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FIRST-YEAR EVALUATION OF A NONDESTRUCTIVE ASSAY SYSTEM FOR THE EXAMINATION OF ORNL TRU WASTE

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ABSTRACT

The Oak Ridge National Laboratory has been selected as the demonstration site for a new transuranic neutron assay system (NAS) developed at the Los Alamos National Laboratory. In addition, in order to meet specific ORNL program objectives, an upgraded segmented gamma-ray drum scanner has been integrated into the nondestructive assay (NDA) system to serve as a radioisotope identifier and as a quantitative assay backup to the NAS. A verification study, wherein selected waste drums will be emptied into glove boxes and their contents sampled and subsequently gamma-ray assayed, will take place in FY 1984. Results will be compared to those obtained from the NDA techniques. The NAS uses pulsed-neutron interrogation (differential-dieaway technique) and passive neutron measurements to determine the fissile component and an upper-limit estimate of the total TRU activity contained in each waste drum. Of the 171 waste drums assayed to date, nine drums were determined to contain less than 10 nCi/g TRU isotopes. An additional number of drums (approximately 20%) are expected to be categorized as non-TRU, which is presently defined as less than 100 nCi/g TRU concentration. This requires a detailed analysis of the data which includes waste matrix compensation, systematic qualitative and quantitative gamma-ray analyses, and interpretation of neutron multiplicity data. Reproducibility of the active assay measurements on a single waste drum indicate agreement to $\pm 3\%$ relative error.

System components have performed very well. One ^3He proportional counter tube and one detector preamplifier have been replaced since operations began in April 1982. After redesign and replacement of the internal voltage supply of the LeCroy 3500 multichannel data acquisition system, persistent computer downtime was significantly reduced. Total system uptime has been approximately 95%. Overall system performance has been excellent.

INTRODUCTION

Oak Ridge National Laboratory (ORNL) transuranic (TRU) waste is packaged in 208-L (55-gal) stainless steel drums and retrievably stored in underground weather-resistant structures under controlled surveillance (see Fig. 1). Due to the limited space available for storage, it is advantageous and cost effective to reduce the volume of TRU waste retrievably stored. Waste containing less than 100 nCi/g transuranics (alpha-emitting radionuclides of atomic number greater than 92 and half-lives greater than 20 y)¹ may be disposed of as low-level waste, while waste containing greater than 100 nCi/g transuranics must be retrievably stored.

Classification of the ORNL waste into "TRU" and "non-TRU" is required for proper disposition. The task of classifying the waste is further complicated by the score of TRU isotopes contained in the waste. Four independent on-site TRU-waste-generating streams have contributed to the large array of TRU isotopes detected in the waste. The isotopes present, both TRU and non-TRU, in a significant fraction of waste drums (greater than 20% of the drum population) include $^{237}\text{Np}/^{233}\text{Pa}$, ^{239}Pu , $^{241}\text{Pu}/^{237}\text{U}$, ^{242}Pu , ^{241}Am , $^{243}\text{Am}/^{239}\text{Np}$, ^{242}Cm , ^{244}Cm , ^{249}Cf , ^{60}Co , $^{106}\text{Ru}/^{106}\text{Rh}$, ^{125}Sb , ^{134}Cs , ^{137}Cs , ^{144}Ce , and ^{154}Eu . Some 45 additional radionuclides in lesser quantities have been identified.

ORNL-PHOTO 4862-80

Fig. 1. ORNL TRU Waste Retrievable Storage Area

ORNL has been selected as the demonstration site for a new transuranic neutron assay system developed at the Los Alamos National Laboratory. The major objectives of the cooperative interlaboratory program are to field test, calibrate, and evaluate the neutron assay system, to provide a demonstration and training facility for personnel from other DOE sites and contractors, to reduce the volume of TRU waste stored at ORNL, and to provide positive identification of the radionuclide content of the ORNL TRU waste.

In order to meet these objectives, a two-tier, passive and active, nondestructive assay (NDA) system utilizing the Los Alamos National Laboratory neutron interrogation assay system² and an upgraded gamma-ray drum scanner has been employed in the TRU Waste Drum Assay Facility (TWDAF) (see Fig. 2).

Verification of the NDA techniques will be attempted through the physical sampling of selected TRU waste drums in the TRU Waste Drum Sampling Facility (TWDSF). The destructive assaying involves unsealing and removing the lid of a waste drum and then transferring the contents into a glove box. The contents will be segregated into the following categories: cellulose, plastics, metals, and glass and ceramics. A predetermined number of samples will be obtained from each waste category and the samples transferred out of the glove box for assay by gamma-ray spectroscopy. The assay results thus obtained will be compared to the nondestructive assay results.

This report provides a detailed description of each NDA system operated in the TWDAF and a description of the TWDSF. Neutron and gamma-ray assay results are presented and discussed. A summary of ORNL's operating experience and system performance for the neutron assay system is also included.

ORNL-PHOTO 3029-82

Fig. 2. ORNL TRU Waste Drum Assay Facility (TWDAF)

TRU WASTE DRUM ASSAY FACILITY

The ORNL TRU Waste Drum Assay Facility (see Figs. 2 and 3) is located in the ORNL Solid Waste and Storage Area No. 5 (SWSA 5) adjacent to the TRU Waste Retrievable Storage site (see Fig. 4). The interior building modifications in preparation for assaying TRU waste drums were completed in March 1982. Some of the modifications included installation of building heating and air conditioning, 120-V and 208-V electrical circuits, one-foot-thick concrete shielding walls, additional office space, and an electric forklift recharging unit. The floor plan of the facility is given in Fig. 5.

Approximately once a week 32-40 TRU waste drums are transferred from the staging area, Building 7823 (Fig. 4), to the TWDAF, Building 7824. The drums are nondestructively assayed to determine their TRU isotopic identity and content. The NDA examination includes a passive neutron and a pulsed neutron interrogation using the NAS and a gamma-ray examination using the gamma-ray drum scanner (GRDS).

The sequence of events in the TWDAF for a typical waste drum is outlined below and is shown pictorially in Fig. 6.

