Conf-830660--1

ORNL TRU WASTE ASSAY SYSTEM*

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*Research sponsored by the Office of Defense Waste Management, U.S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation.

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INTRODUCTION

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. 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 TRU-waste-generating streams have contributed to the large array of TRU isotopes. The isotopes present both TRU and non-TRU, in significant quantities include ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242}Cm, ^{246}Cm, and ^{252}Cf. Some 45 additional radionuclides in lesser quantities have been identified.

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 interrogation 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 assay system utilizing the LANL neutron interrogation assay system and an upgraded gamma-ray drum scanner has been employed. This paper describes each assay technique and the results obtained from the examination of authentic ORNL TRU waste.

TRU WASTE ASSAY SYSTEM

The TRU waste assay system uses pulsed-neutron interrogation (differential dieaway technique) and passive neutron measurements. It has the capability of measuring the total passive neutrons, passive coincident neutrons, and passive neutron multiplicities. A detailed description of the system may be found in the references.[2,3]

The pulsed-neutron interrogation determines the fissile component of the drum's TRU isotope content. The coincident 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.

The passive neutron measurements provide an upper-limit estimate of total TRU activity contained in each drum. Estimates of total spontaneous fission activity and total fissile mass are also obtained. This information is usually sufficient for categorization of a waste drum.

GAMMA DRUM SCANNER

The gamma drum scanner used in the assay system is an upgraded Canberra Segmented Drum Scanner Model 2220B. The effects of vertical and radial inhomogeneities are reduced, respectively, by scanning individual horizontal segments along the vertical axis of the drum and by rotating the drum on a turntable at a constant speed. Each active assay scan is corrected for the drum's varying mass absorption coefficient by measuring a gamma source through an empty drum and the waste matrix.

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

RESULTS

The ORNL drums surveyed by the TRU waste assay system in FY82 are presented in Table 1. In this summary, the total fissile mass is expressed in milligram equivalents of ^{235}U, while the passive neutron data are summarized in terms of a total neutron source in units of neutrons/second (n/s).

Table 1. ORNL TRU waste drum data summary[4]

	Fissile mass, Uranium-235 equivalent (mg)					
Neutrons/s	0–15	15–50	50–100	100–300	300–1000	>1000
<10^1	8	–	1	–	1	–
10^1–10^2	–	–	–	3	–	1
10^2–10^3	1	–	1	1	7	7
10^3–10^4	2	3	2	–	5	7
10^4–10^5	6	8	4	6	2	7
10^5–10^6	6	9	1	2	1	1
>10^6	4	1	5	1	–	–
TOTAL	27	21	14	13	16	23

Of the 114 drums assayed to date, nine drums were determined to contain less than 10 nCi/g TRU isotopes based on the combined passive and pulsed active neutron measurements. These TRU waste drums are listed in Table 2.

Table 2. ORNL waste drums containing <10 nCi/g TRU activity

Drum ATN	Total mass (mg ^{235}U eq.)	Total neutron source (n/s) strength
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[a]	74 ± 5	<3
2101	17 ± 2	<5
1923	2 ± 1	<5

[a]The passive data were used to establish the principal TRU isotope to be ^{233}U in this drum.

To determine which drums contain greater than 10 nCi/g and less than or equal to 100 nCi/g TRU activity, a more detailed analysis of the neutron and gamma data is required.

Through an analysis of the neutron multiplicity data, one will be able to discriminate between neutrons resulting from (α,n) reactions and those originating from spontaneous fission events. For example, a high triples-to-doubles ratio (P3/P2) would indicate the presence of ^{244}Cm or ^{252}Cf. Integrating the passive gamma-ray scan data with the multiplicity data would enable the isotopes to be identified. Work is in progress in this area.

