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TRU WASTE ASSAY INSTRUMENTATION  
AND  
APPLICATIONS IN NUCLEAR FACILITY DECOMMISSIONING

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Introduction

A vital part of any comprehensive radioactive waste management program is the ability to determine the types and amounts of radioactivity in wastes. Without such ability, it is impossible to direct cost effective and timely waste management programs for waste generators and repositories, the decontamination and decommissioning of outdated nuclear facilities, and the exhumation of old radioactive waste burial grounds. Heretofore, only administrative controls could be used for TRU waste sorting, often resulting in overestimates of TRU waste quantity, thereby significantly increasing the amount of materials that had to be handled as retrievable wastes, which requires expensive handling techniques and engineered storage.

To ameliorate this problem, a comprehensive program is in progress at the Los Alamos National Laboratory for the development of sensitive, practical, nondestructive assay techniques for the quantification of low-level transuranics (TRU) in bulk solid wastes. The program encompasses a broad range of techniques, including sophisticated active and passive gamma-ray spectroscopy, passive neutron detection systems, pulsed portable neutron generator interrogation systems, and electron accelerator-based techniques. The techniques can be used with either low-level or high-level beta-gamma wastes in either low-density or high-density matrices. Waste package containers range in size from 2 ft<sup>3</sup> cardboard cartons to 4x4x7' wood and metal crates. Through the use of such techniques, the capability now exists to cost effectively sort and segregate wastes by TRU content.

### Combustible Waste Package Counter

One of the first instruments designed and built for the assay of transuranic (TRU) wastes at the 10 nCi/g level was the Multi-Energy Gamma Assay System (MEGAS).<sup>1</sup> The original MEGAS has been significantly upgraded (MEGAS II),<sup>2-4</sup> as shown in Fig. 1. MEGAS II operates in a segmented mode, which allows the determination of hot spots within waste packages. The photon detector is a 127-mm diameter by 1.6-mm thick NaI crystal, which optimizes the TRU detection capability using L x rays and gamma rays having an energy less than 100 keV. The detection limit at the 3 $\sigma$  level above background for <sup>241</sup>Am is less than 5 pCi/g and for <sup>239</sup>Pu is less than 1 nCi/g for a 500-s count. Packages contain low density, combustible type wastes in a 2 ft<sup>3</sup> cardboard carton. Total mass of the container and wastes is generally less than 10 kg.

The presence of beta- and gamma-ray emitting fission products decreases the TRU detection limit for the NaI detector. The addition<sup>3,4</sup> of a high resolution, hyperpure, planar germanium detector, 1000-mm<sup>2</sup> active area, 12-mm thick, allows the assay of TRU isotopes even in the presence of several mR/h gamma and beta backgrounds. A tabulation of measured detection limits for the hyperpure germanium detector is presented in Ref. 3. Using these data, it is estimated that TRU assay at the 10 nCi/g level can still be made even in the presence of 400  $\mu$ Ci of <sup>137</sup>Cs (65 nCi <sup>137</sup>Cs/g).

As shown in Fig. 1, four banks of polyethylene moderated <sup>3</sup>He neutron detectors have been added. The measured detectability limit (3 $\sigma$  level above background, 1000-s count, total neutron count) for these neutron detectors is 400 nCi/g for weapons grade plutonium oxide. Because <sup>3</sup>He detectors are relatively insensitive to photons, they can operate even in the presence of high fission product backgrounds (1-10 R/h). The neutron counters were added to assure that significant quantities (~100 mg) of plutonium, even if well shielded, would be detected. The technology for this development has been transferred to other DOE facilities and to the commercial instrumentation sector.

### Large Package and Crate Counter

Much of the plutonium and uranium waste generated in the nuclear industry is ultimately packaged in 208-l barrels or large crates having

typical dimensions of 1.0 m or more on a side. An active-passive  $4\pi$  neutron counting system has been developed to assay/screen these large containers for their TRU and uranium content. This crate counter is made from discrete moderated  $^3\text{He}$  neutron detector modules which are easily arranged into a variety of assay chamber geometries. Very large objects and debris from decommissioning programs can be easily accommodated in the counter.