1. The drum is placed on a platform scale to determine its mass in kilograms (see Fig. 7).
2. The drum is scanned by a suitably calibrated, portable, cadmium-wrapped neutron slab detector to determine the gross neutron emission rate (n/s) (see Fig. 7).
3. Depending on the results obtained from Step 2, the drum is designated either a "cold" ($<5 \times 10^4$ n/s) or a "hot" drum ($>5 \times 10^4$ n/s) and suitably labeled.
4. The drums are actively and passively scanned by the NAS. The "hot" drums are run through the NAS first.

Fig. 3. TWDAF Neutron Assay System (NAS) and Gamma-Ray Drum Scanner (GRDS)

∞

Fig. 4. ORNL Solid Waste Storage Area No. 5 (SWSA 5)

▨ SOLID CONCRETE BLOCK SHIELDING WALL

---- CHAIN LINK FENCING

DIMENSIONS IN FEET

Fig. 5. ORNL TRU Waste Drum Assay Facility - Building 7824, Floor Plan

TRU WASTE DRUM ASSAY FACILITY— DRUM FLOW DIAGRAM

Fig. 6. TWDAF Drum Flow Diagram

Fig. 7. Neutron Assay Screening Station

5. The drum is then passively gamma-ray scanned using the segmented GRDS. The "hot" drums are then removed from the building to reduce the neutron and gamma-ray background experienced during examination of the "cold" drums. The gamma-ray system possesses the capability of quantitative scanning, but this is performed only infrequently as a backup to the NAS.
6. After the drum is examined by the neutron and gamma-ray assay systems, it is appropriately labeled as "TRU" or "NON-TRU" and stored (see Fig. 8) for transfer to the staging area.

The operational procedures used in the TWDAF for the sequence of events outlined above are given in Reference 3.

NEUTRON ASSAY SYSTEM

The TRU waste neutron assay system (see Figs. 9-11) uses pulsed active neutron interrogation with detection of prompt-fission neutrons (differential-decay technique) to assay the drum's fissile component and passive neutron measurements to estimate all other TRU isotope contributions. The passive mode of the NAS possesses the capability of measuring the total passive neutrons, passive coincidence neutrons, and passive neutron multiplicities resulting from the spontaneous fission neutron and (α ,n) components contained in the waste.

The assay chamber is a rectangular parallelepiped with a square cross-section of 67 cm on a side and 102 cm high (see Fig. 12). A layer of graphite (10.8 cm thick) encloses the assay chamber. The graphite walls (including chamber door) are surrounded by 30.6 cm of polyethylene, while the roof of the chamber is enclosed by 35.7 cm of polyethylene. The graphite and polyethylene serve to moderate the fast neutrons produced by the neutron generator positioned inside the assay chamber (see Fig. 9) and also serve as personnel shielding.

Fig. 8. TWDAF Drum Storage

Fig. 9. Neutron Assay System

Fig. 10. Neutron Assay System

Fig. 11. NAS Detector Electronics

Fig. 12. Neutron Assay System

The interrogating neutrons are provided by a small pulsed D+T (Deuterium-Tritium) neutron generator (see Appendix A) located in one corner of the chamber at approximately mid-drum height (see Fig. 13). The neutron generator produces approximately 10^6 14-MeV neutrons per pulse with a pulse repetition rate variable from 1 to 100 Hz. The pulse duration is 10–15 μ s. The average expected lifetime of the D+T tube is approximately 5×10^7 pulses, or approximately 10,000 assays at 2000 pulses per assay.

The pulses of 14-MeV neutrons are moderated in the graphite and polyethylene. The resultant thermal neutrons induce fissions in the fissile material contained in the waste. Cadmium-shielded packages of 3–91 cm $\times$ 5.08 cm, 2-atm ^3He proportional counters are located at the center of each vertical wall. Similar packages of 61 cm $\times$ 5.08 cm, 2-atm ^3He proportional counters are located above the top graphite layer and below the bottom graphite layer.⁴ These cadmium-shielded packages constitute the active prompt-neutron detection system.

The cadmium-shielded neutron detectors recover from the initial 14-MeV pulse overload in approximately 0.5 ms and are insensitive to the interrogating thermal neutrons. The ensuing prompt-fission neutrons induced by the thermal-neutron flux are detected with 3.5% efficiency. To monitor the interrogating thermal-neutron flux, a low-efficiency, bare (unshielded) ^3He proportional counter is placed within the assay chamber (see Fig. 13). The ratio of the prompt-fission neutron detector counts to the interrogating flux monitor counts is proportional to the amount of fissile material present in the waste drum. One must assume that the interrogating thermal flux is essentially the same inside and outside the drum.

Fig. 13. NAS Assay Chamber

Of course, neutron moderating and absorbing materials contained in the waste package affect the neutron sensitivities of the detectors.

Table 1 demonstrates the variation in fissile mass detection sensitivity with varying waste matrices.

Table 1. Variability of NAS sensitivity to plutonium-239

Drum matrix	Weight (kg)	Plutonium-239 sensitivity (nCt ^{239}Pu /g waste)
Empty drum	32	0.58
Wet rags	200	0.19
Sand	200	0.19
Iron	220	0.34
Concrete	500	0.62

Figure 14 shows a multiscale time history⁴ of a 2000-pulse neutron assay measurement of an ORNL waste drum (208-L) containing ^{235}U . The plotted points are the sum of all the background-corrected cadmium-shielded detector counts vs time. The region between 0.3 and 5 ms is dominated by the prompt fission neutron response. Two components are evident after a two-exponential fit to these data: 1) the earlier component with a half-life of $\sim 200 \mu\text{s}$ corresponds to the fission events induced by neutrons thermalized directly in the matrix and 2) the longer half-life component ($t_{1/2} = 420 \mu\text{s}$) corresponds to interrogation by thermal neutrons originating in the assay chamber walls. This two-component response is characteristic of waste matrices containing significant amounts of hydrogen.

To provide for passive neutron, coincidence, and neutron multiplicity counting, additional bare ^3He proportional counters were embedded in

Fig. 14. Neutron Multi-Channel Scale Time-History
for Waste Drum Containing Gram Quantity ^{235}U

the polyethylene layers adjacent to the graphite. Each such bare proportional counter provides roughly 2.3 times the response of a corresponding proportional counter within the cadmium-shielded package. There are four bare proportional counters (two sets of two) placed along each vertical wall and in the chamber's ceiling. No bare proportional counters were placed at the chamber's bottom, where two shielded packages are in place. Separate counting electronics, including a preamplifier, amplifier, and discriminator, are provided for each of the four sides and the top set of bare proportional counters. A second, bare, low-pressure ^3He proportional counter is positioned outside the assay chamber opposite the neutron generator. It monitors the pulses of 14-MeV neutrons produced by the generator.

