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TEST AND EVALUATION OF A HIGH-SENSITIVITY ASSAY SYSTEM FOR BULK TRANSURANIC WASTE*

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ABSTRACT

During the past year, we tested and evaluated the performance of an assay system that accommodates 55-gal. drums of transuranic waste at Oak Ridge National Laboratory. This neutron assay system provides a routine assay of fissile transuranic isotopes to the 1-mg sensitivity level with a pulsed active neutron interrogation based on the differential dieaway technique. A highly sensitive passive neutron measurement determines the content of spontaneous fission transuranic isotopes in each drum as well as an upper-bound estimate of the total alpha activity. All components of the combined, pulsed active and passive neutron assay system performed well on a routine basis during the test-and-evaluation period. We performed more than 400 combined passive and active assay measurements of waste drums at Oak Ridge. One-fifth of the initial set of waste drums measured contain less than 100 nCi/g of total transuranic isotopes and thus qualify locally as nontransuranic waste.

INTRODUCTION

The neutron assay system consists of both pulsed active neutron interrogation (differential dieaway technique)[1-3] and passive neutron measurements, including passive neutron coincidence, passive neutron totals, passive neutron multiplicity determinations, and reduced variance. A description of the system, including the physics of the measurements, may be found in articles by Kunz et al.[4] and Caldwell et al.[5]

The pulsed active portion of the assay determines the fissile component of a drum's transuranic (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 quantity. The reduced variance measurement is useful in quantifying very intense spontaneous fission sources.

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The TRU waste at Oak Ridge is complex in isotopic makeup with more than 20 TRU isotopes having been identified.[6] This complexity in isotopic makeup creates a correspondingly complex assay problem. For some waste drums it will not be possible to quantify all TRU isotopes present, although the additional information provided by the segmented gamma-ray scanning system[6] will be helpful. In all cases, however, the neutron assay system provides an upper limit estimate of total TRU activity. In addition, estimates of total spontaneous fission activity and total fissile mass are obtained. This set of information, while incomplete, will suffice to determine the appropriate waste category for most drums, i.e., non-TRU (less than 100 nCi/g), or TRU (greater than 100 nCi/g).

The bulk of the test-and-evaluation data were taken for a set of 114 potential TRU waste drums currently held in storage at Oak Ridge. In general, several independent measurements were made of each drum.

INSTRUMENTATION, DATA ACQUISITION, AND DATA PROCESSING

Passive Neutron

The basic instrumentation is described in the references.[4,5] The ^{3}He proportional counter neutron detection system comprises 11 modules, each module having a separate preamp, amplifier, and discriminator. The counts occurring in each module are recorded separately. These data prove to be quite useful for a variety of purposes, such as tracing noise problems in individual modules.

The output of the 11 modules is summed for our most sensitive totals, coincidence, and multiplicity information data set. For this summed set (designated system totals) the detection efficiency is about 14%. In addition, the six cadmium-wrapped modules are summed and this set (designated shielded totals) is also processed as a separate entity with totals, coincidence, and multiplicity data recorded. The shielded total efficiency is about 7.2%.

The passive data described above are acquired simultaneously and routinely for all waste drums and

calibration measurements. Coincidence and multiplicity backgrounds for both summed sets are measured simultaneously with foregrounds. Data processing occurs at the end of a passive neutron data acquisition run, generally about 500 s, using specialized software written for the LeCroy 3500 data acquisition system. No operator intervention other than run initiation is required. All data—foreground, background, and processed sets—are recorded on magnetic disk for a permanent retrievable record.

Such an elaborate passive neutron data acquisition and processing system is required for the Oak Ridge waste because of the large dynamic range of passive neutron sources. Of the 114 potential TRU waste drums studied to date, 32% contained neutron sources of less than 3×10^3 n/s (these drums include those most likely to qualify for the less than 100 nCi/g designation). Another 25% contained sources between 3×10^3 and 4×10^4 n/s. The remainder (43%) contained sources greater than 4×10^4 n/s, including 10% with sources in excess of 10^6 n/s.

