratory has spent the last few years developing and applying proven and emerging technologies for non-intrusive characterization of low level, transuranic, and mixed waste. These technologies include both nondestructive evaluation and nondestructive assay methods using a variety of x-ray and γ-ray imaging techniques including:

- Real Time Radiography (RTR);
- Digital Radiography (DR);
- Transmission Computed Tomography (TCT);
- Active and Passive CT (A&PCT);
- Single-Photon Emission Computed Tomography (SPECT);
- X-ray fluorescence (XRF);
- Spectroscopy Radiation Detection (SRD).

Here we describe five major activities related to non-intrusive characterization of nuclear waste. These activities are in support of the U.S. Department of Energy (DOE), Environmental Restoration and Waste Management (ER/WM) needs and objectives.

## MIXED WASTE OPERATIONS

The DOE Office of Technology Development Robotics Technology Development Program (RTDP) is an Integrated Program, utilizing complementary skills across the DOE Complex to ensure that safer, faster and cheaper technologies are available for processing DOE wastes. The objective of the RTDP Mixed Waste Operations (MWO) program is to develop robotics and automation technology for existing and planned DOE mixed waste facilities that store, characterize, and treat mixed waste (waste that is radioactive and otherwise hazardous).[GRA94]

One of the major focus areas within the MWO program is Item Characterization and Sorting. Characterization of mixed wastes requires that all hazardous, non-conforming, and radioactive materials be identified, localized, and quantified. With such information, decisions can be made regarding whether the item is treatable or has been adequately treated. Much of the required information can be gained without taking representative samples and analyzing them in a chemistry laboratory.

A characterization systematic plan was developed to satisfy the MWO requirements using non-intrusive inspection methods currently available at LLNL.[ROB94a] The characterization methods used in this project were radiography, computed tomography, γ-ray spectroscopy, and x-ray fluorescence.

Mock waste items were selected and characterized to unambiguously identify hazardous inorganic materials on the surfaces. The presence of heavy metals within the waste items can also be ascertained. The material volumes for simple objects were determined using 3D computed tomography and surface areal densities were measured using x-ray fluorescence.