A LeCroy 3500 system is used for data acquisition and analysis (see Fig. 15). The time histories from the main detector packages and the flux monitor are recorded on magnetic diskettes for archival storage.

The pulsed-neutron interrogation determines the fissile component of the drum's TRU isotope content. The coincidence passive neutron measurement determines the spontaneous fission component; and the passive total neutron measurement, after accounting for the spontaneous fission contribution, can be used to estimate all other TRU isotope contributions. The neutron multiplicity measurements serve to identify which spontaneous fission isotopes are present in significant quantities. This information is usually sufficient to categorize a waste drum.

GAMMA-RAY DRUM SCANNER

The Gamma-Ray Drum Scanner used in the assay system is an upgraded Canberra Segmented Drum Scanner Model 2220B (see Figs. 16 and 17).

Fig. 15. TWDAF Control Room

Fig. 16. Gamma-Ray Drum Scanner

Fig. 17. Gamma-Ray Drum Scanner

It consists of the following components: a collimated solid state Ge(Li) detector, computer-based multichannel analyzer, and a dual-axis positioning system.

The scanner was originally designed to assay ^{235}U and/or ^{239}Pu in drums filled with low-density wastes such as rags, towels, and rubber gloves. The scanner will measure five or more grams of ^{235}U or ^{239}Pu to an accuracy of better than $\pm 20\%$ for densities up to 200 kg/m^3 .

The GRDS uses a segmented scanning technique wherein an 8000-channel gamma-ray energy spectrum of each individual horizontal segment (presently, seven segments per 208-L drum) along the vertical axis of the drum is collected and stored on magnetic diskettes by the LeCroy 3500 multichannel analyzer. The segmented horizontal scanning reduces the effect of vertical inhomogeneity. Also, the drum is rotated to reduce the effects of axial inhomogeneity. The operator has the option of viewing one, none, or all of the spectra collected. The seven segments are then summed to render a qualitative spectrum emitted by the contents of the drum.

For quantitative assaying, each measurement must be corrected for the varying mass absorption coefficient encountered in the waste drum. The attenuation correction is made by measuring a source of known intensity (transmission source), emitting gamma-rays with energies similar to the assay gamma-rays, through the waste drum and measuring the decrease in intensity. From the fundamental law of gamma-ray attenuation,⁴ the sample transmission T is related to the sample linear attenuation coefficient, μ , by the relation $T = e^{-\mu x}$, where x is the thickness of the sample penetrated. Three assumptions are made:

1. The material to be assayed and the matrix material are reasonably homogeneous.
2. The particles of assay material are small enough to ignore self-attenuation within individual emitting particles.
3. Multiple scattering and neutron buildup factors are ignored.

The sample transmission, T , is defined as the fraction of gamma-rays from the external (transmission) source that pass through the sample unabsorbed and unscattered. It is assumed T is measured along a diameter of the container and that the detector views the sample along the normal to the midpoint of the axis.

The present assay scheme provides for all waste drums to be qualitatively scanned for gamma-ray-emitting radioisotopic identification. However, only those drums which cannot be positively identified through the neutron measurements as possessing less than 100 nCi/g TRU concentration will be quantitatively scanned.

TRU WASTE DRUM SAMPLING FACILITY

A validation study has been undertaken to determine the ability of the NDA technique to accurately predict the mean concentration of transuranic isotopes contained in each waste drum. The population to be sampled consists of approximately 2000 drums containing materials [decontamination debris (DD), contaminated equipment (CE), dry solids (DS), and other] which have been used in the handling of radioactive substances. The operation is expected to operate for approximately two years with an average of one drum per week of waste being examined.

A statistical sampling plan (see Ref. 6) has been suggested as a means of validating the NDA measurements. Every drum will be measured by the NDA system, and a subpopulation of drums will also be examined by hand inspection in the TRU Waste Drum Sampling Facility (TWDSF). For those drums which are examined by both methods, the measurements will be compared, and the proportion of agreement between the matched observations will be determined and will form the basis for the validation.

Approximately 150 drums will be selected for hand inspection according to the drum sampling plan. To become eligible for destructive assaying in the TWDSF, the waste drum will have met the following criteria:

1. must have been both actively and passively assayed in the Neutron Assay System (NAS);
2. must have been passively scanned by the segmented GRDS;
3. must contain less than 10 g fissile mass;
4. must possess a total dose equivalent rate (beta-gamma and neutron) less than 200 mrem/h;
5. must possess a mass less than 114 kg;
6. must have a neutron emission rate less than 5×10^4 n/s;
7. must comply with the glove box operating standards for transuranium elements given in Ref. 7.

The sampling plan also provides a method to determine the number of samples to be collected from each waste category within a selected drum. Estimates of total number of samples collected range from 50 to 100 for each drum.

The TWDSF is located in the basement of the ORNL Thorium-Uranium Recycle Facility (TURF). It consists of four stainless steel double-faced glove boxes that have been assembled into two double-length

boxes (see Figs. 18 and 19). The boxes are connected to the building's hot off-gas system through high-energy particulate absorber (HEPA) filters and pressure-reducing Maxitrol valves. The first box has affixed to the side panel a drum bag-in port through which waste from the drum will be brought into the box. The box contains a small shredder (see Figs. 20 and 21) to shred and/or crush paper, plastics, and glass and ceramics and a balance to determine the mass of the waste.

The waste material will be segregated into four categories: cellulose, plastics, metals, and glass and ceramics. Each waste type will be bagged and then placed on the balance to obtain its preshredded mass. The shredded material will then be weighed to estimate the loss of material incurred during shredder operations. A predetermined number (see Ref. 6) of samples from each waste category will be obtained and loaded into test tubes, double bagged, and transferred out of the glove box. The waste samples will be assayed using a gamma-ray spectrometer and, as mentioned previously, the results compared to those obtained from the neutron and gamma-ray nondestructive assay techniques.

The procedures describing in detail the operations of the TWDSF can be found in Reference 3.