Figure 2 depicts the construction of the discrete counter modules and the placement to form an assay chamber with internal dimensions of 1.2x1.2x2.4 m. The measured  $4\pi$  detection efficiency in this configuration for a bare californium source is 14%. Separate counting electronics are provided for each of the two chambers in each of the six modules for a total of twelve independent signals. The relative singles count rates from different portions of the  $4\pi$  system are used for geometry and matrix corrections. Figure 3 shows the ratio of the count rates from the two end modules as a function of source position along the length of the assay chamber. Similar ratios have been measured for the two sets of opposite side modules.<sup>5</sup> The three ratios can serve to determine the approximate location of a source.

All neutron detection systems suffer in the presence of matrix materials, particularly hydrogenous materials. A flat response (10%) was measured for  $^{252}\text{Cf}$  neutron sources moderated by thicknesses of polyethylene ranging from 0 to 7.5 cm. A compensation technique for greater effective hydrogenous moderators is based on the differential energy sensitivity of the count rates in the inner and outer chambers of each module.

For plutonium contaminated wastes, the passive  $4\pi$  coincident measurement generally determines the  $^{240}\text{Pu}$  mass. If the  $^{240}\text{Pu}$  to total Pu ratio is known, this measurement determines the total Pu mass. Measured  $3\sigma$  detection sensitivity is about 10 mg  $^{240}\text{Pu}$ .<sup>5,6</sup>

The active part of the crate counter is the differential die-away pulsed neutron technique discussed next and elsewhere.<sup>7</sup> Preliminary detection limits for the active crate counter are 5-10 mg for either  $^{239}\text{Pu}$  or  $^{235}\text{U}$ .<sup>6</sup> Such crate assay systems are now under design and construction for implementation at other DOE facilities.

### Drum Assay System

The Los Alamos National Laboratory has also developed an accurate, high sensitivity assay instrument for the assay of TRU waste in 208-l barrels. The assay chamber of this differential die-away system consists of a graphite and polyethylene structure with a small, pulsed D-T neutron generator inside. Both cadmium covered and bare  $^3\text{He}$  neutron detectors are incorporated in the chamber, being placed external to the graphite but within polyethylene. The graphite and polyethylene moderate 14 MeV neutrons, which are completely thermalized in 0.7 ms. The thermal neutrons die away in the interrogation cavity with a half life of about 0.76 ms. The interrogating thermal pulse lasts a long time in the chamber and induces thermal neutron fission in any fissile material present in the waste barrel. This system has a measured  $^{239}\text{Pu}$  sensitivity of less than 1 mg in a 208-l barrel. A complete description of this system, including its application to mixed wastes (curium, californium, plutonium, uranium, americium, neptunium) and matrix correction methods, is presented elsewhere.<sup>8,9</sup> A drum assay system has recently been developed and fabricated by Los Alamos and installed at the Oak Ridge National Laboratory for test and evaluation purposes under field conditions.

### Gamma Assay

An ideal supplement to the pulsed thermal neutron interrogation system is gamma-ray spectroscopy. This is particularly true for waste containing many isotopes. Gamma-ray spectroscopy is sensitive to most radioactive isotopes, including fission products, not only TRU isotopes that either have a significant spontaneous fission or neutron-induced fission cross section. Some specific examples of isotopes that cannot be assayed via neutrons that can be assayed using gamma-ray spectroscopy are  $^{243}\text{Am}$  and  $^{237}\text{Np}$ , which have daughters,  $^{239}\text{Np}$  and  $^{233}\text{Pa}$ , respectively, that have energetic gamma rays. An isotope in the grey area is  $^{241}\text{Am}$ , which has a low fission cross section and spontaneous fission rate and a very intense, but low energy ( $E_\gamma = 60 \text{ keV}$ ), gamma ray.

