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WASTE CHARACTERIZATION: WHAT'S ON SECOND?

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## WASTE CHARACTERIZATION: WHAT'S ON SECOND?

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### INTRODUCTION

Waste characterization is the process whereby the physical properties and chemical composition of waste are determined. Waste characterization is an important element of a waste certification program in that it provides information which is necessary to certify that waste meets the acceptance criteria for storage, treatment, or disposal. Department of Energy (DOE) Order 5820.2A and WIPP-DOE-069 list and describe the germane waste form, package, and container criteria for the storage of both solid low-level waste (SLLW) and transuranic (TRU) waste, including chemical composition and compatibility, hazardous material content (e.g., lead), fissile material content, radioisotopic inventory, particulate content, equivalent alpha activity, thermal heat output, and absence of free liquids, explosives, and compressed gases.

At the Oak Ridge National Laboratory (ORNL), the responsibility for waste characterization begins with the individual or individuals who generate the waste. The generator must be able to document the type and estimate the quantity of various materials (e.g., waste forms - physical characteristics, chemical composition, hazardous materials, major radioisotopes) which have been placed into the waste container. Analyses of process flow sheets and a statistically valid sampling program can provide much of the required information as well as a documented level of confidence in the acquired data. A program is being instituted in which major generator facilities perform radionuclide assay of small packets of waste prior to being placed into a waste drum.

Prior to removing a filled waste drum from a generator facility, an inspection of the container, documentation, and related items is performed. Table 1 presents a summary of the items reviewed. After the inspection, the waste containers are transferred to a central waste certification facility, the Waste Examination Assay Facility (WEAF). The WEAF serves both to verify a portion of the generator's waste characterization data, such as the radioisotope inventory and absence of free liquids and compressed gases, and to perform certain other characterization measurements, such as fissile content and thermal heat output.

### WASTE CHARACTERIZATION AT THE GENERATOR FACILITY

The process of characterizing wastes must begin at the point where waste is generated. In order to certify that a given waste package meets the applicable waste acceptance criteria for whatever treatment, storage, or disposal facility that is accepting the waste, the process in which the waste is generated must be examined.

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Table 1. Checklist Items for Use at Drum Pick-Up

- a. Radiation tag (UCN-2785) attached and properly filled out.
- b. Documentation complete (UCN-2822), log-in sheet, accountability transfer form, if required.
- c. Check for physical damage (visual).
- d. Bolt ring and gasket properly installed and in place.
- e. Closure bolt torqued to 45 ft-lbs.
- f. Jam nut in proper location and tight.
- g. Drum checked to be 17-H.
- h. Bar code installed and in place.

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In addition to characterization of waste according to its radionuclide content, the waste generator must also provide assurance that constituents which are prohibited by the waste acceptance criteria are being excluded. Table 2 shows a list of several items which must be excluded from SLLW, based on current and draft waste acceptance criteria for the Oak Ridge Reservation. Additional items may be included on the prohibition list as the waste acceptance criteria for new treatment, storage, and disposal facilities are developed.

According to guidance given by the Nuclear Regulatory Commission (NRC) and reinforced in DOE Order 5820.2A, the acceptable techniques for characterizing wastes are:

1. Direct or indirect measurement. Direct measurement would include techniques such as gamma spectroscopy or sampling the waste stream. Indirect methods may include using gross measurements, such as surface exposure rates, in conjunction with scaling factors. The indirect methods would be applicable to a well defined waste stream and would have to be substantiated through use of direct measurement techniques.
2. Material accountability. Essentially a bookkeeping method, accountability uses the difference between material input for a process and the amount of that material that is present in the product, including the amounts known to be consumed or converted in the process and the quantity remaining as contamination on surfaces or inside pipes and ducts. To be used as a basis for waste characterization, material accountability would have to be substantiated through direct measurement techniques.

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Table 2. Constituents Prohibited from Solid Low-Level Waste

free liquids  
oils and oily wastes  
PCB contaminated materials  
pesticides (herbicides, insecticides, etc.)  
compressed gases  
hazardous waste as defined in RCRA  
  arsenic  
  barium  
  cadmium  
  chromium  
  lead  
  mercury  
  selenium  
  silver  
  toxicity characteristic compounds  
  ignitable waste  
  reactive waste  
  corrosive waste  
  cyanide or sulfur bearing waste  
pyrophoric materials  
chelating agents in excess of 0.1% by weight  
respirable particles in excess of 1% by weight  
pathogenic, infectious, toxic, or poisonous materials  
explosive materials

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3. Process knowledge. Through an understanding of the material input to a process and the manner in which the material is manipulated or handled, the waste may be characterized by use of a model which accounts for the quantities which will be included in the waste from that process. The model may be rather simple or complex, depending on the process involved, but also must be substantiated.

At ORNL, a study project is being conducted on one of the major low-level waste (LLW) streams to develop a measurement procedure to segregate uncontaminated waste from LLW and to determine the concentration of beta and gamma emitting radionuclides in the waste. The first step in the procedure is the segregation of waste streams.

At the facility, there are several waste streams contaminated with radionuclides that emit only weak beta particles, such as  $^3\text{H}$ . The procedure being developed is not designed to detect these weak beta emissions. Therefore, the initial stage of the process must be to segregate those waste streams from the streams that can be used in the demonstration project. To accomplish this, a series of administrative controls and controls on the waste generating processes are being instituted to ensure that the streams do not become commingled.