The TWDSF will begin examination of ORNL waste drums after specific restrictions and safety criteria have been satisfied. These criteria have been established by the ORNL Radiation Operations Committee (ROC) and the Office of Operational Safety to ensure the safety of facility operators and the containment of radioactive materials within the facility's glove boxes.

Some of these criteria were listed previously in this section (see drum eligibility criteria for TWDSF). Additional restrictions and safety criteria are listed below.

Fig. 18. ORNL TRU Waste Drum Sampling Facility

Fig. 19. ORNL TRU Waste Drum Sampling Facility

Fig. 20. View Through Drum Portal

Fig. 21. Back View of Glove Box Assembly

1. Respiratory protection shall be worn during drum transfer into and out of glove boxes.
2. Levels of transferrable alpha contamination outside the glove boxes must be maintained less than 20 dpm/dm².
3. HEPA filters will be DOP tested semiannually.
4. Differential glove box pressure gages and meters shall be calibrated annually.
5. All health physics instruments must be maintained and calibrated periodically.
6. Facility operators shall be trained; and their training shall be documented in glove box, drum transfer, and emergency procedures.
7. Glove box procedures — in particular, drum-loading procedure — must be verified by subcommittee of the ROC.*
8. Glove box hot off-gas lines must have minimal impact on existing facility's system.
9. An adequate fire-suppression system must be installed in the glove boxes.

Compliance with these and other restrictions and safety criteria has delayed the drum examination schedule.

Criteria 7, 8, and 9 listed above remain to be satisfied before final approval for waste drum examination is granted by the ROC. This is expected to be accomplished in early 1984.

WASTE MATRIX COMPENSATION

The results obtained from any neutron or gamma-ray assay system are subject to various matrix effects. A considerable amount of matrix

*Two nonradioactive drums have been run through the TWDSF to train personnel and uncover procedural deficiencies.

information is endemic to the pulsed active neutron assay measurements conducted by the NAS.

The matrix effects on both the interrogating (thermal) and signal (prompt-fission) neutrons are attributable to 1) thermal neutron absorption and 2) fast neutron moderation.

The thermal neutron absorption effects are due to the thermal neutron absorption properties of the different materials contained in the waste drum. For example (Ref. 9), a drum containing 220 kg of iron scrap would reduce the measured thermal neutron half-life within the assay chamber from 700 μ s (empty chamber half-life) to approximately 370 μ s. In general, the greater the drum's total macroscopic absorption cross-section, the greater will be the interrogating thermal neutron flux reduction.

The primary moderator effect is that of producing additional thermal neutrons directly within the drum (these are in addition to the interrogating thermal neutrons coming from the assay chamber walls). Hydrogenous materials through elastic scattering reactions with the 14-MeV neutrons and iron and lead inelastic and (n, 2n) reactions will add an extra component to the interrogating thermal flux.

A technique based on the pulsed thermal neutron flux monitor data for matrix compensation has been established.¹⁰ Two parameters, termed matrix identifier factors, can be extracted from the data that, respectively, are single functions of the matrix-moderating mass and the matrix absorber mass. These two parameters, the extrapolated zero-time thermal neutron flux intensity (A_0) and the thermal neutron lifetime ($T_{1/2}$), are obtained from an exponential least squares fit to the thermal neutron flux monitor data.

The data shown in Fig. 22 are presented in terms of the differential matrix identifier quantities; that is, the differential-time zero flux

Fig. 22. Waste Drum Matrix Correction Factors

intensity (ΔA_0) relative to an empty drum versus the differential thermal neutron lifetime ($\Delta T_{1/2}$) relative to a 208-L drum containing neither bulk material nor any activity (empty drum). The presence of absorbing materials in a drum produces a decrease in $T_{1/2}$ and, consequently, $\Delta T_{1/2} = T_{1/2} \text{ (empty drum)} - T_{1/2} \text{ (waste drum)}$.

A representative sample of ORNL TRU waste drums is depicted by the black dots. The 14 matrix standards are outlined by ovals. Some of the matrix standards studied included 208-L drums filled with aluminum scrap, sand (SiO_2) plus vermiculite, polyethylene scraps, raschig rings, dry rags, wet rags, iron scrap, concrete, and various mixtures of raschig rings and polyethylene scraps. The numbers outlined by the ovals are the measured matrix correction values required to obtain the proper assay of a standard fissile sample positioned within the indicated standard matrix (see Fig. 23).

A less empirical approach to obtaining the waste matrix correction factors has recently been undertaken. The matrix identifier quantities A_0 and $T_{1/2}$ generate a series of equations called the matrix response factors, K . For a given range of $T_{1/2}$, K is a single function of A_0 . For example,

$$\text{for } 360 \mu\text{s} < T_{1/2} < 400 \mu\text{s}, K = 1.2 + 0.0012 (A_0 - 2800).$$

No corrections are required for $T_{1/2} > 450 \mu\text{s}$; in other words, ($K = 1.0$). The linearity expressed by the above equation will exist for drums containing up to hundreds of grams of fissile material. One must keep in mind that the system was designed to assay for low-fissile-content waste (1-mg to 1-g range) and that problems can arise in high-fissile-content waste. However, the linearity deteriorates when the fissile material is not homogeneously distributed throughout the waste matrix but exists in significant dense concentrations. Also, assay measurements become difficult for matrices containing dense, highly enriched fissile source materials such as highly

Fig. 23. Interior View of Calibration Drum

enriched uranium and plutonium. The data presented in Table 2 illustrate the above concerns. The effect shown is attributable to the ability of the source thermal neutrons to penetrate only a thin layer of atoms on the surface of the highly enriched uranium sources. The large majority of uranium atoms are, therefore, not exposed to the thermal neutron flux and, consequently, do not fission (self-absorption).

The uranium oxide (U_3O_8) sources used in obtaining the data presented in Table 2 contained 99.887% enriched ^{235}U — a very high enrichment. The fissionable ^{235}U was, therefore, in a dense concentration and not "diluted" with essentially nonfissionable ^{238}U (thermal neutron fission cross-section of only 3 μb as compared to 584 b for ^{235}U), as was the case for the D-38 calibration source (see table). Two computer programs were used to calculate the milligram equivalents of ^{235}U contained in each source (for this case no additional calculations were required to convert to a different isotope), DDT-2 and ANEUT.