Active/passive gamma-ray spectroscopy can quantify the radioactive wastes in a barrel.<sup>10,11</sup> The major problem is characterizing the matrix to make the necessary corrections to the gamma-ray signatures. There are

two subtly different techniques. One is to use external sources, identical to the isotopes in the barrel, to over-ride the passive signal, to give the effective attenuation at the desired energies. The other technique relies on the fact that above about 150 keV, the attenuation coefficient varies smoothly and slowly as a function of energy. This technique characterizes the matrix as a function of energy over a large energy range. The upper part of Fig. 4 shows the linear attenuation coefficient  $\mu$  as a function of energy for an actual waste barrel ( $\rho = 0.22 \text{ g/cm}^3$ ). The line in the figure is a linear least squares fit to the data. There is no a priori reason to exclude a linear relationship between  $\mu$  and  $\ln(E_\gamma)$ . The barrel supposedly has paper and laboratory glassware in it. For comparison, the lower part of Fig. 4, using the same scales, shows the linear attenuation coefficient as a function of energy for pyrex glass ( $\rho = 0.25 \text{ g/cm}^3$ ). There is very good agreement, both in magnitude and in slope, between the two plots. There is a small curvature to the data at low energies, but the data are approximately linear above about 150 keV.

#### LINAC Applications

An electron linear accelerator (LINAC) can be the heart of a complete assay system. Photofission interrogation offers good sensitivity for TRU, but because of the similarity of photofission cross sections for both fissile and fertile (e.g.  $^{238}\text{U}$ ) isotopes and other high Z materials, such as lead, identification of specific nuclides is difficult. Thermal neutron interrogation offers high sensitivity for fissile isotopes but essentially none for fertile isotopes. A combination of neutron and photon interrogation can separate the fissile and fertile isotopes.<sup>12</sup>

Photons are produced in a bremsstrahlung target that stops the electron beam. The photon beam then passes through a polyethylene slab to harden the photon spectrum. A portion of the higher energy photons above various reaction threshold energies will produce photoneutrons. A beryllium converter can also be used to significantly increase the photoneutron flux. Photoneutrons and prompt photofission neutrons will thermalize in a few tens of microseconds and will persist as thermals for hundreds of microseconds, during which time they will generate thermal neutron fissions among the fissile TRU. Prompt fissions from thermal fission are separated in time from the photoneutrons and can serve as a quantitative signature. The detection method is the differential die-away system described earlier and elsewhere.<sup>7-9</sup>

While the thermal fissions are produced in near simultaneity with the photofission events, the prompt and delayed neutrons from the two fission processes can be resolved in a single detector. This is illustrated in Fig. 5, where the neutron count rate from a 1 g  $^{239}\text{Pu}$  sample irradiated by a 12 MeV bremsstrahlung burst is plotted versus time after burst. Curve "a" shows the prompt neutron count rate to persist for about 8 ms, with a nearly constant delayed neutron count rate continuing to the next burst. Curve "b", obtained with the  $^{239}\text{Pu}$  wrapped with cadmium, shows the delayed neutrons to be only weakly affected by the cadmium, whereas the prompt neutrons are essentially absent, demonstrating the predominantly photofission origin of the delayed group. For a 200-s LINAC interrogation run, the 3 $\sigma$  detection limit is better than 1 mg  $^{239}\text{Pu}$ , which is less than 1 nCi/g of waste for a 105 kg matrix of aluminum scrap in a 208-l barrel.<sup>13,14</sup> Barrels of concrete, bitumen, sand, and other matrices have also been studied with applications in decommissioning and exhumation programs.

While the LINAC is being used as an interrogation source, it can simultaneously be used to produce a radiograph or picture of the waste container and contents. A radiograph indicates where and what inhomogeneities are in the barrel. Of a purely qualitative nature, a radiograph gives an excellent fingerprint of the barrel, which can be used for shipper/receiver verification that a given barrel has not been tampered with.

Figure 6 shows a one dimensional radiograph of a composite wedge of wood, aluminum, polyethylene, and copper. This transmission scan was taken at an electron beam energy of 6 MeV, 150 pps, about 275 ma beam current, and a 4  $\mu\text{s}$  wide beam pulse. The scanning table was moving past the bremsstrahlung target at about 3 cm/s. The data represent 30 sweeps in a multichannel scale mode, 10 ms/channel dwell time. The detector was a plastic scintillator (NL 102) coupled to a photodiode. Spatial resolution in these preliminary measurements is about 1 cm. This can be optimized by changing the speed of the scanning table, the dwell time per channel of the multichannel scale, and the repetition rate of the LINAC.