After separating the beta-gamma emitters from the unsuitable wastes, the material will be screened for radioactivity in an instrument called a Waste Curie Monitor. This device consists of a small cabinet lined on four sides with sheets of plastic scintillator material. A plastic bag containing the waste to be monitored is placed inside the cabinet, the door is closed, and a short count (on the order of a few minutes) is made. The plastic scintillators provide an integrated count of all the gamma and hard beta emissions from the waste. Provided the lower limits of detection are chosen appropriately, this screening step will indicate whether or not the waste is contaminated. Since the current operation of this generator facility assumes that all wastes coming from the process area is contaminated, using this instrument should be able to reduce the amount of waste that must be handled as radioactive.

If the waste is contaminated, further analysis will be performed. For gamma emitting nuclides, a germanium-based gamma spectrometry instrument will be used to quantify the radionuclides in the waste. For the radionuclides that emit only beta particles, the waste streams have been chosen so that a defined ratio exists between the quantity of pure beta and beta-gamma emitters. By periodically verifying the ratio, the concentration of all radionuclides in this particular waste stream can be determined through this procedure.

Other characterization methods used by waste generators rely heavily on process knowledge to estimate the quantity of radionuclides in the waste. Although the application of process knowledge to waste characterization involves subjective evaluation, the estimates and procedures for performing the estimates are well documented. Each small package of LLW placed into a waste container is recorded on a log sheet maintained by the generator. The log description of the waste packet includes the date generated, the person placing the waste in the container, estimates of the radionuclide content, physical measurement of the surface radiation levels, and a general description of the waste form. By maintaining this running documentation for each container, the contents are more easily characterized than would be possible by examination of the bulk form.

Additional documentation is prepared when the waste generator needs to transfer the container. A Request for Disposal form is prepared which summarizes and totals the information contained on the individual log records. Both the Request for Disposal and the log sheets are a part of the documentation which accompanies the waste container to assay, treatment, storage, and disposal facilities. Further characterization is performed at the central facilities.

## WASTE CHARACTERIZATION SAMPLING PROGRAM

A significant quantity of solid contact-handled transuranic (CH-TRU) and LLW packaged in 208 l (55 gallon) drums is stored at ORNL. The characterization data available for many of these drums was obtained under different criteria and procedures than are now in force. Therefore, some of the data needed to certify the wastes against the appropriate acceptance criteria either do not exist, or do not provide sufficient accuracy, precision, and level of detail. ORNL has not yet instituted a formal waste characterization sampling program for these drums. However, a CH-TRU waste drum sampling program was conceived in the early 1980's to be used as an independent validation of the passive/active neutron (PAN) assay measurements made at the WEA. [References 1 and 2]

Under the sampling plan, a glove box facility was constructed to obtain samples from selected CH-TRU waste drums. The contents of the drums would be emptied into a glove box and segregated into four categories: cellulose, plastics, glass and ceramics, and metals. Each type of material, excluding metals, would be weighed and homogenized. A specified number of samples, approximately 100 per drum, of different waste types, designated by "content codes" (e.g., decontamination debris, contaminated equipment, dry solids) would be collected and transferred out of the glove box. The collected samples would then be assayed by gamma-ray spectrometry and the results compared to the PAN assay measurements. The proportion of agreement between the matched observations would form the basis for validation of the neutron assay technique.

The statistical sampling program would begin by obtaining a stratified random sample of drums. (Number of drums = n) The variance of the proportion of agreement would also be estimated. The question remaining is how large should n be? The probability that the absolute difference between the estimate of proportion of agreement and the true proportion of agreement is greater than or equal to an arbitrary factor supplied by the experimenter is given by the following equation:

$$\Pr ( |P - P| \geq d ) = \alpha$$

Alpha represents the confidence interval,  $100 * (1 - \alpha)\%$  (e.g.,  $\alpha = 0.01$  corresponds to a 99 percent confidence interval), while d represents the proportion of agreement sought by the experimenter (e.g., for  $d = 0.2$ , the experimenter seeks a relative difference between the two measurements of 20 percent).

For example, with a choice of  $\alpha = 0.05$  and  $d = 0.08$ , 150 drums out of a population of 1,800 would be destructively sampled. There are not sufficient numbers of drums in certain waste content codes for a statistically valid program. Consequently, all drums within that waste content code were to be sampled. Other content codes encompass a large number of drums. A stratified random sample of these drums at the generator sites was chosen to ensure that a wide range of drum activity concentrations would be included in the sample. The strata limits were determined by the cumulative square root of the frequency method [Reference 1].

The number of samples to be collected from each waste category (i.e., metals, plastics, cellulose, glass and ceramics) within a selected drum is based upon the width of the confidence interval desired, determined by the size of the difference between the assay measurement results for the two methods which would be considered significant. The values of proportion, by weight, of the waste categories in the drum and the variance of the activity concentration distribution of an isotope within a waste category could be estimated from a pilot sample.

The observations from the destructive and nondestructive assay can then be used to determine if the nondestructive assay (NDA) method provides accurate estimates of the mean concentration of the isotopes present in the waste container. At this time, the sampling program has not been instituted at ORNL.

#### CENTRAL WASTE CERTIFICATION FACILITY

The ORNL central waste certification facility is the WEAFF, Building 7824, located in Solid Waste Storage Area (SWSA) 5. It was established in 1982 as the ORNL central certification facility for the NDA and nondestructive examination (NDE) of CH-TRU waste. In November of 1984, an NDE instrument, real-time radiography (RTR), was added to the WEAFF instrument inventory for examination of physical contents of waste drums. Currently, all CH-TRU and LLW drums generated at ORNL are examined using the RTR unit. In addition, all CH-TRU waste drums are assayed with the PAN and segmented gamma scanning (SGS) systems described below. Only those LLW drums generated at facilities which also generate CH-TRU or remote-handled (RH) TRU waste are currently being assayed. However, in the development of the LLW certification program, plans are being made for all LLW drums to be assayed, primarily due to new requirements imposed in DOE Order 5820.2A.