Table 2. NAS measurements of uranium-235 sources

^{235}U source (mg ^{235}U)	Calculation method	Average detected ^{235}U (mg)	Percent of known value
25	DDT-2	17 ± 2^a	69
	ANEUT	17 ± 3^b	66
50	DDT-2	31 ± 3	62
	ANEUT	32 ± 3	63
75	DDT-2	46 ± 2	62
	ANEUT	46 ± 1	61
100	DDT-2	59 ± 1	59
	ANEUT	60 ± 4	60
150	DDT-2	75 ± 2	57
	ANEUT	84 ± 2	56

Table 2. NAS measurements of uranium-235 sources (cont.)

^{235}U source (mg ^{235}U)	Calculation method	Average detected ^{235}U (mg)	Percent of known value
200	DDT-2	111 ± 3	56
	ANEUT	116 ± 3	58
250	DDT-2	140 ± 3	56
	ANEUT	141 ± 3	56
300	DDT-2	171 ± 5	57
	ANEUT	172 ± 3	57
350	DDT-2	205 ± 1	59
	ANEUT	206 ± 5	59
400	DDT-2	233 ± 4	58
	ANEUT	236 ± 3	59
500	DDT-2	473 ± 4	95 ^c
	ANEUT	476 ± 3	95

^aStandard deviation of six measurements

^bStandard deviation of five measurements

^cD-38 source in which ^{235}U is diluted with ^{238}U

From the table one observes that the highly enriched and fission-dense ^{235}U sources do indeed create problems for the neutron assay system.

The D-38 source, however, being diluted with essentially non-fissionable ^{238}U , is accurately assayed.

DISCUSSION OF RESULTS

To date, 171 ORNL TRU waste drums have been assayed by NAS. The assay includes the pulsed active neutron interrogation which determines the transuranic fissile mass content, and the passive neutron measurements

which determine the spontaneous fission transuranic isotope content as well as an upper-bound estimate of the total contained alpha activity. The passive neutron scan also includes neutron coincidence and neutron multiplicities measurements.

Table 3 depicts 160 of the assayed drums arranged in categories according to their total passive neutron count rate (n/s) and the fissile mass content (milligram equivalents of ^{235}U). These data are uncorrected for any matrix effects (neutron absorber and moderator effects). The passive and pulsed active measurements can be performed at the 20% level of accuracy without the use of matrix compensation. Approximately 70% of the waste drums require little or no matrix compensation; and, consequently, the results should not change significantly. The remaining eleven drums contain high spontaneous fission and/or (α/n) neutron sources and difficult-to-detect amounts of fissile mass and, consequently, are listed separately in Table 5.

If the neutron source intensity is sufficiently low (<20 n/s) and if, simultaneously, the fissile mass of the drums is less than 100 mg, these data suffice to define the drum as non-TRU.¹¹ Nine drums (5% of the drum population) have been classified as non-TRU using the above definition.

Table 3. NAS data summary for ORNL TRU waste drums¹²

Passive neutron count rate (n/s)	Fissile mass ^a (milligram equivalents of ²³⁵ U)						Total
	0-15	15-50	50-100	100-300	300-10 ³	>10 ³	
<10 ¹	8	0	1	0	1	0	10
10 ¹ - 10 ²	0	0	0	3	0	2	5
10 ² - 10 ³	1	1	2	2	8	12	26
10 ³ - 10 ⁴	4	4	2	0	6	12	28
10 ⁴ - 10 ⁵	11	11	5	6	2	13	48
10 ⁵ - 10 ⁶	10	12	2	3	1	3	31
>10 ⁶	4	1	5	1	1	0	12
TOTAL	38	29	17	15	19	42	160

^aSee Appendix B for method of calculation.

Table 3 indicates that approximately 50% of the drums are potential candidates for the non-TRU category based only on a fissile inventory of less than 100 mg (assuming that the fissile signal is due entirely to ²³⁹Pu). However, all but a few of these drums contain large passive neutron sources (>10⁴ n/s). To separate the spontaneous fission and (α ,n) non-TRU contributions from the TRU content in these drums is a formidable task. Use of neutron multiplicity data and gamma-ray quantitative spectral analyses can lead to the certification of more waste drums containing less than 100 nCi/g TRU concentration (perhaps up to 20% of drum population). For example, drums containing ²⁵²Cf and/or ²⁴⁴Cm with negligible amounts of other alpha emitters would be classified as such.

Of the 171 drums assayed thus far, nine drums have been determined to contain less than 10 nCi/g TRU isotopes (well below the TRU definition) based on the combined passive and pulsed active neutron measurements. These low-level waste drums are listed in Table 4. This represents 5.3% of the drum population assayed thus far.

Table 4. ORNL waste drums containing less than 10 nCi/g TRU activity

Drum ATN ^a	Total TRU isotopic mass (mg eq. ²³⁵ U)	Total neutron source strength (n/s)
1945	3 ± 1	<4
1921	2 ± 1	<4
1924	2 ± 1	<4
1922	5 ± 2	<2
1919	6 ± 2	<2
1208	2 ± 2	<2
1788 ^b	74 ± 5	<3
2101	17 ± 2	<5
1923	2 ± 1	<5

^aATN denotes drum accountability number.

^bThe passive gamma-ray data were used to establish the principal TRU isotope to be ²³³U.

The waste drums listed in Table 5 emit a very high neutron background level (greater than 1×10^5 n/s) which tends to mask the fissile mass content (if any) of the drum. However, a reduced variance calculation can extract the spontaneous fission neutrons from these data leaving only the fissile mass content of the drum. Work is in progress in this area.

Table 5. Strong passive neutron source waste drums

Drum ATN ^{<i>a</i>}	Neutrons/s
2283	7.58×10^5
2291	1.44×10^6
2292	2.77×10^6
2098	3.34×10^6
2357	6.89×10^5
2345	6.09×10^6
2092	1.25×10^6
2401	6.37×10^6
2404	4.78×10^6
2080	1.89×10^5
2407	1.10×10^5

^{*a*}ATN denotes drum accountability number.

Table 6 illustrates the internal consistency of the pulsed active neutron measurements which were made on a single drum spanning a three-month period. This intrinsic repeatability demonstrates the basic high quality of the neutron data.

