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Nondestructive Assay of Boxed Radioactive Waste

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
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Westinghouse
Hanford Company Richland, Washington

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NONDESTRUCTIVE ASSAY OF BOXED RADIOACTIVE WASTE

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ABSTRACT

This paper describes the problems related to the nondestructive assay (NDA) of boxed radioactive waste at the Hanford Site and how Westinghouse Hanford Company (WHC) is solving the problems. The waste form and radionuclide content are described. The characteristics of the combined neutron and gamma-based measurement system are described.

INTRODUCTION

Solid radioactive waste must be classified before treatment and disposal methods can be chosen. After treatment and before disposal, the radionuclide contents of a container must be certified.

The two principal types of radioactive waste at the Hanford Site are transuranic (TRU) and low-level waste (LLW). Transuranic waste contains >100 nCi/g of long-lived transuranics. Low-level waste at the Hanford Site contains <100 nCi/g of TRU and is further subdivided into Categories 1, 3, and "greater than 3." Category 1 LLW must contain <10 nCi/g of TRU radionuclides, plus meet other limiting requirements for fission and activation product content. Categories 3 and greater than 3 contain >10 nCi/g but contain <100 nCi/g of TRU and greater quantities of fission and activation products than Category 1.

Nondestructive assay is the technique used to quantify the principal actinides, fission products, and activation products in a waste container. These systems count the gammas from key emitters (typically ^{137}Cs and ^{60}Co) and neutrons originating from spontaneous and induced fission. A waste characterization program develops ratios of all the radionuclides in a waste stream to these key emitters. Using these ratios, the quantities of all the other alpha, beta, and soft gamma-emitting radionuclides present in a container are calculated. With this knowledge of the radionuclide inventory, a container can be placed in the proper waste category.

DESCRIPTION OF THE HANFORD SITE'S BOX WASTE

The Hanford Site's box waste may contain failed equipment, concrete, dirt, glass, paper, plastic, steel, wood or lead, or combinations of them. Boxed waste is divided into two general categories: newly generated waste, which is either TRU or LLW and retrievably stored TRU and suspect TRU.

Most newly generated LLW is packaged in 208-L drums, in B-25 boxes, and plywood boxes 2.4 m long, 1.2 m wide, and 1.2 m high. Newly generated TRU waste will be packaged in 208-L drums and in standard waste boxes (SWB). The B-25 box and the SWB are constructed of steel with walls approximately 4 mm thick. The maximum weight of each of these boxes and their contents (Figures 1 and 2) is limited to 1,800 kg.

Since 1970 all waste suspected of being or known to be TRU has been placed in retrievable storage pending availability of a permanent repository. Retrievably stored TRU waste at the Hanford Site is contained in 208-L drums and in boxes ranging from as small as a 1 m cube to as large as 6 m long, 2.7 m wide, and 4.3 m high. Boxes can be constructed of plywood, steel, or concrete. Box weight ranges from 100 kg to in excess of 36,000 kg. A single retrievable box may contain kilogram quantities of thorium or uranium and milligram quantities of plutonium. Some boxes contain milligrams of plutonium and tens of Curies of mixed fission products. Some boxes may contain

FIGURE 1. B-25 LARGE SCALE ARRAY WASTE CONTAINER.