The route of waste drums through the WEAFF begins with an RTR examination, followed by NDA examinations, if required. If a drum is found to contain any items which are prohibited by waste acceptance criteria, called "nonconformance" items, or if the drum itself is found to be defective, the drum is color coded with orange tape and a "REJECT" tag is affixed to it. The drum is then returned to the waste generator for repackaging. A nonconformance report (NCR) is written and accompanies the drum. Copies of the NCR are forwarded to the appropriate quality assurance representatives and a copy is retained at the WEAFF. The NCR is also entered into a computerized tracking system where resolution of and corrective actions for the NCR are monitored by the ORNL Quality Department. Table 3 shows a list of nonconformance items which can be detected by RTR examination.

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Table 3. Nonconformance Items Detectable by Real-Time Radiography Examination

Free liquids  
Compressed gases  
(compressed gas cylinders and aerosol cans),  
Lead  
Thin-walled primary waste containers  
Improperly-positioned lid gaskets

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The CH-TRU waste drums are examined to ensure compliance with Waste Acceptance Criteria (WAC) of the Waste Isolation Pilot Plant (WIPP) and ORNL [Reference 3]. After the examination is completed and shows compliance, the CH-TRU waste drums are retrievably stored for eventual shipment to the DOE geological TRU waste repository, the WIPP, located near Carlsbad, New Mexico.

The present routine operation of the WEAF includes NDA and NDE of solid CH-TRU waste and SLLW packaged in drums, certification of these wastes against waste acceptance criteria, issuance of nonconformance reports for drums which fail RTR inspection, and preparation and maintenance of data bases on drums of waste.

#### REAL-TIME RADIOGRAPHY

The RTR, a TFI Corporation system, uses a 320 kV<sub>cp</sub> (maximum) variable x-ray tube head. Most waste drum examinations are conducted using an applied voltage of 100 to 150 kV. The duration of an RTR examination of a typical laboratory waste drum containing plastics, cellulose, and small amounts of metal is approximately one-half hour.

The ORNL RTR radiation control enclosure, or examination chamber, dimensions are 5 ft. x 8 ft. x 8 ft. The examination chamber walls are fabricated of laminated steel, plywood, and elemental lead. The 3/4 in. thick plywood is bonded to the lead sheeting of the side walls and top for structural support and for securing the panels to the chamber frame. The panels of plywood and lead are sandwiched between two sheets of 1/16 in. steel.

The primary shielding for the wall and top of the examination chamber are 1/2 in. thick lead sheets. The side of the chamber to which the x-ray beam is directed is constructed with 3/4 in. thick lead. The additional shielding is required to terminate the x-ray beam and minimize the x-ray penetration to the areas adjacent to the examination chamber.

The chamber door is electrically driven, its motion governed by electrical limit switches at both the open and closed positions. The chamber door contains a 12 inch square leaded glass window and a Bureau of Radiological Health (BRH)-approved safety interlock which interrupts current to the transformers should the door accidentally open.

3. detection limit: a stated limiting value which designates the lowest concentration or mass that can be estimated or determined with confidence and which is specific to the analytical procedure used.
4. calibration: the determination of the values of the significant parameters by comparison with values indicated by a reference instrument or by a set of reference standards.

#### SEGMENTED GAMMA SCANNING (SGS)

The WEAFF SGS unit is a modified Canberra Model 2220-B. The photon detector used is a high-resolution, hyperpure germanium (HPGe) solid state detector.

Originally, the system was capable of quantitatively assaying  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , exclusively. After completion of recent modifications, the instrument can now acquire data and provide estimates of the quantities of any gamma-emitting isotopes present in amounts above the detection limit. The modifications included expansion of the nuclide search library, using a higher-capacity multi-channel analyzer, rewriting the peak search software, and using a mixed oxide  $^{152/154}\text{Eu}$  transmission source.

Besides fission product isotopes (e.g.,  $^{60}\text{Co}$  and  $^{137}\text{Cs}$ ), the number of individual TRU isotopes or their daughters that can be assayed with SGS is relatively large, with  $^{233}\text{U}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{237}\text{Np}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$  among the more common ones. In each case, one or more characteristic moderate-to-high energy gamma rays are emitted in sufficient intensity to permit estimates of quantities in low-to-moderate density waste packages as large as 208 L drums.

To minimize assay errors due to axial inhomogeneities, assays are performed in segments along a waste package's vertical axis. Radial inhomogeneities are minimized by rotating the drum during the assay measurement. The detector is shielded in such a manner so as to allow the waste drum to be scanned in segments. Typically 10 to 20 segments are used.

Gamma-ray attenuation is measured for each segment with a transmission source usually located on the detector casing. The energy of this source is selected to match that of the gamma-ray energies being measured. ORNL uses a mixed  $^{152}\text{Eu}/^{154}\text{Eu}$  oxide source for its large array of gamma-emitting radioisotopes (50 keV to 1600 keV), while  $^{75}\text{Se}$  is typically used for  $^{239}\text{Pu}$  assays.

Counting electronics allow dynamic counting rate ranges of factors of  $10^4$  to  $10^5$  or more. Dead-time corrections are measured with a second small, low-energy source positioned near the detector. Waste packages are automatically rotated about their vertical axes and cycled through the required segment heights with standardized, computer-controlled electronic motors and precision mechanical turntable and elevator hardware.