To further complete an assay, the LINAC can be used to identify matrix constituents using the thermal neutron capture reaction ( $n, \gamma$ ) and a hyper-pure germanium (HpGe) gamma detector. Preliminary measurements have identified cadmium and aluminum. The HpGe detector was severely affected

by the gamma flash from the LINAC and was paralyzed for several milliseconds after the flash. Thus, the prompt capture gamma rays were missed and only a few gamma rays from thermal neutron activation were detected. Efforts are under way to reduce the paralysis time of the detector.

Similar matrix studies can be made using other external neutron sources, such as  $^{252}\text{Cf}$ , or even the internal neutron sources contained within the waste. Obviously, these methods do not suffer from detector paralysis problems. Table 1 shows the experimentally measured detectability limit (3 $\sigma$  level above background, 1000-s count) for various elements located in the central region of a 208-l barrel ( $\sim$ 100 kg). These measurements employed a 50- $\mu\text{g}$   $^{252}\text{Cf}$  neutron source and a highly collimated 16% efficient Ge(Li) detector. Thermal neutron capture is particularly sensitive to neutron poisons. A detectability limit below 1 g ( $\sim$ 10 ppm) is achieved for all the neutron poisons except lithium. The poor sensitivity for lithium is because the primary neutron absorbing lithium isotope,  $^6\text{Li}$ , captures neutrons without emitting gamma rays. The technique can be used to identify hazardous and toxic materials other than radioactive materials (e.g. the heavy metals, mercury and cadmium).

#### Summary

The Los Alamos TRU waste assay program is developing measurement techniques for TRU and other radioactive waste materials generated by the nuclear industry, including decommissioning programs. Systems are now being fielded for test and evaluation purposes at DOE TRU waste generators. The transfer of this technology to other facilities and the commercial instrumentation sector is well in progress.

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TABLE 1. Elemental Thermal Neutron Capture Gamma-Ray Sensitivities for 208-Liter-Drum Assays.

Element	Number of Gamma-Ray Lines <sup>a</sup>	Detectability Limit <sup>b</sup>
Hydrogen <sup>c</sup>	1	14.2 g
Helium	0	
Lithium <sup>d</sup>	7	3.2 kg
Beryllium	7	8.6 kg
Boron <sup>d</sup>	7	150 mg
Carbon	3	40.3 kg
Nitrogen <sup>c</sup>	43	1.7 kg
Oxygen	0	
Fluorine	11	5.6 kg
Sodium	51	176 g
Magnesium	18	286 g
Aluminum <sup>c</sup>	51	605 g
Silicon	27	970 g
Phosphorus	60	2 kg
Sulfur	33	400 g
Chlorine	41	15.3 g
Potassium	88	280 g
Calcium	46	792 g
Scandium	87	21.8 g
Titanium	39	45.2 g
Vanadium	62	68.2 g
Chromium	56	202 g
Manganese	76	48.2 g
Iron <sup>c</sup>	42	508 g
Cobalt	59	29 g
Nickel	49	122 g
Copper	66	95 g
Zinc	71	1.2 kg
Cadmium <sup>d</sup>	38	420 mg
Gadolinium <sup>d</sup>	17	879 mg
Mercury	41	3.2 g

<sup>a</sup>When usable, escape peaks are included.

<sup>b</sup>Counting time 1000-s, three standard deviations above background.

<sup>c</sup>Possible interference with measurement system components.

<sup>d</sup>Neutron poison.

FIGURE 1. MEGAS II showing NaI detector, hyperpure germanium detector,  $^3\text{He}$  neutron detector banks, and data acquisition and analysis equipment.