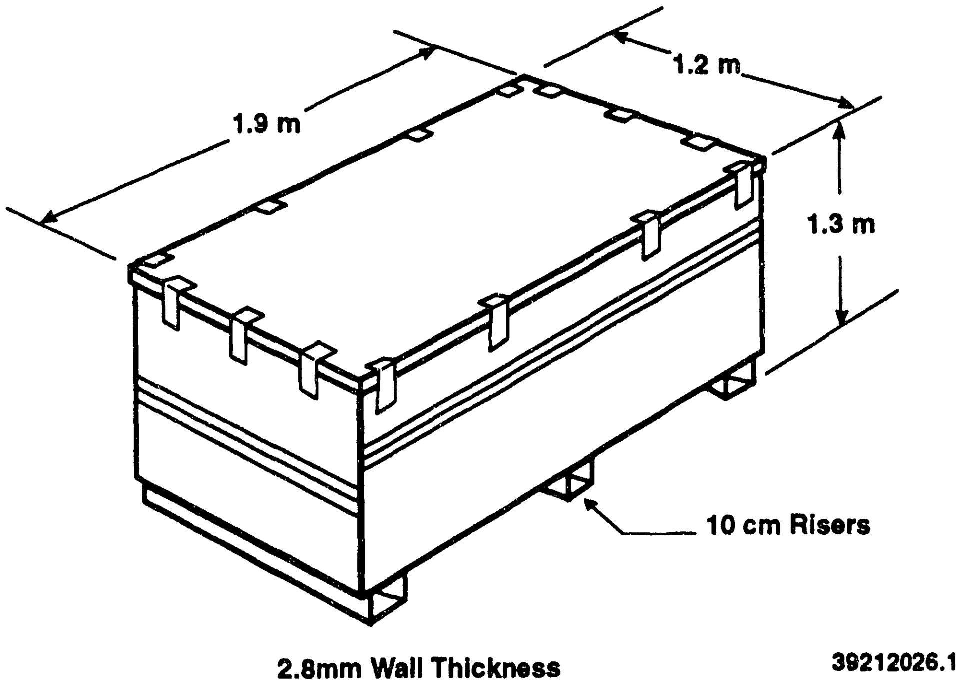
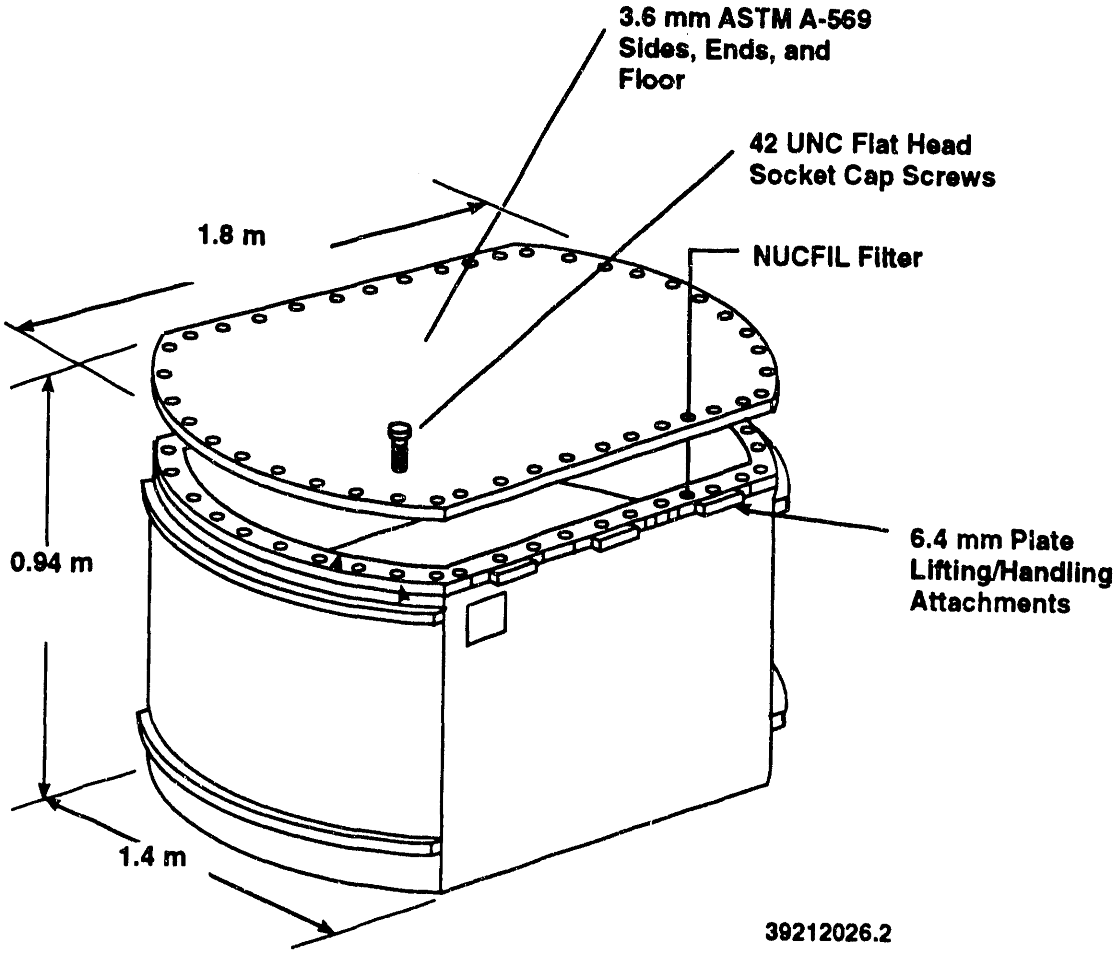