Some of the factors affecting assay measurements are particle self-absorption and nonhomogeneity of the assayed item ("lumping"). Two conditions must be met to optimize assay results: (1) the particles containing the radionuclide must be small to minimize self-absorption of emitted gamma radiation, and (2) the mixture of material within a package segment must be reasonably uniform in order to apply an attenuation correction factor. The attenuation correction factor is computed from a single measurement of gamma ray transmission through the segment. Variations in waste composition and density within a vertical segment lead to indeterminate errors. Such variations should be minimized through strict scrap and waste segregation procedures.

A combination of analytical error analysis [Reference 11] and experimental usage over many years has determined that transmission factors greater than or equal to 0.5 percent are required for accurate SGS assays, although some researchers have reported lower transmission factors. Subject to that limitation, any chemical or physical waste form may be assayed using the SGS method. The physical density of a waste package, under the limitation of transmission factor size, depends greatly on the package size (i.e., the radial distance from the gamma-emitting source(s) to detector). Four-liter packages having densities as high as 2 g/cm<sup>3</sup> meet the criterion, whereas 208 L packages are limited to densities of 0.5 g/cm<sup>3</sup> or less. To assure compliance with these limits, the SGS software package includes an automatic warning, recorded on paper and magnetic media for archival purposes, indicating when the transmission factor for any sector falls below the prescribed limiting value. The routine practice is to calculate a contribution from that sector based on the lower-limit transmission (e.g., 0.5 percent).

The reason for maintaining the assay value, rather than disregarding it, is that most SGS transmission failures occur for only one sector out of the 10 to 20 drum sectors assayed. This sector, on the average, contains only a small fraction of the waste drum's total TRU inventory of gamma-emitting isotopes. Estimating the TRU content for one, two, or three such failed segments in this fashion results in only a small overall assay error for the waste drum. Since the SGS assay value for a transmission failure is truly a lower limit, and passive neutron assays generally provide upper limit assay values, especially for TRU isotopes, the combination of SGS and passive neutron assay methods tends to bracket the true concentration of radionuclides in the waste.

Some waste matrices are inherently unsuitable for SGS analysis. Such forms may contain 'lumps' of radionuclides, that is, radionuclides contained in small volumes of waste having a localized density substantially different from the bulk density of the rest of the container. The particle dimensions that constitute a lump vary with the energy of the radiation being measured. For example, a plutonium metal sphere 0.02 cm in diameter will absorb approximately 4% of the 414 keV, <sup>239</sup>Pu gamma rays produced. Approximately 15% of the 186 keV gamma rays of <sup>235</sup>U will be absorbed in a uranium metal sphere of the same diameter.

As mentioned previously, another condition that will cause measurement problems occurs in containers with several irregular regions, highly variable in density, that prevent the calculation of a valid attenuation correction based on the transmission measurement. In case of such a condition, an analytical method less sensitive to nuclide and matrix densities, such as passive neutron coincidence counting (PNCC), should be employed.

Careful inspection of the transmission and nuclide peak areas for each segment may provide clues when a measurement should be suspect. Sudden, discontinuous changes in the transmission values for adjacent segments or high counts for isolated segments are examples of signals indicating possible problem items.

### SGS Assay Precision and Bias

Repeatability of results in typical SGS systems, when operated and calibrated according to the recommended procedures, is limited only by radioactive decay statistics and related counting errors. Counting statistics, in turn, are a strong function of TRU isotopic loading and counting time.

Reference 7 discusses SGS precision and bias in detail. The precision of a SGS assay is a function of the precision of the measured peak areas for each segment. The precision of an assay is normally better when the following conditions can be obtained:

- Long count time
- High transmission source activity
- Low gamma attenuation at the energy being measured

The precision of an assay is not strongly related to the presence of an ideal matrix and nuclide density. However, measurement bias depends primarily on small particle size and homogeneity. Negative bias will be encountered when the nuclide is present in lumps that attenuate their own radiation to a greater extent than the surrounding material. Positive bias can result from low transmission items with over-corrected end effects. Items containing high-density areas may be biased either high or low or be unbiased, depending on the relative position of the high density area and the nuclide of interest. In the majority of measurement situations, however, it is expected that when biases exist, measurement results will be lower than true values.

SGS and destructive assay comparison studies of several waste forms indicate SGS assay biases of 10 percent or better at the 95 percent confidence level [Reference 12]. Assay biases for low-density waste matrices contained in 208 L drum packages are 5 percent or better. In small packages, SGS assay biases of better than 0.5 percent have been reported [Reference 5]. The basic assay formalism associated with the SGS method, that is, transmission correction and the use of small segments, is conducive to very accurate results if recommended procedures are correctly followed. Heterogeneous matrices and isotopic concentration can have a severe effect on assay bias.

## WEAF NONDESTRUCTIVE ASSAY INSTRUMENTS

### Assay Overview

Two NDA instruments are used at the WEAF: PAN assay and SGS. ANSI N15.20 [Reference 4] defines NDA to be:

"The observation of spontaneous or stimulated nuclear radiations, interpreted to estimate the content of one or more nuclides of interest in the item assayed, without affecting the physical or chemical form of the material.

active assay - Assay based on the observation of radiation(s) induced by irradiation from an external source.

passive assay - Assay based on the observation of naturally occurring or spontaneous nuclear radiation(s)."

The NRC in NRC Regulatory Guide 5.11 [Reference 5] describes the applicable NDA passive measurements:

"Radiations attributable to alpha particle decay, to gamma-ray transitions following alpha and beta particle decay, and to spontaneous fission have served as the basis for practical passive NDA measurements."

Gamma rays, neutrons, and alpha particles, as well as other subatomic particles, are emitted by various radioisotopes. NDA techniques based on detection of each emitted radiation have been developed and used for CH-TRU and LLW bulk-waste assay.