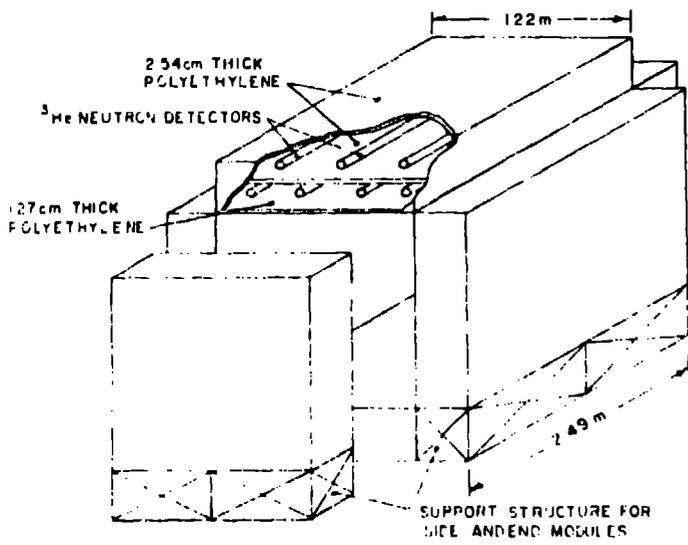


FIGURE 2. Modular  $4\pi$  neutron assay chamber.

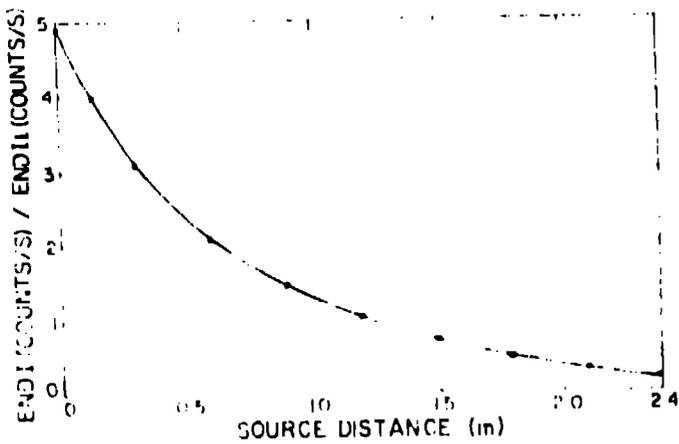


FIGURE 3. Ratio of count rates for the end modules of a  $4\pi$  neutron assay chamber for a source moved along the length of the assay chamber.

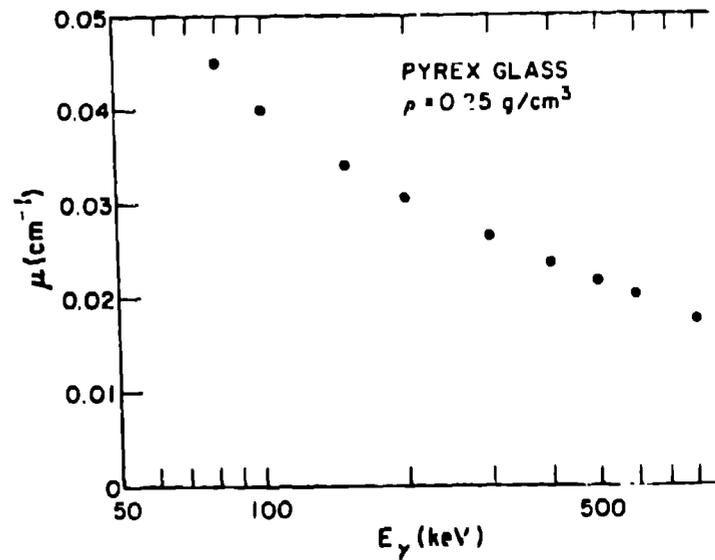
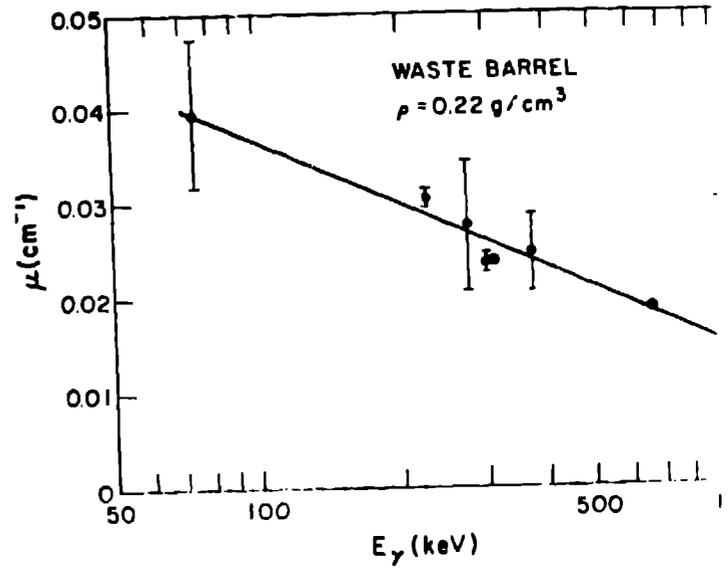