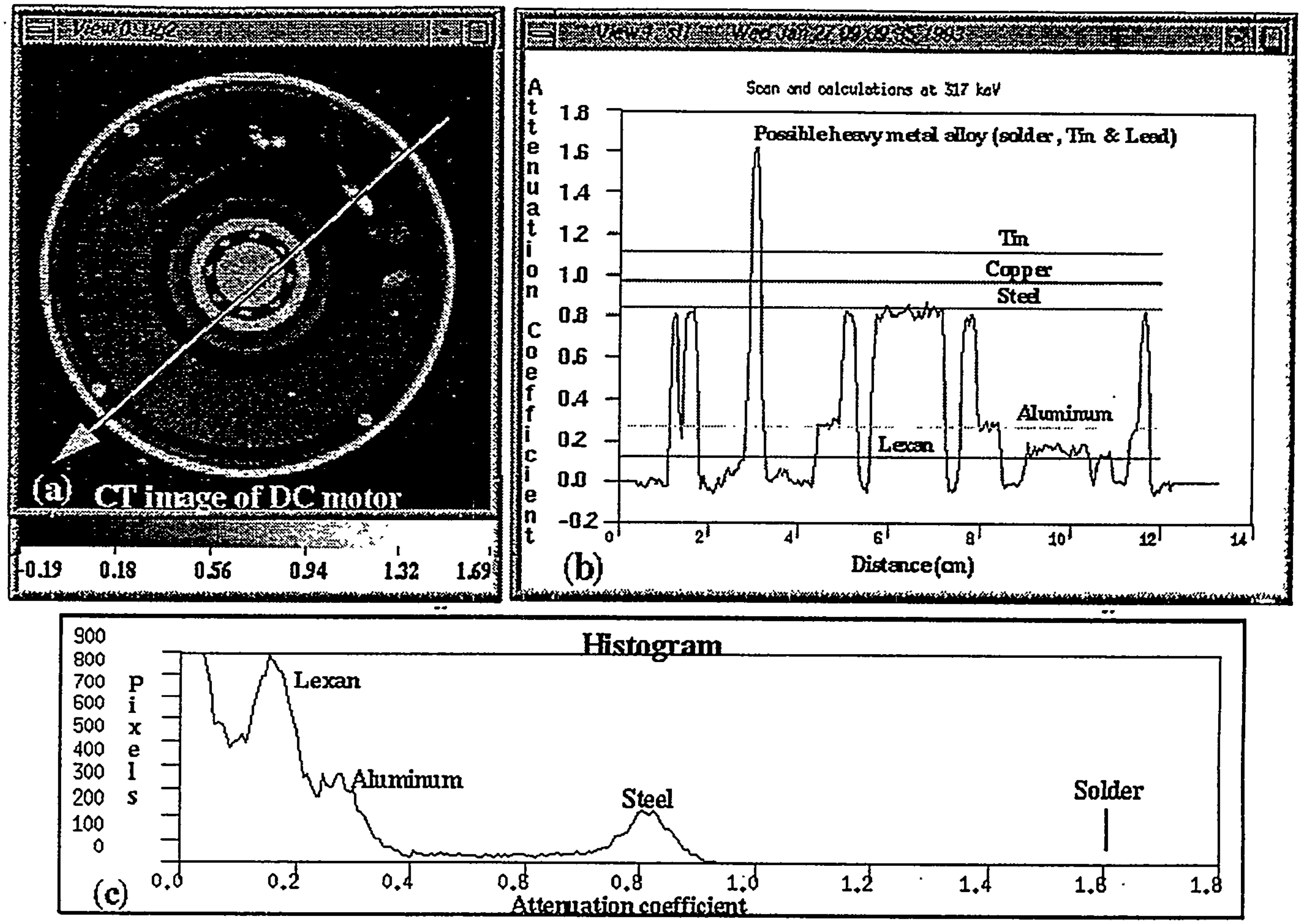



*Figure 1: (a) Representative TCT image of a DC motor. The colorbar below the image provides a scale relating the different shades of gray in the TCT image to the linear attenuation coefficient in $cm^{-1}$. The attenuation variations are related to changes in density ($\rho$) and/or effective-atomic number (Zeff). (b) A 1D profile through the TCT image shows the variation of the linear attenuation coefficient within the motor. The measured attenuation coefficient is compared to theoretically calculated values of various materials (labeled and shown by flat lines). This enables us to determine whether the imaged item is homogeneous or heterogeneous and weather the item contains heavy metals, combustible, or unknown material(s). (c) A histogram of the TCT image can also be used to identify the materials within the individual waste item and indicate their relative amounts.*

Figure 1 shows an example of how various materials can be identified using the variations in pixel intensities within a TCT image. In determining the volume of various materials, segmentation was used to group pixels into regions based on intensity similarity and connectivity. Connectivity is a method that creates partitions in an image based on a pixel's neighborhood. Segmenting the objects in a 3D image provides a more accurate determination of volume because the intensity overlap from other objects within the image is reduced.

The minimum detectable limits for various characterization methods were calculated and confirmed. These detection limits depend upon everything else that is present within the waste item, but range down to sub-milligram per square centimeter levels for heavy elements using radiographic and tomographic methods and below the microgram per square centimeter level (on the surface) using x-ray fluorescence.

## MIXED WASTE MANAGEMENT FACILITY

The Mixed Waste Management Facility at LLNL is a pilot-level demonstration facility that is slated to be completed in 1997. Environmentally acceptable treatment processes that are alternatives to incineration of low-level organic mixed waste will be developed at this facility. The facility will support all essential functions required of a fully integrated plant, including characterization.