FIGURE 2. TRUPACT II STANDARD WASTE BOX
STYLE 1 - BOLTED CLOSURE.



several hundred grams of plutonium with various quantities of other actinides and fission products.

PROBLEMS WITH BOX NONDESTRUCTIVE ASSAY SYSTEMS

Drums of waste have an axis of symmetry; they can be rotated during assay to help average the effects of distribution and heterogeneity. Boxes have no such symmetry, they come in a greater variety of sizes and weights, and they have a greater diversity of content and material distribution than drums. It is for these reasons that the performance of earlier box NDA systems has been marginal. In addition, gamma energy analysis and passive neutron counting do not have the sensitivity to classify boxes of waste as LLW at the 100 nCi/g level, much less at the 10 nCi/g point for Category 1 LLW. Active neutron interrogation using the Los Alamos-developed Differential-Dieaway technique was attempted some years ago with mixed results. Active neutron interrogation remains the technique of choice for making low-level TRU measurements.

THE HANFORD SITE APPROACH TO BOX NONDESTRUCTIVE ASSAY SYSTEMS

In the past, WHC has used portable equipment to make neutron and gamma measurements on boxes. These measurements provided estimates of the fissile content of a box, which was enough to satisfy safety requirements. Because the measurements were not good enough to permit it, no attempt was made to classify the waste as LLW. All boxed waste that originated in areas where transuranic contamination was likely were treated as TRU.

A new portable system is being specified for retrieval operations. Combined neutron and gamma measurements will be made using "slab" detectors. These slabs will measure about 1.3 m wide x 1.5 m high x 20 cm thick, will weigh about 600 kg each, and will be used in pairs. The neutrons are counted using eight ^{3}He tubes in each slab. Gammas are counted with a pair of sodium iodide detectors in each slab. Neutron coincidences are counted to separate the random neutrons that originate from α,n reactions from those resulting from spontaneous fission.

In practice, the slabs will be placed on opposite sides of a box and neutrons from spontaneous fissions and gammas will be counted. The slabs will be moved from position to position as the counting process is repeated. This process continues until the whole surface of the box has been surveyed. When counting is complete an average will be calculated using algorithms that consider the patterns of counting and the isotopics of the known TRU constituents. The result will provide improved estimates of radionuclide contents , however, they will not provide the capability to make LLW/TRU determination. Figure 3 depicts the portable method.

THE BOX WASTE ASSAY SYSTEM

Permanent disposal of TRU waste requires the inventory of all radionuclides present in a container. Westinghouse Hanford Company has embarked upon a program capable of classifying and certifying the contents of boxed waste. Westinghouse Hanford Company has placed a contract with Pajarito Scientific Corporation of Los Alamos, New Mexico to design and construct a box waste assay system (BWAS). This system will combine passive and active neutron (PAN) measurements, and high sensitivity-low resolution sodium iodide gamma spectral measurements with high resolution (high-purity germanium [HPGe]) gamma spectral measurements.

The BWAS will handle boxes as large as 2 m long, 1.5 m wide, and 1.5 m high, and weighing as much as 4,500 kg. The system will make the TRU/LLW determination plus, within the LLW classification, the Category 1 and 3 determinations. The specified accuracies for the system are listed in Table 1.

A scanning approach that combines gamma transmission and neutron absorption/moderator corrections of gamma and neutron data will be used to calculate the quantities of the radionuclides present in a box of waste. The box will be loaded onto a turntable where a computer-controlled conveyor moves it into the assay chamber and increments it past the detectors. After the first active neutron and gamma assays are complete, the box is moved back onto the turntable where it is rotated 180° and re-enters the chamber where the active neutron and gamma assay process is repeated. The passive neutron measurement is made last. Figure 4 provides an overall view of the system.