Table 6. Background-corrected pulsed active neutron data
for Drum 1772

Date	Corrected counts ^a	A _o	T _{1/2}
7/16/82	3.37	4500	509
7/22/82	3.33	4264	521
7/22/82	3.30	4232	524
7/22/82	3.30	4232	521
7/22/82	3.33	4255	521
7/22/82	3.34	4367	519
9/22/82	3.28	4530	518
9/22/82	3.28	4613	514
Average	3.32 ± 0.03	4387 ± 134	518 ± 4

^aThis term is directly proportional to the fissile
mass content of the waste drum.

Discussion of Results: Gamma-Ray Drum Scanner Analyses

Qualitative gamma-ray scanning of 171 ORNL TRU waste drums has identified a multitude of gamma-ray-emitting isotopes (TRU and non-TRU) contained in the waste. Table 7 shows the isotopic (TRU and non-TRU) distribution (alpha-emitting isotopes and fission products) for 114 of the waste drums scanned.

Table 7. Distribution of isotopes in ORNL waste drums¹³

Isotope	Frequency of occurrence (drums)	Isotope	Frequency of occurrence (drums)
<u>Alpha-Emitting Isotopes</u>			
²¹¹ Pb	8	²³⁹ Pu	36
²²⁷ Ac	1	²⁴¹ Pu/ ²³⁷ U	40
²²⁷ Th	1	²⁴¹ Am	101
²²⁸ Th	42	²⁴³ Am/ ²³⁹ Np	67
²³¹ Pa	1	²⁴⁴ Cm	54
²³³ U	11	²⁴⁵ Cm	2
²³⁵ U	9	²⁵⁰ Bk	5
²³⁷ Np/ ²³³ Pa	29	²⁴⁹ Cf	44
²³⁸ Pu	2		
<u>Fission Products</u>			
⁶⁰ Co	39	¹³⁴ Cs	60
¹⁰⁶ Ru/ ¹⁰⁶ Rh	43	¹³⁷ Cs	97
^{110m} Ag	13	¹⁴⁴ Ce	25
¹²⁵ Sb	25	¹⁵⁴ Eu	53

All isotopes listed in Table 7 with the exception of ²⁴⁴Cm were identified directly through detection of an emitted gamma-ray. The gamma-rays emitted by ²⁴⁴Cm have a very low probability of occurring ($< 2 \times 10^{-5}$ gamma-rays per 100 alpha decays) and, therefore, cannot be detected. However, ²⁴⁴Cm alpha decays to ²⁴⁰Pu with high probability. The energetics of a curium-plutonium quantum mechanical system are sufficient for plutonium K X rays to be emitted. However, the K X-ray region of a spectrum (75 keV to 120 keV) is complicated and oftentimes obscured by the lead K α X rays produced by the detector's

collimator/shield. To alleviate this problem, the detector's lead shielding has been lined with a thin sheet of cadmium and a thick sheet of copper (3/8-in. thick). These low atomic number (low Z) materials absorb the lead X rays and minimize the interference with the low-energy gamma-rays.

Quantitative studies of the waste drums using the GRDS have been initiated. A mixed-europium source, $^{152,154}\text{Eu}$, has been selected as the transmission source. The energy region spanned by this source (123 keV to 1408 keV) and its relatively long half-life [$t_{1/2}(^{152}\text{Eu}) = 13.4$ y; $t_{1/2}(^{154}\text{Eu}) = 8.2$ y] makes it an ideal transmission source. Since the gamma-ray transmission varies smoothly and slowly as a function of energy for gamma-rays whose energies are greater than 120 keV, a plot of $(-\ln T)$, where T is the gamma-ray transmission as a function of $(\ln E_\gamma)$, is a smooth curve. A linear least squares fit to the transmission data when plotted in this manner permits interpolation of the transmission for any desired gamma-ray energy.

As seen from Table 7, ^{154}Eu is a fission-product isotope contained in a number of drums. Consequently, a complete analysis of the drum would require two scans, a passive (no transmission source) and an active scan with the europium transmission source. The difference between the photopeak area from the two scans provides the transmission measurement.

Table 8 presents the results obtained from the gamma-ray transmission measurements of five selected waste drums.

The results¹³ obtained for the TRU isotopes ^{241}Am and ^{239}Np that are better than a 100-nCi/g sensitivity indicate that these isotopes can be quantitatively identified in the desired range. The NAS cannot yield quantitative results for the isotopes ^{241}Am and ^{239}Np since the thermal neutron fission cross-sections are vanishingly small (3.2 b and <1 b, respectively). Many of the fission products seen in these

Table 8. Assay results for selected TRU waste drums¹³

Isotope	Drum (contents in nCi/g)				
	1921	2008	2015	2046	2151
²²⁸ Th	-	-	-	-	(2.06 ± 0.23) × 10 ⁻²
²³⁵ U	-	-	-	-	(6.47 ± 2.59) × 10 ⁻⁴
²³⁷ U	-	(2.06 ± 0.34) × 10 ⁻¹	(1.63 ± 0.36) × 10 ⁻¹	(3.33 ± 0.66) × 10 ⁻¹	(1.29 ± 0.26) × 10 ⁻²
²³⁹ Pu	-	(4.35 ± 0.91) × 10 ²	(2.17 ± 0.54) × 10 ²	(6.19 ± 1.36) × 10 ²	(5.69 ± 1.14) × 10 ¹
²⁴¹ Pu	-	(8.56 ± 1.71) × 10 ³	(6.42 ± 1.28) × 10 ³	(1.32 ± 0.20) × 10 ⁴	-
²⁴¹ Am	3.70 ± 1.48	(4.93 ± 1.97) × 10 ²	(1.75 ± 0.70) × 10 ²	(5.91 ± 2.36) × 10 ²	(1.14 ± 0.46) × 10 ²
²³⁹ Np	-	-	(7.61 ± 1.54) × 10 ²	-	-
¹²⁵ Sb	-	(3.26 ± 1.86) × 10 ⁻³	-	-	-
¹³⁷ Cs	(1.8 ± 0.18) × 10 ⁻³	(1.11 ± 0.11) × 10 ⁻³	(5.85 ± 0.59) × 10 ⁻³	(8.53 ± 0.85) × 10 ⁻⁴	-

drums are detected at the 100-nCi/g level, including ^{60}Co , ^{125}Sb , $^{134,137}\text{Cs}$, and ^{154}Eu . The fission-product isotopes cannot be assayed with a neutron interrogation system.