The Solid Waste Processing group within the MWMF project has employed the NDE section at LLNL to non-intrusively characterize 102 low level waste drums for the following three criteria:

- Determine the volume fraction of organic combustibles within the waste drums to determine if the drum should be processed through the MWMF;
- Identify items within the drums and evaluate their shapes and sizes to verify that the planned MWMF handling equipment is appropriate;
- Evaluate the contents for general preknowledge concerning drum opening operations.

LLNL has previously developed a scanner for characterizing TRU and LLW drums using RTR, DR, and TCT for other applications.[MAR94] This scanner was slightly modified and used for the MWMF effort. The scanner is called the Waste Drum Radiographic Inspection System (WDRIS) and is shown in Figure 2. It consists of a Phillips 320 kVp x-ray source, staging, and a large area imaging system. The large area imaging system consists of 40" x 34" scintillating screen ($Gd_2O_2S$) that is lens coupled to an 8-bit COHU 4910 CCD camera.

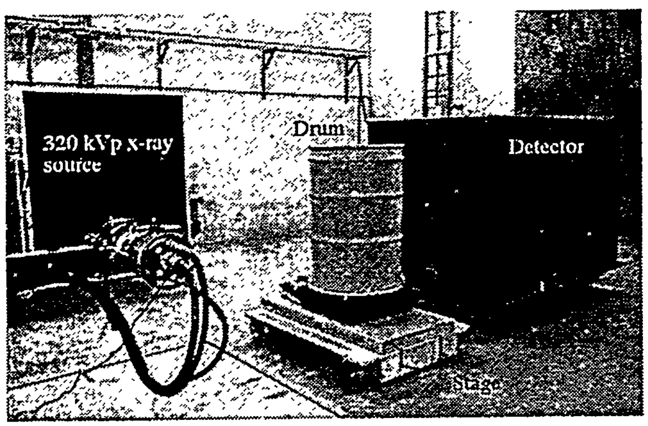



*Figure 2: Medium energy RTR, DR and TCT drum scanner located at LLNL*

Real-time data acquisition was acquired at RS-170 (30 frames per second) and still-view (DR) acquisition was done with a 30 second integration time and no frame averaging. RTR data was acquired over a 360° rotation at 2 vertical positions and DR data was acquired at 0°, 120°, and 240° at 2 elevations for each drum. The throughput for this scanner was limited by regulatory and transportation logistics and was approximately 12 drums/day at an approximate cost of $200/drum. Interactive data analysis was performed on each drum and decisions were based on waste item identification and relative attenuation properties (see Figure 3). The characterization of 102 LLW drums yielded the following results:

- 43 contained greater than 90% combustible material;
- 39 contained 50% to 90% combustible material;
- 14 contained less than 50% combustible material;
- 6 contained liquid.