MATRIX COMPENSATION

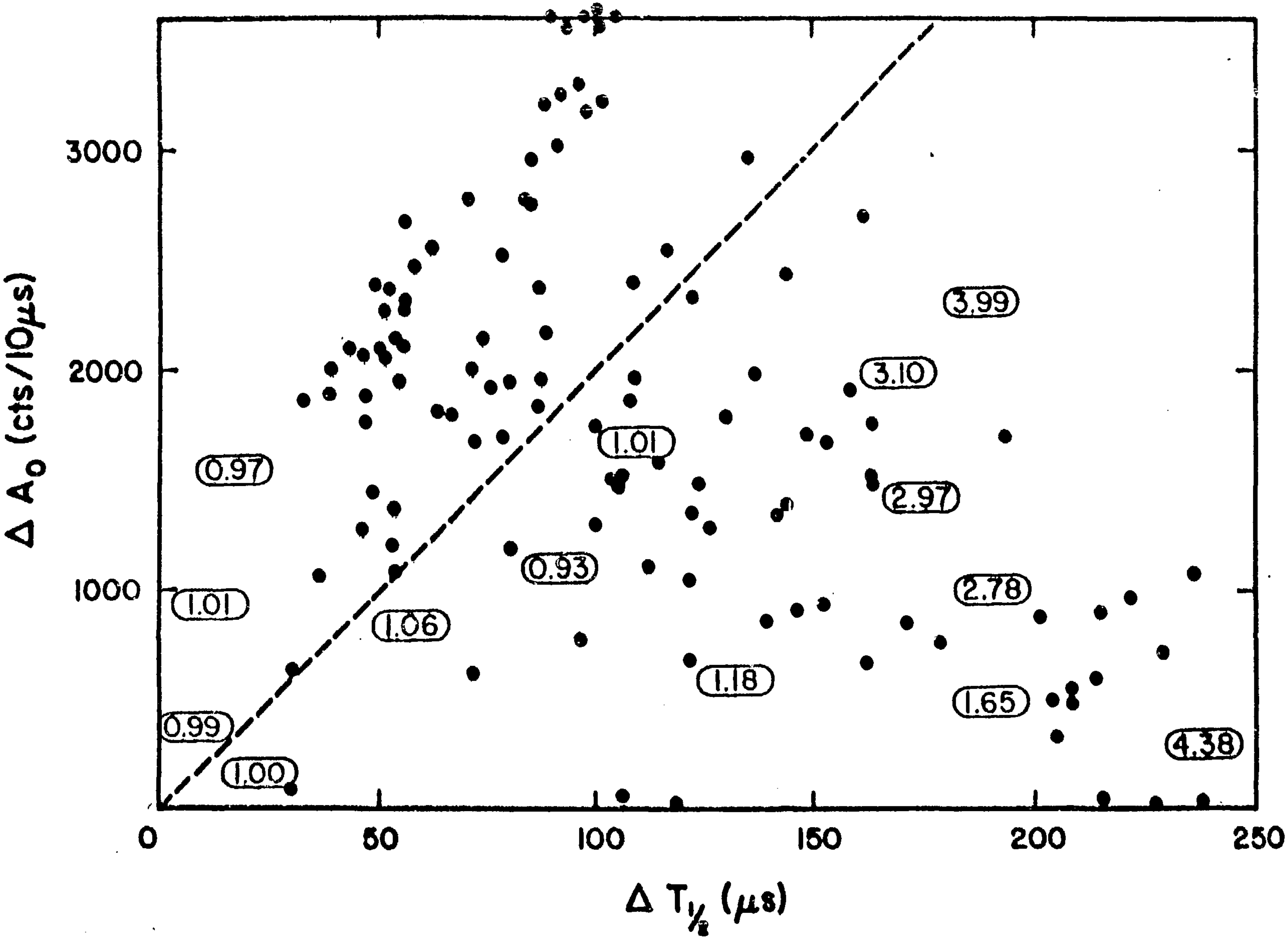
Matrix compensation corrects for the effect of the bulk content of the drums (combustibles, glassware, iron, etc.) on both passive and active neutron measurements.

A technique based on the pulsed-thermal-neutron flux monitor data for matrix compensation has been established. Two parameters 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_o) 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. 1 are presented in terms of the differential-time zero flux intensity (ΔA_o) relative to an empty drum versus the differential thermal neutron lifetime ($\Delta T_{1/2}$) relative to a 208-L drum containing no bulk material nor any activity (empty drum). The presence of absorbing materials in a drum produces a decrease in $T_{1/2}$ and, consequently, $\Delta T_{1/2} = T_{1/2}$ (empty drum) $-$ $T_{1/2}$ (waste drum).

Fig. 1. **ORNL DRUM ASSAY**

MATRIX CORRECTION FACTORS



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The 113 ORNL TRU waste drums are depicted by the black circles. The 14 matrix standards are outlined by ovals. The numbers are the measured matrix correction values required to obtain the proper assay of a standard fissile sample within the indicated standard matrix. It appears that only a relatively small portion of the ORNL waste drums will require significant matrix corrections.

REPRODUCIBILITY OF THE ACTIVE NEUTRON MEASUREMENTS

Table 3 illustrates the internal consistency of the pulsed active neutron measurements made on a single drum spanning a three-month period. This intrinsic repeatability demonstrates the basic high quality of the neutron data.

Table 3. Active neutron data for drum 1772

Date	SHTOT/FM	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	4332	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

NEUTRON MULTIPLICITY MEASUREMENTS

Table 4 presents samples of neutron multiplicity data obtained using SYSTOT (most sensitive) grouping of detectors. The term P3/P2 (ratio of triples to doubles) is a unique signature of a spontaneous fission isotope. Three of the waste drums depicted in the table indicate P3/P2 values similar to that of ^{252}Cf and thus contain appreciable amounts of that isotope. Drum 1798 has a low P3/P2 value and therefore probably contains ^{240}Pu or ^{242}Pu.

Table 4. ORNL TRU waste drum neutron multiplicity data

Drum/source	Neutrons/second	P3/P2	P1/P2
^{252}Cf	900	0.107 ± 0.004	5.9 ± 0.1
2021	1.6×10^3	0.098 ± 0.004	7.9 ± 0.1
1925	1.5×10^2	0.078 ± 0.008	10.2 ± 0.4
1947	2.3×10^2	0.090 ± 0.008	14.2 ± 0.6
1798	1.7×10^3	0.056 ± 0.008	27 ± 1
1960	1.3×10^3	0.086 ± 0.018	88 ± 8

The P1/P2 ratio (ratio of singles to doubles) is a sensitive function of the relative nonspontaneous fission neutron source content. Drum 1960's P1/P2 value indicates an excess of nonspontaneous fission TRU isotopes relative to spontaneous fission TRU isotopes.

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