For low-density waste (0.2 g/cm^3), the total time to complete an assay will not exceed 75 minutes. The heavier and more difficult matrices will require more time, but in no case will the total time be permitted to exceed 2 hours.

Neutron detectors, consisting of 0.6- and 1.8-m-long ^{3}He tubes, are arranged in single or double rows within polyethylene slabs in 8- or 15-tube "packages." There will be three 1.6-m packages in the roof and floor and two in the wall opposite the neutron generator. There will be two 1.8-m packages and two 0.6-m packages in the wall adjacent to the neutron generator. All of the ^{3}He tubes will be cadmium shielded.

Active neutron interrogation techniques make the low-level (10 and 100 nCi/g) TRU measurements. The box is incremented past a pulsed neutron generator that produces 10^6 14-MeV neutrons per pulse. The neutron generator is housed in a moderating assembly external to the assay cavity. The moderating assembly can be moved vertically in increments. This vertical incrementing action, coupled with stepping the box past the source, provides a crude picture of the location of the materials undergoing induced fission. This "active" phase provides a measure of the fissile isotopes (principally ^{235}U and ^{239}Pu) present in each segment of the waste.

FIGURE 3. SLAB COUNTER APPLICATION.

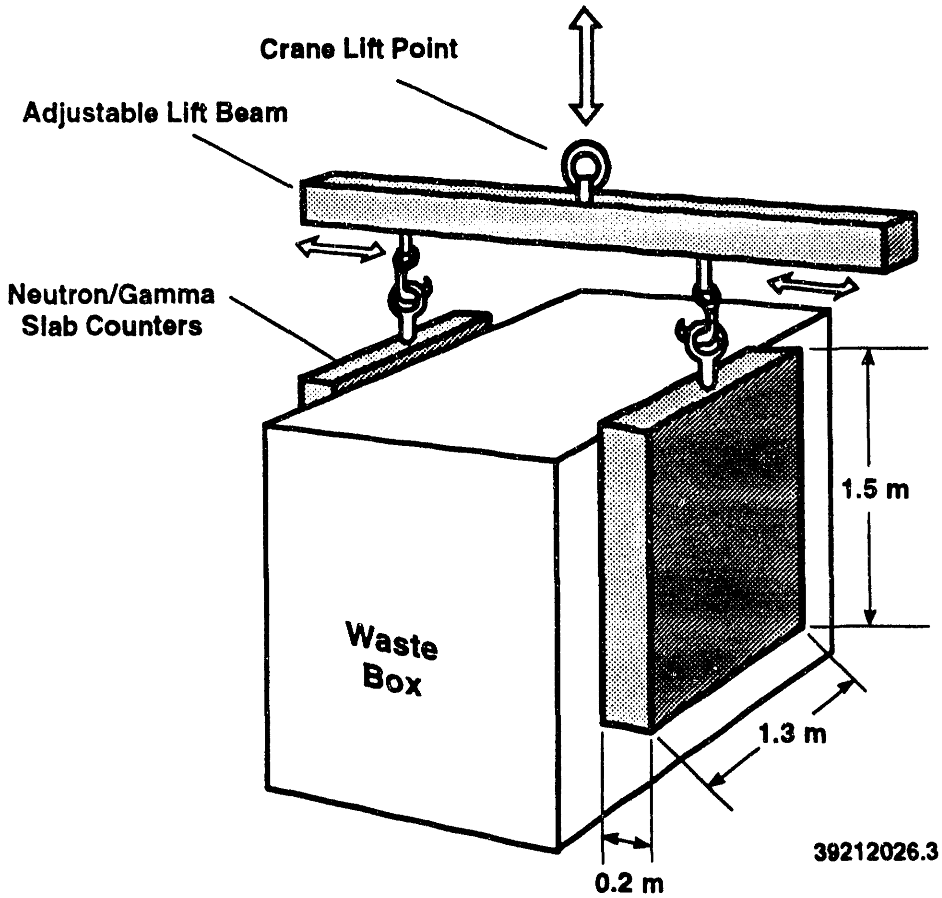


FIGURE 4. BOX WASTE ASSAY SYSTEM PERSPECTIVE.