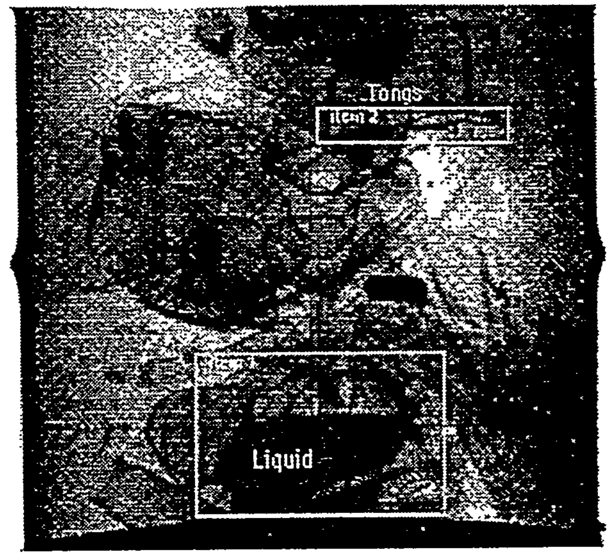
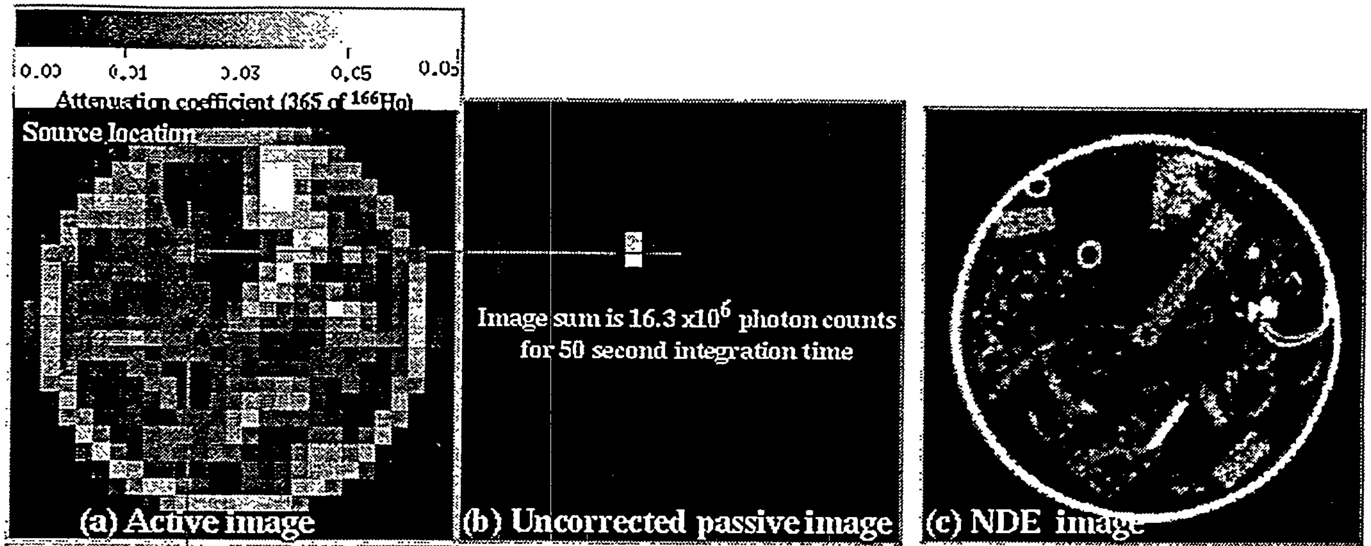


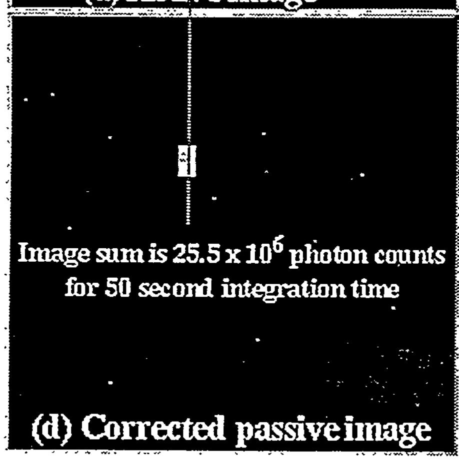

*Figure 3: DR of a LLW drum containing liquid*

## BURIED WASTE INTEGRATED DEMONSTRATION

The Buried Waste Integrated Demonstration (BWID) program supported the applied research, development, demonstration, and evaluation of advanced technologies identified for comprehensive remediation of buried waste for the ER/WM. Through the BWID program, LLNL has developed the active and passive computed tomography technology, a comprehensive and accurate gamma-ray NDA method, that can identify all detectable radioisotopes present in a container and measure their activity.[ROB94b] 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 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's within the drum as transuranic or low-level, according to their measured activities and to repository regulations.