An example of synergism between the NAS and the GRDS can be found in the isotopes ^{228}Th and ^{233}U . Uranium-233 emits approximately 40 gamma-rays, but with probabilities of only 4×10^{-5} gamma-rays per 100 alpha decays. The gamma-rays emitted from the decay of ^{228}Th , however, are much more prominent. Due to the thorium-uranium fuel cycle, the presence of ^{228}Th indicates the possible presence of ^{233}U . The ^{228}Th information from the gamma-ray spectrum for Drum 2151 alerts the NAS to the possible presence of ^{233}U , even though ^{233}U gamma-rays are not detected. The NAS is more sensitive to ^{233}U than is the gamma-ray system.

TECHNOLOGY TRANSFER

One of the major objectives of this program, as stated previously, is to provide a demonstration and training facility for personnel from other DOE sites and contractors. In accordance with this directive, ORNL has hosted many visitors to the TWDAF. The chronologically arranged listing below demonstrates the significant amount of interest engendered by the neutron and gamma-ray assay systems.

April 1982 - Fourteen Japanese business persons interested in radioactive waste operation visited the site on April 15. D. B. LeClair and W. C. Frankhauser visited the site on April 29.

May 1982 - Five scientists from the People's Republic of China toured the site on May 4; Mr. J. Martin of the USNRC toured the site May 13. J. J. Beauchamp and T. W. Wright of the ORNL Statistical Analysis group visited the site May 14. Deputy Assistant Secretary for Nuclear Materials, Dr. C. Gilbert, visited the site May 21. J. D'Ambrosia, DOE/HQ, reviewed the facility on May 26. J. R. Welch and K. Gasper of Rockwell-Hanford visited the site on May 27.

June 1982 - A. T. Duff of the Australian AEC visited the site on June 1. G. D. Lehmkuhl and R. T. Scott of Rockwell RFP/TWSO reviewed the program June 10-11. K. Burke of the USNRC; M. S. Glen, R. A. Almero, R. E. Raera, C. C. Reyes, R. A. Savalleno, and A. Ver Albano of the Phillipine AEC toured the site on June 11. D. D. Fischer of Rockwell-Hanford and J. P. Hinckley of EG&G, Idaho, visited the site June 17-18. B. J. Campbell, N. Allen, and R. J. Green of DOE; and R. L. Chim, L. M. Gray, D. J. Inman, and H. C. Austin of ORNL toured the site on June 17. A. K. Jowdy, W. D. Woods, and T. L. Gregory of R. M. Parsons; A. L. Ayers and J. E. Langford of EG&G, Idaho; R. R. Wright and M. Littleton of DOE-Idaho; and W. W. Pitt of ORNL visited the site June 22. A review of the system was presented to the representatives of the Korean AERI June 22.

July 1982 - J. K. Wobser, G. M. Traverso, J. H. Griffin, and J. M. Redfield of SRL visited the site on July 29 to discuss SRL's proposed plans for construction of an NAS at their site.

October 1982 - K. E. Plummer and R. S. Lee of AGNS visited the site on October 20 for an orientation of the TWDAF.

November 1982 - A. K. Williams, W. B. Sumner, and L. L. Thomas of AGNS visited the site November 11. R. G. Kurtz and M. E. Hughes of Rockwell International, RFP, visited the TWDAF November 2-3 for a demonstration of the system. E. Ricci and L. Fields of ORGDP visited the TWDAF on November 16 for a brief tour and demonstration of the NAS.

January 1983 - R. S. Lee, G. A. Huff, and L. J. Jordan of AGNS visited ORNL January 7 for a tour and demonstration of the TWDAF. A program review was prepared and presented to G. Oertel on January 27 at ORNL.

February 1983 - R. A. Wolle of the Tennessee State Governor's Office visited ORNL February 15 for a tour and demonstration of the TWDAF. G. D. Hurley and L. S. Kee of EG&G, Idaho, visited the site February 22. An abstract entitled "Test and Evaluation of a High-Sensitivity Transuranic Waste Assay System" was submitted to the Institute of Nuclear Material Management's Twenty-Fourth Meeting to be held July 10-13, 1983. A summary entitled "An Integrated Neutron and Gamma NDA Technique for Examination of ORNL TRU Waste" was submitted to the Canadian Nuclear Society's Fourth Annual Conference to be held June 15, 1983.

March 1983 - J. Hinckley of EG&G, Idaho, visited ORNL March 30 for a tour and discussion of the improvements to the TWDAF.

April - May 1983 - The report The Statistical Sampling Plan for the TRU Waste Facility, ORNL/CSD/TM-203, was submitted for publication. Twelve scientists from the National Academy of Sciences visited the TWDAF on April 21 for a tour and demonstration. M. Price of the UKAEA visited the site on May 12. Twenty-four Japanese scientists visited the TWDAF May 20 for a tour and demonstration. A paper entitled "ORNL TRU Waste Assay System" was presented at the TRU Waste Assay Workshop held in Los Alamos, New Mexico, May 23-24, 1983.

June 1983 - A report coauthored by Los Alamos National Laboratory and ORNL personnel entitled Multi-Isotopic Gamma-Ray Assay System for Alpha-Contaminated Waste, LA-UR-83-1931, is in final review before publication. A report entitled ORNL TRU Waste Drum Assay and Sampling Facilities: Operational Procedures Manual, ORNL/CF-83/253, was written and is being reviewed prior to publication. George Lehmkuhl, TWSO Rocky Flats Plant, visited ORNL June 14-17 to review the progress of the TWDAF and TWDSF. A paper entitled "An Integrated Neutron and Gamma NDA Technique for Examination of ORNL TRU Waste," ISSN-0227-0129, was presented at the Fourth Annual Canadian Nuclear Society Meeting held June 15 in Montreal, Canada.

A monthly newsletter entitled "TRU Waste Assay Facilities Update: A Monthly Report on Operational Activities at ORNL" has recently been initiated. This publication informs those interested parties concerning the general activities, engineering and design, NAS and GRDS system hardware, and computer software changes occurring within the TWDAF. Also included are sections covering the TWDSF and technology transfer (documentation and visits to either facility). The first newsletter, which was a chronological review of the period March 1982 through June 1983, was published in September 1983.