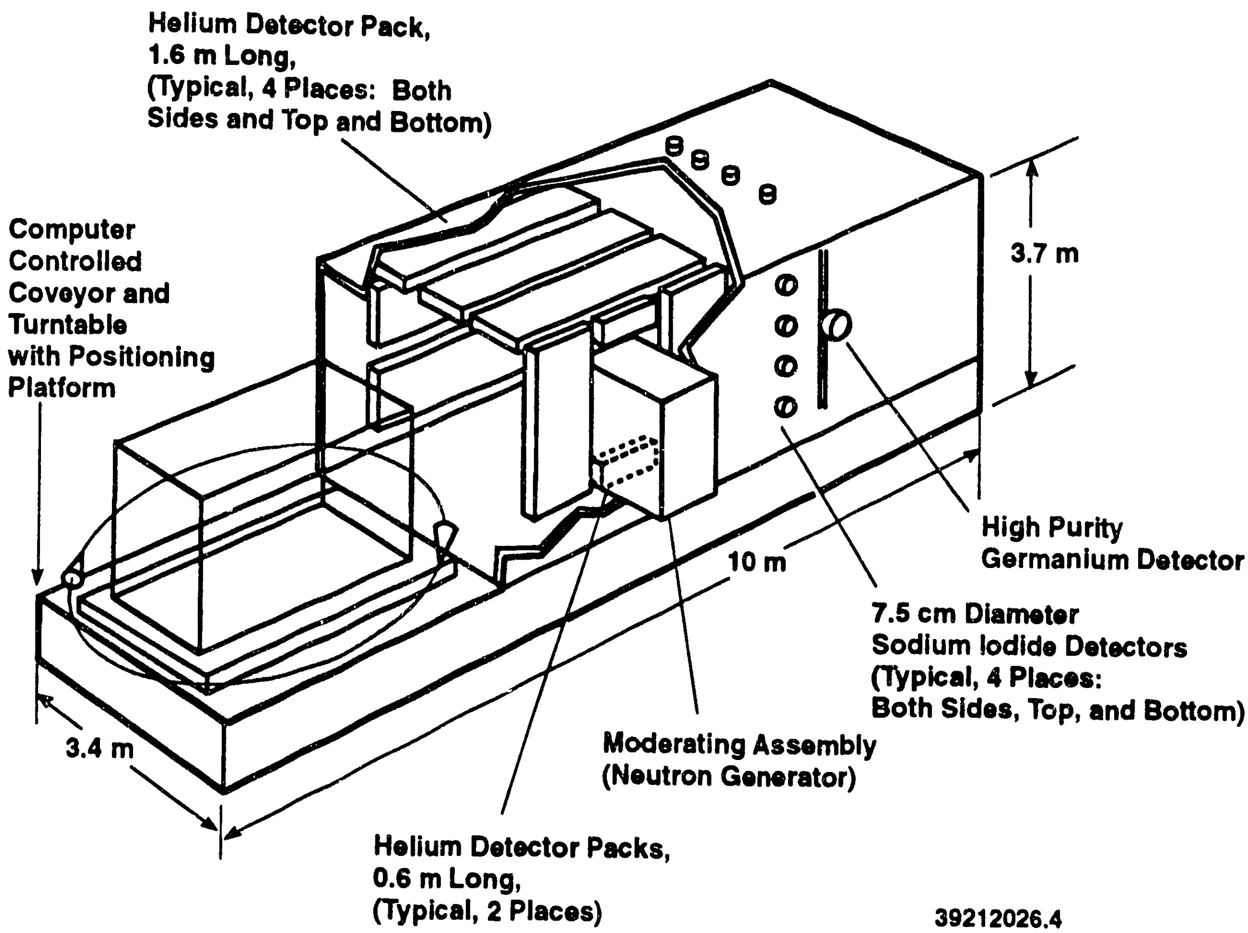


TABLE 1. BOXED WASTE ASSAY SYSTEM ACCURACY REQUIREMENTS

Measurement method	Measurement level	Waste contents	Measurement uncertainty
TRU active	10 nCi/g (plutonium)	Lab waste (0.1 g/cm^3)	± 5 nCi/g
TRU active	10 nCi/g (plutonium)	Moderate absorber (0.2 g/cm^3)	± 7 nCi/g
TRU active	100 nCi/g (plutonium)	Lab waste	± 40 nCi/g
TRU active	100 nCi/g (plutonium)	Moderate absorber	± 50 nCi/g
TRU passive	10 g (plutonium)	Lab waste	± 4 g
TRU passive	400 g (plutonium)	Moderate absorber	± 75 g
TRU GEA	400 g (plutonium)	Heavy matrix (1.3 g/cm^3)	± 40 g
Fission/active Prod GEA	20 pCi/g (^{60}Co, ^{137}Cs)	Lab waste	± 10 pCi/g
Fission/active Prod GEA	2 mCi (^{60}Co, ^{137}Cs)	Medium matrix (0.2 g/cm^3)	± 500 μCi/g

GEA = gamma energy analysis
TRU = transuranic.

Passive coincidence counting, using the ^{3}He detectors, measures the spontaneously fissioning actinides. Coincidence counting separates the multiple neutrons originating from spontaneous fission from single neutrons. The single neutrons originate from α,n reactions. The passive phase measures the ^{240}Pu present in plutonium waste.

The gamma energy analysis part of the system will measure gammas with energies from 40 keV to 2 MeV. Gamma measurements will be made at the same time as the active measurements, as the box is incremented through the chamber. Sixteen sodium iodide detectors, four each located on top, bottom, and both sides of the chamber, will be used to provide broad spectrum and medium resolution, but high-sensitivity measurements of all gamma emitters of consequence. One or two movable HPGe detectors will be used to provide high-resolution gamma source identification and may aid in the determination of plutonium isotopic composition. The data from the HPGe detectors also will aid the deconvolution of the sodium iodide measurement data. Two sets of four cobalt and cesium sources will provide horizontal and vertical transmission correction information.

The BWAS will, based on the measurements and characterization information, calculate the total fission and activation product inventory of the box. Where gamma measurements indicate the presence or absence of radionuclides contrary to characterization information, the BWAS will provide notice to the operator. The same holds true for ratios between those supplied by characterization information and those that may be measured. The BWAS will also compare the values derived from the three different measurement processes in calculating TRU content and to determine which of the three to use.