**Top section of mock–waste drum with ~ 35% average attenuation for 356 peak of $^{133}$Ba passive source**

| | Curies | Photon counts | Error |
|---|---|---|---|
| Ideal | 68.2 μ Ci | $26.19 \times 10^6$ | |
| Measured | 66.4 μ Ci | $25.5 \times 10^6$ | -2.6% |

*Figure 4: Representative reconstructed NDA ACT (a) and PCT (b&d) images, and high resolution NDE TCT (c) images of a full size 208-liter simulated waste drum. For the ACT image, the colorbar (a, top) provides a scale relating the different gray shades in the image to the linear attenuation coefficient in $cm^{-1}$. The attenuation variations in the ACT and TCT images are related to changes in density ($\rho$) and/or effective-atomic number (Zeff). For the PCT images, the gray shades represent the photon counts accumulated at each pixel during the integration period.*

The A&PCT system produces two- and three-dimensional images of cross sections of the container contents. LLNL has collaborated with the University of California at San Francisco, School of Medicine, Department of Radiology to develop the emission image-reconstruction software for this scanner. Figure 4 shows the reconstructed images and results obtained from scanning a mock-waste drum[CAM94] containing a $^{133}$Ba calibration point source using the A&PCT technology. The corrected emission of the $^{133}$Ba source was measured to within 3% of the calibrated source.

## WASTE INSPECTION TOMOGRAPHY

The Waste Inspection Tomography (WIT) system is a mobile semi-trailer with a multi-modality sensor capability for the NDE and NDA of nuclear waste drums.[BER95] The WIT system is being co-developed by Bio-Imaging Research, Inc. (BIR), Lincolnshire, Illinois and LLNL. The trailer contains DR, CT, SPECT and A&PCT scanning systems, all designed to evaluate waste drums as large as 110 gallons and weighing up to 1600 pounds. The WIT project is funded by a Program Research and Development Award (PRDA) contract through the ER/WM Office of Technology Development (OTD/EM-50). As part of the WIT project, LLNL has been transferring the A&PCT technology to BIR and will be participating in the evaluation of the overall system.

Figure 5 shows the rear portion of the WIT trailer where the scanning systems are housed. In the first phase of this project, LLNL has configured, analyzed and characterized the A&PCT spectroscopy hardware that is used for the WIT A&PCT scanner. LLNL has transferred the A&PCT data acquisition and reconstruction software to BIR. The software and hardware has been integrated with BIR staging controls and A&PCT data has been successfully acquired, reconstructed and analyzed. LLNL will participate in a second phase of the WIT project with BIR. Phase II is a twelve month program for further integration and DOE site demonstration of WIT.


*Figure 5: Rear portion of the Waste Inspection Tomography system (photo courtesy of BIR, Inc.).*

## SPENT FUEL AND WASTE MANAGEMENT TECHNOLOGY DEVELOPMENT

In cooperation with the Idaho National Engineering Laboratory (INEL) and the Spent Fuel and Waste Management Technology Development program, LLNL has performed a series of studies for examining the potential of using digital radiography, conventional transmission computed tomography, and active and passive computed tomography for non-destructive inspections of high-level waste (HLW) in glass-ceramic form.[ROB95] The INEL is currently developing a process to immobilize high-level waste in glass-ceramic and package this form in stainless-steel canisters. The packaged waste form must be characterized before it is sent to a repository.

The goal of this project was to demonstrate the feasibility of using radiography and computed tomography to characterize HLW canisters for verification of dimensions, concentricity, density and consistency, fill height, and to detect form defects (e.g. cracks, voids and inclusions). X-ray projection and CT simulation tools were used as a principal means for assessing the initial feasibility of scanning the canisters. INEL fabricated two 1/24-scale simulated waste canisters with controlled defects to be characterized on LLNL NDE/NDA systems. It was not possible to fabricate full-scale canisters at the time of this feasibility study. Experimental data was acquired on these canisters and compared to the simulation predictions to verify the results. Figure 6 shows 3D rendered images acquired from one of the fabricated canisters containing controlled defects of glass and boron pellets, titanium cubes, and drilled holes.