TWDAF OPERATING EXPERIENCE AND SYSTEM PERFORMANCE

Approximately one year was required for adequate planning of all program phases before installation of the NAS at ORNL. The design of the TWDAF required three months to complete. Construction of the facility, which included installation of building heating and air conditioning, 120-V and 208-V power for the computer, electric forklift recharger, wallboard and suspended ceiling, lighting, office expansion, concrete shielding walls, heat-detection and fire-alarm system, and resurfacing the floor took place in approximately six months.

Before the NAS was delivered to ORNL, ORNL personnel traveled to Los Alamos National Laboratory for training in the principles of the differential die-away technique and actual hands-on experience with the instrumentation. This allowed the writing of preliminary procedures and safety assessments for quality assurance documentation essential for all ORNL operations and thus resulted in large time savings. The installation of the NAS assay chamber and its associated detector hardware, signal and neutron generator electronics, and computer hardware went smoothly. It was virtually completed in two days.

A normal supply of spare parts is essential to the operation of a facility of this type. We maintain a stock of ^3He proportional counters for the detector packages and spare computer boards for the

LeCroy 3500 data acquisition system. Also, we possess a complete functioning neutron generator package in addition to the one operating at present in the assay chamber. Spare NIM and CAMAC modules, such as linear amplifiers and dual gate generators, are stored on site.

To date, only one detector preamplifier and one ^3He proportional counter tube have been replaced in the NAS. The original neutron generator tube was replaced soon after operations began in April 1982. However, after further investigation, it was soon evident that the neutron generator control circuitry had malfunctioned, not the neutron generator tube. The entire control panel was replaced and has operated flawlessly ever since. The neutron generator has operated steadily with neutron output levels constant within $\pm 10\%$ of an average value.

The electronic noise levels (noise in addition to that caused by neutrons) encountered within the NAS have been minimized by installing additional grounding wire throughout the aluminum frame of the assay chamber. Also, cross-communication between the power and signal cables, which were enclosed in separate conduits, has been eliminated by transferring the detector linear amplifiers from the control room to atop the NAS chamber and, thereby, considerably decreasing the length of the signal cables from the preamplifiers to the linear amplifiers.

One persistent problem involving the stability of the LeCroy 3500 data acquisition system still remains. The system tended to "freeze up" or "crash" and not respond to operator commands. The entire system was then returned to LeCroy and the internal power supply replaced. The "freeze-up" problem has diminished, but still remains. The problem is tenable and does not cause significant operational shutdowns (approximately 5% downtime due to computer malfunction).

From our operating experience with the NAS and GRDS, some invaluable lessons have been learned and are summarized below.

1. Adequate shielding is required on NAS chamber and adjacent storage areas to minimize interference from stored waste drums.
2. Uninterruptable, clean power is a necessity for the LeCroy 3500 data acquisition system.
3. The assay chamber's aluminum support frame must be well grounded in order to minimize contributions to the electronic noise level associated with the detector packages.
4. Power and signal cables must be prevented from cross-communication to minimize their contribution to the electronic noise level.
5. Adequate air conditioning is required for the electronic equipment and the computer system.
6. A supply of standard sources for equipment calibration checks (^{235}U , ^{239}Pu , ^{252}Cf , etc.) are needed.

Overall, for the period April 1982 to September 1983, the NAS and GRDS have performed very well and with a high degree of dependability.

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APPENDIX ANAS D+T Neutron Generator System¹⁴

The Sandia neutron generator system, Model MA165, used in the ORNL neutron assay system, consists of a neutron generator package and an electronic control chassis.

The system will produce approximately 1.0×10^6 neutrons per pulse (10–15 μ s-wide pulses) at a pulse repetition rate of 1 to 100 Hz for a preset number of pulses up to 9,999. The control chassis houses two high-voltage pulse-forming networks, a reservoir control circuit which maintains neutron tube gas pressure, a pulse generator, and logic circuitry to control and count the neutron bursts that are produced.

The neutron generator package used in the NAS assay chamber consists of a neutron tube (called a Zetatron), a 600-gauss permanent magnet, a 120-kV pulse transformer, and two resistors. These components are housed in a 10-cm OD $\times$ 42.5-cm-long lucite cylinder which is filled with fluorinert, a dielectric fluid which is used for high-voltage hold off and heat conduction.

A schematic of the Zetatron is shown in Fig. 24. The reservoir control circuit located in the control chassis initiates a pulse which heats the reservoir containing the zirconium deuterotritide (50% D, 50% T) powder and produces deuterium-tritium gas. The ion source is then pulsed to determine if the gas pressure has increased enough to produce ionization of the deuterium-tritium gas.

When the proper gas pressure has been reached and stabilized, a high voltage can be applied to the target of the tube in order to accelerate the ions into the target (molybdenum substrate with a scandium

ZETATRON

Fig. 24. Deuterium-Tritium Neutron Generator Tube (Zetatron)

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deuterotritide film) and produce 14-MeV neutrons. A portion of each ionizing pulse is fed back to the reservoir control circuit, which then utilizes the signal to maintain the correct operating gas pressure in the neutron tube.

The expected lifetime of the neutron generator tube is approximately 5×10^7 pulses (~100-200 h at 100-Hz pulse rate). This corresponds to approximately 10,000-20,000 assays (2000 pulses per assay).

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APPENDIX B

The equation used in calculating the fissile mass contained in each waste drum is given below. This equation has been incorporated into the newest version of the pulsed active neutron interrogation computer software program called ANEUT. It yields accurate results for approximately 70% of the waste drums assayed. However, the remaining 30% of the waste drums require a matrix correction factor to give accurate fissile mass content.

$$\text{Fissile Mass (milligram equivalents of } ^{235}\text{U)} = K_1 \left(\frac{A-B}{C} \right) - K_2 ,$$

where

A $\equiv$ total counts from shielded detector packages in the time interval 0.71 ms to 4.70 ms,

B $\equiv$ total counts from shielded detector packages in the time interval 5.71 ms to 9.70 ms,

C $\equiv$ total counts from assay chamber flux monitor in time interval 0.71 ms to 4.90 ms,

K₁ $\equiv$ proportionality constant determined from calibration standard (K₁ = 500 mg),

K₂ $\equiv$ background correction (K₂ = 13 mg) determined from empty drum in assay chamber.

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