The passive gamma (SGS) and passive neutron coincidence counting (passive portion of the PAN) methods are techniques which are described by the American Society for Testing of Materials (ASTM), American National Standards Institute (ANSI), NRC, and American Society of Mechanical Engineers (ASME) standards, guidelines, and regulations.

Characteristics of any assay measurement include precision, bias, detection limit. Proper calibration methods must also be employed to ensure unbiased assay results are obtained. Definitions of each of the above terms are given below and were obtained from reference 6.

1. precision: a generic term used to describe the dispersion of a set of measured values.
2. bias: a persistent positive or negative deviation of the method average from the correct value or accepted reference value. (Also referred to as "constant" or "systematic error.")

## WEAF PASSIVE-ACTIVE NEUTRON ASSAY SYSTEM

PAN assay systems consist of two independent neutron assay units: passive and active. The combination of passive and active neutron assays within a common system provides a unique set of information.

The passive assay method described below is an adaptation of the PNCC method using "self-measurement" matrix corrections. Two complete passive assay detection systems are maintained with separate counting electronics. The passive coincidence measurement provides quantitative information on isotopes with even mass numbers, such as  $^{240}\text{Pu}$ , which are present in the waste container. The passive singles neutron count rate, which is the difference between total neutron rate and that due to spontaneous fission events, provides semi-quantitative information on alpha particle emitters present in the waste container, such as  $^{241}\text{Am}$ . The active assay provides quantitative information on the  $^{239}\text{Pu}$  and other fissile isotope constituents of the waste.

For example, for weapons-grade plutonium, the passive coincidence and active assays provide independent total Pu assay values. This fact has been extremely important in verifying the accuracy or determining the bias of the PAN assay measurement technique. [Reference 8]

### PAN Instrumentation:

A cross-sectional view of the ORNL PAN detection system, showing the schematic "interwoven" layout of the two distinct types of neutron detection packages, bare  $^3\text{He}$  and cadmium (Cd)-shielded  $^3\text{He}$  detector tubes, is shown in Figure 1.

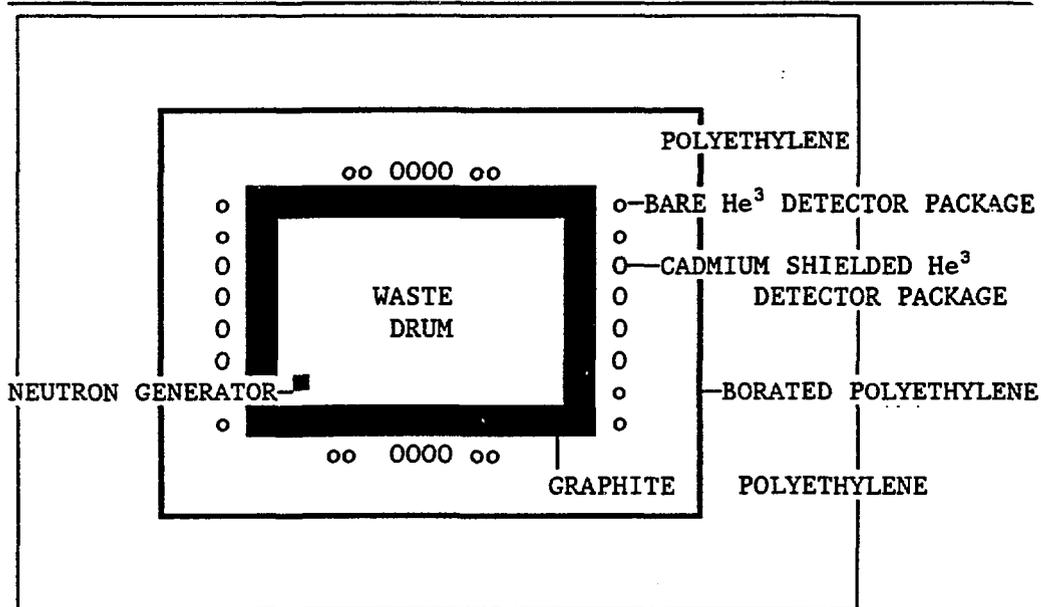


Figure 1. Cross Section View of Passive/Active Neutron Assay System

### Passive Assay Portion

The passive portion of the PAN assay system uses the two types of detectors to determine a moderator index (MI) used to make systematic matrix corrections, and optimize counting statistics depending on the actual relative neutron sources encountered.

For low count-rate waste containers all counts detected by the neutron detectors are summed to yield the lowest assay limit of detection possible. All detector count rates, acquired by both bare and shielded detectors, are summed electronically to obtain a "System Totals" neutron detection efficiency of approximately 12 percent.

For waste containers with higher TRU isotopic loadings (e.g., 100 g or more) coupled with strong (alpha, n) backgrounds, the cadmium-shielded detectors are summed independently, and the "Shielded Totals" count rate is formed with a resulting neutron detection efficiency of 2.9 percent. However, this detection package possesses a much faster "die-away" or "neutron-collection" time, approximately 15 microseconds, about six times faster than that of the slower "Systems Totals". At low count rates the slower collection time is of no consequence, since accidental coincidences due to (alpha, n) reactions are small. Therefore, the Systems Totals provides not only a more sensitive but also statistically more precise passive assay measurement.

At higher data collection rates this is no longer true. The faster die-away time of the Shielded Totals becomes more important statistically than the neutron detection efficiency. As a consequence, at high neutron count rates the Shielded Totals Coincidence rate is used to obtain the more precise passive assay measurement value.

The cross-over count rate (i.e., the count rate at which the assay measurement value obtained by the Shielded Totals supplants the Systems Totals) has been experimentally determined to be approximately 2000 counts per second (cps Systems Total count rate). This value is used in the assay algorithm. There is a substantial range in which either Systems Coincidence or Shielded Coincidence rates both provide precise assay values.