There was good agreement between the simulation study and the experimental data. The simulation code was then used to predict the required parameters necessary for characterizing a full-scale waste canister. These predicted parameters provided a foundation for determining the specifications and system design of a scanner that can be used for the characterization of full-scale waste canisters at INEL.

## SUMMARY

LLNL has successfully assembled a multi-disciplinary team to research, develop and applied both proven and emerging technologies for non-intrusive characterization of low level, transuranic, and mixed waste. Both NDE and NDA assay methods utilizing a verity of x-ray and γ-ray imaging techniques have been used. These technologies have been applied to characterize real TRU and LLW drums, individual (singulated) mock-waste items, and 1/24-scale, high-level mock-waste canisters. A unique assay system using active and passive computed tomography has been developed. A&PCT can non-intrusively and accurately identify and quantify radioisotopes within waste drums and other containers. LLNL is actively transferring the A&PCT and other technology throughout DOE and the private sector.

## ACKNOWLEDGMENTS

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## REFERENCES

[BER95] R. T. Bernardi and H. E. Martz, "Nuclear Waste Drum Characterization with 2 MeV X-ray and Gamma-ray Tomography,' *Proceedings of SPIE X-ray and Ultraviolet Sensors and Applications Conference* (Preprint), San Diego, California, July 13-14, 1995.

[CAM94] David C. Camp, John Pickering, and Harry E. Martz, "Design and Construction of a 208-L Drum Containing Representative LLNL Transuranic and Low-Level Wastes," *Proceedings of the Nondestructive Assay and Nondestructive Examination Waste Characterization Conference*, Pocatello, Idaho, February 14-16, 1994; UCRL-JC-115672, Lawrence Livermore National Laboratory, Livermore, CA, February 1994.