FIGURE 4. Linear attenuation coefficient vs.  $E_\gamma$  for an actual waste barrel having  $\rho = 0.22 \text{ g/cm}^3$  (upper). Theoretical linear attenuation coefficient vs.  $E_\gamma$  for pyrex glass having  $\rho = 0.25 \text{ g/cm}^3$  (lower).

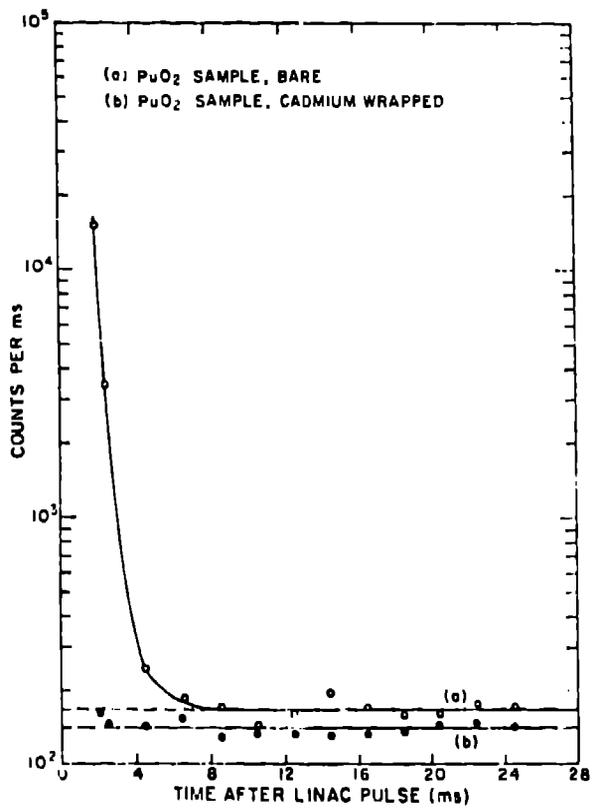


FIGURE 5. Neutron count rate vs. time from simultaneous photon and neutron interrogation of 1 g  $^{239}\text{Pu}$ .

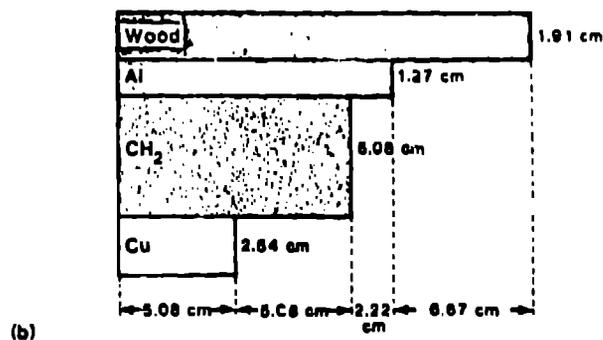
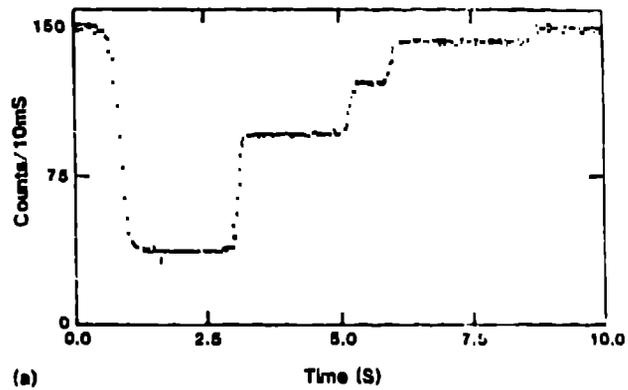


FIGURE 6. Transmission scan (a) of the wood-Al-CH<sub>2</sub>-Cu wedge shown in (b).