### Active Assay Portion

The active portion of PAN systems performs a high-sensitivity, pulsed thermal neutron interrogation assay of waste drums. As shown schematically in Figure 1 a small 14-Mev neutron generator has been placed within the assay chamber between the waste drum and moderating walls to provide short pulses (5-10 microseconds) of high-energy interrogating neutrons. In approximately 0.5 ms all original fast neutrons in this interrogating pulse have been thermalized by multiple collisions with the graphite and polyethylene walls and with moderating materials within the waste drum. This "thermalized interrogating pulse" persists with a half-life of about 400 us, during which time induced fissions within the waste drum are produced, primarily in <sup>239</sup>Pu or other fissile isotopes. These events, in turn, result in a burst of prompt-fission spectrum neutrons emitted by each fissioning nucleus.

The cadmium-shielded detection packages have been designed to reject an external thermal neutron flux to 1 part in  $10^7$ , but to respond sensitively to fission spectrum neutrons. The same shielded detector packages shown in Figure 1 detect about 10 percent of all induced fission events produced within typical waste drums.

An additional measurement feature not shown in Figure 1, but discussed at length in reference 8, is the set of thermal flux monitors, one cadmium-shielded and collimated and the other bare, that are also positioned inside the assay chamber between the waste drum and the moderating walls. As discussed at length in reference 8, the ratio of these flux monitors is highly sensitive to the total neutron absorption characteristics of the waste drum contents. This ratio is used to form a drum "Absorption Index" (AI).

### PAN Assay Matrix Corrections

Two types of matrix effects can interfere with the active neutron measurements: absorption and moderation [Reference 8]. The absorption effects occur almost entirely as an attenuation of the interrogating thermal neutrons, caused by the presence of various neutron poisons (e.g., boron, cadmium, etc.) within the waste matrix.

Moderation effects occur at two stages of the measurement. The original burst of 14-Mev neutrons can be moderated to a considerable extent during passage through the waste matrix. Generally, this results in a larger thermal neutron interrogation flux than would have been produced in the absence of matrix. After the interrogation flux has produced fission reactions within the waste matrix, the same moderating materials can attenuate the prompt-fission signal neutrons, resulting in a decrease in observed response relative to the no-matrix case. This attenuation of fission-neutrons is also the primary matrix effect for the passive measurement.

The approach to matrix corrections has been to base corrections on measured quantities determined as adjuncts to the primary active and passive TRU assay measurements. The systematic matrix correction algorithm is based on an analytic fit to assay measurements obtained for different positions of the source within a matrix drum. These analytic fits then provide estimates of uncertainty for the active and passive assay data.

The absorption matrix correction approach used by the PAN systems employs a ratio of an unshielded in-chamber flux monitor to a cadmium-collimated, in-chamber flux monitor (designated the barrel flux monitor). This ratio is called the AI. The barrel flux monitor detects those neutrons which have undergone drum matrix interactions. The ratio of the monitors strongly reflects the neutronic properties of the matrix.

#### Absorption Index

$$= \frac{[\text{flux monitor response (0.7-4.7ms)}]}{[\text{barrel flux monitor response (0.7-4.7ms)}]} \quad (1)$$

The moderator index depends upon the responses of the two detection systems (cadmium-shielded and bare) to moderated neutrons. The shielded detectors are insensitive to thermal neutrons, while the bare detectors are very sensitive to the thermal neutron flux. In turn, the thermalized fraction depends very strongly on the moderator density of the matrix. To use this relationship in obtaining matrix correction factors, the ratio is normalized so that a value of zero is obtained when no moderator is present and a small correction is made to account for self-absorption effects.

Moderator Index (MI)

$$= \{1 - [(\text{shielded totals})/(\text{system totals})]/A_0\} \times \{A_1 + A_2 \times \ln(\text{absorption index})\} \quad (2)$$

The term within the first set of brackets is the basic raw spectral data. The term within the second set of brackets is the correction term for matrix absorption effects. The same MI values are used for both active and passive matrix corrections.

In order to obtain data to construct the analytical models of matrix correction factors, nineteen simulated waste matrices were fabricated [Reference 8] and active and passive calibration standards were placed in known locations throughout the waste matrix drums. Both active and passive assay matrix response measurements were obtained as a function of position (radius,  $r$ , and height,  $z$ ) of the standards. The resulting matrix response values varied smoothly as a function of  $r$  and  $z$ . These studies determined that the systematic effects are due only to gross neutron absorber and moderator amounts and are independent of the actual nature of the materials themselves. That is, a drum filled with Rashig rings (borated glass) produces the same responses as a drum filled with vermiculite mixed with an equally absorbing amount of borax.

Most of the observed distributions have been found to fit a power law as given in equation 3:

$$y = A + Br^N \quad (3)$$

where  $A$ ,  $B$ , and  $N$  are the curve fit parameters and  $r$  is the drum radius.

Volume-weighted average values were calculated using this equation, representing the most probable measurement result for either a totally uniform or a totally random distribution of source material within the matrix.

The matrix correction factor (MCFA) for an active assay measurement is a function of the AI and MI.

$$\text{MCFA} = \text{MCFA}(\text{AI}) \times \text{MCFA}(\text{MI}) \quad (4)$$

The MCFA values were fit to the power law (equation 3) as a function of their AI values for the 19 simulated waste matrices. The following set of equations describing the absorption portion of the active assay matrix correction factor were obtained:

$$\text{MCFA(AI)} = 1.00 \quad (5)$$

for the AI less than or equal to 2.72, and

$$\text{MCFA(AI)} = 0.54x(\text{AI})^{0.612} \quad (6)$$

for the AI greater than 2.72.