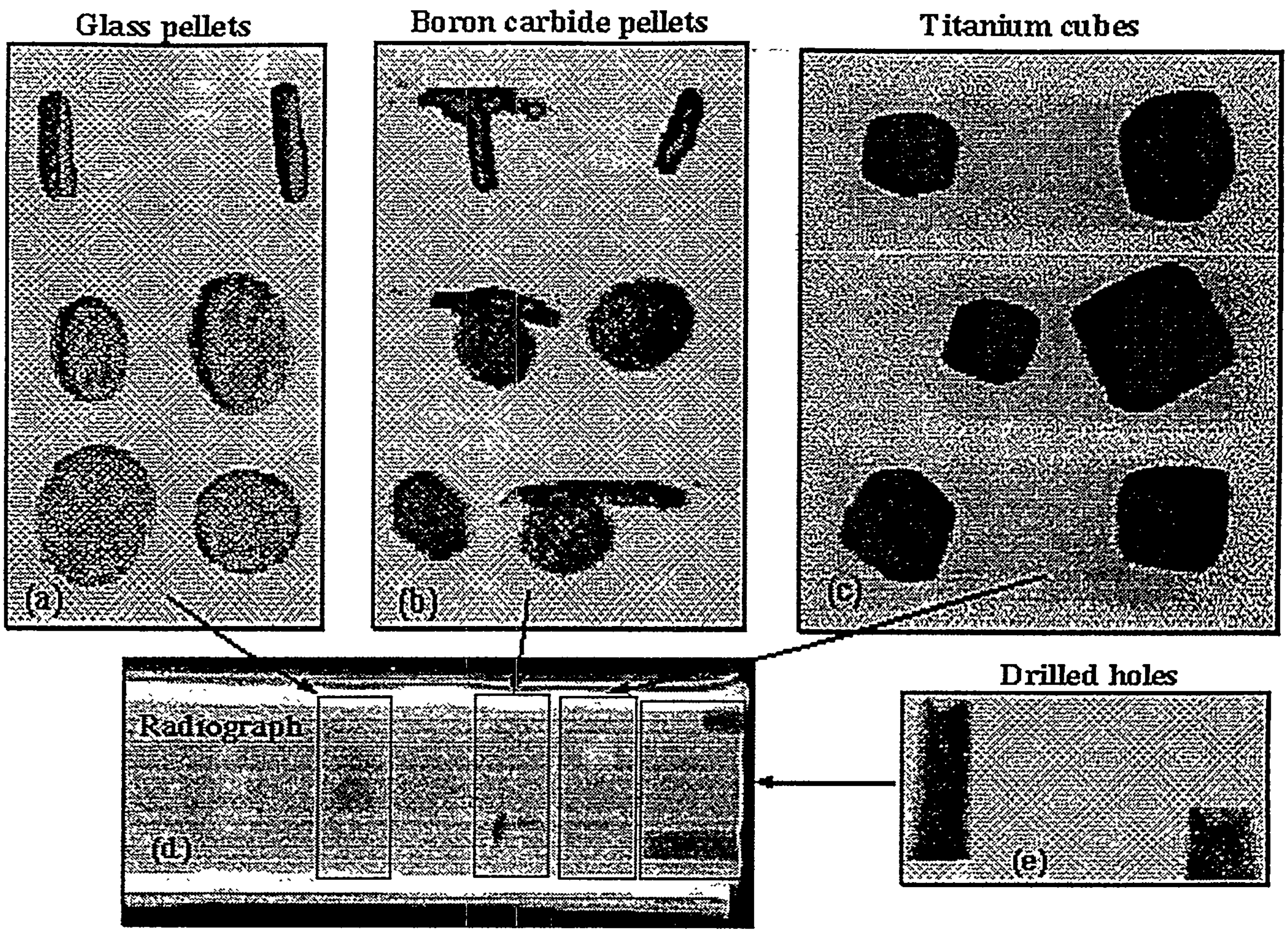


*Figure 6: Four of the controlled defects are rendered (a-c & e) at various angles showing the distribution of the defects within the waste matrix. The rendering process provides a means for computationally striping away materials within the CT volume data set so that anomalies of interest can be analyzed without obstruction. The anomalies can be displayed in different color and opacity that is related to their relative attenuation values. Three rendered views at different angles are shown for each of the three defects shown at the top of the figure (a-c) and one view (e) for the rendered image of the drilled holes. The location of the rendered images within the canister are shown in the radiographic image (d).*

[GRA94] E. Grasz, E. Domning, D. Heggins, L. Huber, R. Hurd, H. Martz, P. Roberson, K. Wilhelmsen, "Summary of LLNL's Accomplishments for the FY93 Waste Processing Operations Program," *Prepared for the U.S. Department of Energy*, 031548002/D-94-012.337m/V-1.4, LLNL, Livermore, CA, March, 1994.

[MAR94] H. E. Martz, D. J. Schneberk, and G.P Roberson, "Real-Time Radiography, Digital Radiography, and Computed Tomography for Non intrusive Waste Drum Characterization," *Proceedings of the Nondestructive Assay and Nondestructive Examination Waste Characterization Conference*, Pocatello, Idaho, February 14-16, 1994; UCRL-JC-115670, LLNL, CA, February 1994.

[ROB95] G.P. Roberson, H. E. Martz, and T. O'Holleran "Feasibility Study for Non-intrusive Characterization of High-Level Waste in Glass-Ceramic Form," UCRL-ID-(to be published), LLNL, CA, 1995.

[ROB94a] G. P. Roberson, R. W. Ryon and N. L. Bull, "Characterization of Mixed Waste for Sorting and Inspection Using Non-Intrusive Methods," UCRL-ID-119298, LLNL, CA, December, 1994.

[ROB94b] G. P. Roberson, H. E. Martz, D. J. Decman, D. C. Camp, S. G. Azevedo and E. R. Keto, "Characterization of Waste Drums Using Non-intrusive Active and Passive Computed Tomography," *Proceedings of the Nondestructive Assay & Nondestructive Examination Waste Characterization Conference*, Idaho State University, Pocatello, Idaho, February 14-16, 1994; UCRL-JC-118317, LLNL, CA, August, 1994.