The original intent was to use the BWAS to perform NDA on all retrieved boxes within its size capability. Policies have evolved and because all retrieved TRU waste must be processed and repackaged to meet new criteria, the requirement for NDA after retrieval no longer exists. The BWAS will still have the ability to handle a wide range of box sizes and weights, but will only be used for the certification of newly generated LLW and TRU (including reprocessed retrieved TRU) boxed waste.

THE BOX WASTE ASSAY SYSTEM TEST PROGRAM

The BWAS will be installed in the U.S. Department of Energy's Hanford Site Waste Receiving and Processing (WRAP) facility in March 1995. After delivery to WHC, in September 1993 and before the WRAP installation, the system will be installed in a separate building at the Hanford Site for an extensive period of characterization, qualification, and calibration.

Testing will cover a wide range of box inventories, from the LLW Category 1 limits, at 10 nCi/g, to the high loadings where box content may reach as much as 300 g of plutonium. Fission and activation product inventories will also be varied over a wide range.

The hundred-gram quantities of plutonium required for system testing and calibration presented many practical problems, of which the most difficult was physical security. To eliminate the security issue, it was decided to load test boxes with previously certified drums of TRU waste. These drums will have plutonium contents ranging from less than a gram to quantities in excess of a hundred grams or more. As a result of the use of drums of waste, the testing program has become much simpler and total test program cost has been reduced.

Discrete sources of cesium, cobalt, and europium, and multikilogram quantities of uranium will be located in different places throughout a test box and an assay will be performed. Drums and sources will then be rearranged within the box and the assay will be repeated. Many different TRU drums, containing from milligram to several hundred gram quantities of plutonium, will be used in different combinations and in three different box types. These boxes will be the B-25, the SWB, and a plywood box measuring 2 m by 1.5 m by 1.5 m. The result will be a system whose performance envelope is well defined and, having used known sources, one which is well calibrated.

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