The moderator portion, MCFA(MI), of the active assay matrix correction factor is obtained by dividing the total measured MCFA values by the calculated MCFA(AI) values obtained in equations (5) or (6).

The analytic representation of these data is thus of the form

$$\text{MCFA(MI)} = 1.00, \quad (7)$$

for the MI less than or equal to 0.40,

$$\text{MCFA(MI)} = 0.483\exp[1.817(\text{MI})] \quad (8)$$

for the MI greater than 0.40.

The passive neutron matrix corrections are determined by systematic drum matrix measurements in a manner similar to the active measurements discussed previously. The passive matrix correction factors, MCFP, are a function of only the MI.

The MCFP analytic fits to the four independent quantities measured during a passive assay scan are given below.

$$\text{MCFP(system totals)} = 1.00, \quad (9)$$

for the MI less than or equal to 0.355,

$$\text{MCFP(system totals)} = -0.16 + 3.28(\text{MI}), \quad (10)$$

for the MI greater than 0.355,

$$\text{MCFP(shielded totals)} = 1/[1 - \text{MI}], \quad (11)$$

$$\text{MCFP(system coincidence)} = [(0.5967)/(1 - \text{MI}) + 0.4187]^2, \quad (12)$$

$$\text{MCFP(shielded coincidence)} = [(0.8902)/(1 - \text{MI}) + 0.2337]^2. \quad (13)$$

The matrix correction equations given above are contained in the present PAN assay systems algorithms used throughout the DOE. The MI varies smoothly with average hydrogen density within a 208 L drum. Sludges display one of the highest average hydrogen densities, with correspondingly high MI's (0.4 to 0.8) of any CH-TRU waste form. Lightly moderating matrices, such as combustibles, have MI's falling typically in the 0.1 to 0.3 region, and miscellaneous metals matrices, which generally contain no moderating materials, have measured MI's near 0.0.

The MCF value is the multiplicative factor required to normalize a given matrix measurement to the empty drum level of PNCC sensitivity. The MCF value varies smoothly as a function of the MI; A graph of MCF versus MI can be used to estimate typical MCF values. For example,

- (a) Miscellaneous metals, MCF = 1.0 (i.e., same sensitivity as with empty drum),
- (b) Combustibles matrix, MCF = 1.35, and
- (c) Sludges, MCF = 3.6.

#### PAN Assay Algorithm and Data Acquisition System

The ORNL PAN unit is equipped with an IBM/PC-based data acquisition systems as described in reference 13. The system operating program (NEUT) controls all data acquisition and contains the assay algorithm.

To begin data acquisition, the operator enters drum identification, content code information, drum weight, and similar data from the PC keyboard, from a bar code reader, or from direct interaction with the site data management computer. This is followed by sequential active and passive neutron assays.

The weight of the drum's contents is used in calculating the specific activity (nCi/g) of TRU isotopes which differentiates between TRU and non-TRU wastes. The content code is used to flag difficult-to-assay matrices or "special case" drums.

Measured data, initialization information, date and time stamp from the internal PC clock, and final analysis results are archived on both floppy and internal hard magnetic disks. An on-line hard copy printout of the assay parameters and results is generated. All background and calibration measurements are routinely recorded and archived in the same fashion as normal assays. Thus, a continuous and traceable record of all data is maintained.

The PAN assay systems are comparatively recent developments in NDA instrumentation, having come into practical use in the last six years. Consequently, ASTM and ANSI standards have not yet been developed for these assay systems. Active assay techniques have been used for approximately 18 years, but 14-Mev thermalized neutron assay, which constitutes the active portion of PAN, is comparatively recent. The passive coincidence portion of PAN is similar to the PNCC assay technique. Therefore, ASTM, ANSI, and NRC standard practices and guidelines for PNCC are followed for that portion of the PAN system.

The assay procedure for PAN units equipped with the IBM/PC data acquisition system is relatively straightforward. An operator inserts a waste drum into the PAN unit and enters all drum identification information through an interactive dialogue with the PAN assay system software, NEUT. Once NEUT has checked the information for correct format, the assay record and programmable electronics hardware are properly indexed and prepared. NEUT then displays a message that the system is ready to begin assay.

At this point, the operator presses the start button on the MA165C neutron generator controller unit to initiate PAN active assay. At the conclusion of the active assay, NEUT automatically records all data and initiates the PAN passive assay. At the conclusion of the PAN passive assay, all data is recorded, analyzed and printed out for immediate inspection. The system is then ready to perform another assay.

#### PAN Assay Precision, Bias, and Limit of Detection

The PAN assay algorithm contains a calculation of measurement error [Reference 8] that combines statistical and estimated systematic errors based on the measured matrix correction factor. For a generally heterogeneous matrix and TRU materials distribution, the larger the indicated matrix correction, the larger the expected assay error. These values are reported with the actual assay values, for both passive and active neutron assays. For many waste streams a typical value for the estimated systematic error (not including the statistical contribution to the error) is 20 percent. When a systematic matrix correction formalism is used, the corresponding error in the passive assay measurement can be decreased to 5 percent or less for dry, combustible, low-hydrogen content waste, such as general laboratory waste.

The passive assay precision is calculated as for PNCC, with the addition of a term representing the uncertainty in the matrix correction factor. The algorithm used in the passive coincidence portion of the PAN calculates a composite assay error based on combining all the effects discussed above, which becomes part of the permanent archived assay record.

The active assay precision estimate includes a systematic error contribution, which is a function of the matrix correction factor (AI and MI). For reasonably homogeneous matrices, AI measurements indicate one sigma relative errors of  $\pm 10$  percent. For nonhomogeneous matrices, the variance is a function of the magnitude of the matrix correction factor [Reference 8]. That is, the larger the matrix correction factor the larger the associated variance of the assay. Distributional errors have been calculated and plotted as a function of the total matrix correction factor. For example, a matrix correction factor of approximately five yields a corresponding relative error (one sigma) of 50% in the assay measurement. Table 4 summarizes PAN assay biases.

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Table 4. PAN Assay Bias

<u>Matrix Correction Factor, MCF</u>	<u>Percentage Error</u>
1.0*	20
4.0	30
5.0	50

\* MCF = 1.0 corresponds to a "no matrix correction" case.

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Extensive comparisons have been performed for passive and active neutron assays of the same drum and a variety of matrix types, such as sludges, job-control wastes, combustibles, graphite scarfings, miscellaneous metals, tantalum crucibles, glassware, molten salts, filter media, dirt, and others. The matrix corrections applied to passive and active assays for a given type of matrix (except where no matrix corrections are necessary) are quite distinct. Thus, the probability of obtaining agreement between active and passive neutron assays for wastes with significant moderator and absorber amounts by accident or coincident matrix correction factors, is virtually negligible. That is, if one obtains agreement, both independent PAN assay techniques are considered to yield unbiased assay measurement values.

The assay limit of detection for the active neutron portion of the PAN unit is approximately 1 mg <sup>239</sup>Pu placed anywhere within a typical 208 L waste drum.

#### Decision-Tree Analysis

Two independent assay values, a "passive" mass and an "active" mass, are obtained with each PAN assay. A "decision-tree analysis" is performed within the assay algorithm to determine the most representative value. The default decision is to choose the active assay for low Pu mass (< 10g) and the passive assay for high ( $\geq 10$  g) high Pu mass.

When specific content code or matrix information is available that indicates, for example, that passive assay results are more reliable than active assay results for that content code, then the algorithm selects the passive assay results. Similar overrides involve the statistical accuracy of a measurement. For example, if the passive measurement has a large error associated with it, then the active measurement is selected.

## CONCLUSION

Waste characterization is the most important part of certifying that wastes meet a given set of waste acceptance criteria. A variety of methods and techniques may be used to characterize wastes, including material accountability, process knowledge, and direct or indirect measurements. In this paper, several techniques for direct measurement of radionuclide content, physical form, and chemical composition were discussed, including measurements which can be made at the generator level before waste is packaged into transportation containers, measurements that made be made on bulk waste containers after packaging, and verification of certain measurements by sample collection and analysis. Several waste characterization approaches are being employed at the Oak Ridge National Laboratory.

## REFERENCES

1. Beauchamp, John J. et al., "Statistical Sampling Plan for the TRU Waste Assay Facility," ORNL/CTD/203, September 1983.
2. F. J. Schultz, et al., "ORNL TRU Waste Drum Assay and Sampling Facilities Operational Procedures Manual," ORNL/CF-83/253, January 1984.
3. "TRU Waste Acceptance Criteria for the Waste Isolation Pilot Plant", WIPP-DOE-069, Revision 2, September 1985.
4. ANSI N15.20-1975, "American National Standard Guide to Calibrating Nondestructive Assay Systems."
5. USNRC Regulatory Guide 5.22, "Nondestructive Assay of Special Nuclear Material Contained in Scrap and Waste," Revision 1, April 1984.
6. ASTM C 859-87, "Standard Terminology Relating to Nuclear Materials".
7. Standard Test Method for Nondestructive Assay of Special Nuclear Material in Low Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning, ASTM C 853. This draft standard has been referenced with permission from ASTM Subcommittee C-26.10.
8. J.T. Caldwell, R.D. Hastings, G.C. Herrera, W.E. Kunz, E.R. Shunk, "The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum-Size Containers", Los Alamos Formal Report LA-10774-MS, September 1986.
9. J. T. Caldwell et.al, "System Evaluation Including Assay Algorithm, Matrix Corrections, and Operational Performance of the Los Alamos Passive/Active Neutron Assay Systems," Los Alamos Technical Report N2-87-222WP, (in preparation as a formal LA-MS series report. Estimated publishing date June 1988).
10. R. B. Perry, R. W. Brandenburg, and N. S. Beyer, "The Effect of Induced Fission on Plutonium Assay with a Neutron Coincidence Well Counter," Transactions of the American Nuclear Society, 15 674 (1972).
11. ASTM C 853-82, "Standard Test Methods for Nondestructive Assay of Special Nuclear Materials Contained in Scrap and Waste."
12. Fleissner, John G. and Hume, Merril W., "Comparison of Destructive and Nondestructive Assay of Heterogeneous Salt Residues," RFP-3876, March 29, 1986.
13. T. H. Kuckertz et.al., "Making Transuranics Assay Measurements Using Modern Controllers," Proceedings 9th ESARDA Symposium on Safeguards and Nuclear Material Management, London UK, pages 389-393, May 1987.
14. C. E. Moss and J. T. Caldwell, "Assay of TRU Wastes Containing (alpha, n) Sources," LA UR 86-2220, June 22, 1986.

REFERENCES (contd.)

15. "Chart of the Nuclides," Knolls Atomic Power Laboratory.
16. F. J. Schultz, et al., Oak Ridge National Laboratory; J. T. Caldwell et al., Los Alamos National Laboratory, "First-Year Evaluation of a Nondestructive Assay System for the Examination of ORNL TRU Waste," ORNL-6007, April 1984.
17. F. J. Schultz, et al., "Neutron and Gamma-Ray Nondestructive Examination of Contact-Handled Transuranic Waste at the ORNL TRU Waste Drum Assay Facility," ORNL-6103, March 1985.