Pulsed Active Neutron

The basis of quantifying fissile TRU isotopes is the Los Alamos-developed differential dieaway technique.[1-5] Short (15 us) pulses of originally 14-MeV neutrons are produced by an internally mounted, small neutron generator system.[8] These pulses are moderated in the drum matrix materials as well as in the graphite and polyethylene walls of the assay system in about 0.5 ms, with a subsequent lifetime (T_1/e) in the waste drum and assay chamber of 0.3 to 0.6 ms. The specially designed prompt fission neutron detection (PFND) modules (same as the shielded modules discussed in the passive neutron section) recover from the direct effects of the 14-MeV pulse in about 0.5 ms.

Thus, during the period of maximum flux of interrogating thermal neutrons within the waste drum, the PFND modules detect induced fission events. For the ^{239}Pu component of the contact-TRU waste, the observed detection efficiency of fission events is about 10%. Figure 1 shows (a) a typical interrogating thermal flux time-history, (b) the PFND system count rate time-history with no fissile isotopes present in a waste drum, and (c) the PFND time-history with about 300-mg equivalent of ^{239}Pu present in a waste drum. The basic fissile assay is obtained from the relationship [(c) - (b)]/(a).

The interrogating thermal neutron flux is measured with a small, bare ^{3}He counter placed within the assay chamber. A similar moderated and shielded ^{3}He counter placed outside the assay system, directly behind the 14-MeV neutron generator, serves to monitor the 14-MeV pulsed neutron output. The time histories of the two flux monitors and the PFND system are recorded with three separate multi-channel scaling units and are stored on magnetic disk for a permanent record. The fissile TRU isotopes contained within the 114 drums studied to date range from 0 to about 50 g in each drum. The intrinsic fissile assay sensitivity is about 1 mg[3,5] using a routine 2000 pulse assay accomplished in less than 1 min of elapsed time.

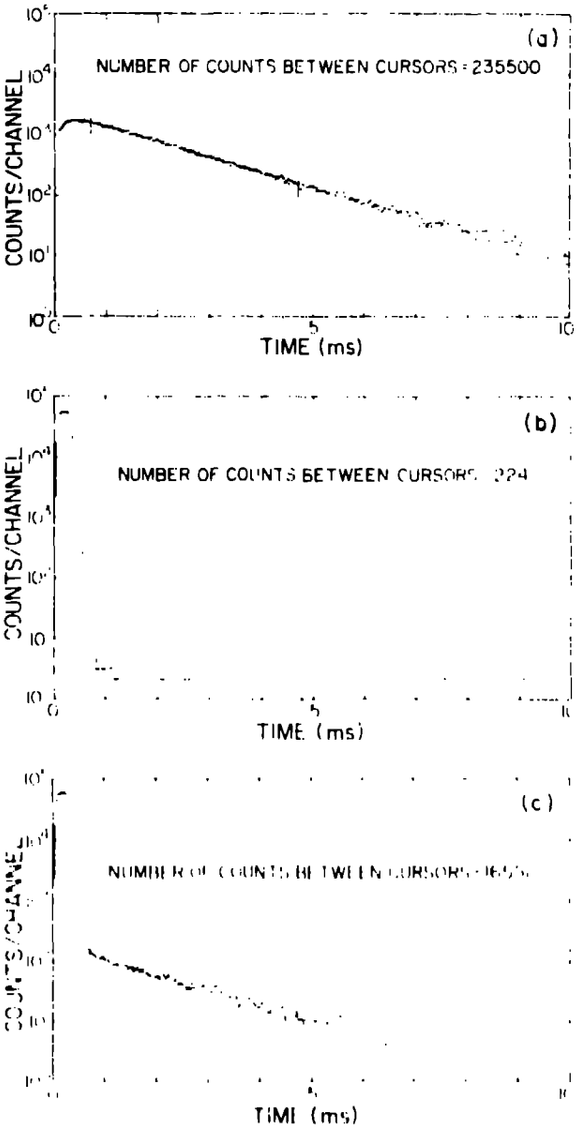

FIG. 1.
Typical time history plots recorded with the LeCroy 3500 data acquisition system. (a) The shape of the thermal neutron flux monitor data changes considerably with different waste matrices and can be used as the basis of a matrix correction to fissile assay values. (b) The PFND data with no fissile isotopes present differs dramatically from (c) PFND data with fissile isotopes present. Generally, fissile mass is proportional to [(c) - (b)]/(a), where all quantities are summed over common limits.

SCREENING PROCEDURE

A batch of potential TRU waste drums delivered to the building in which the assay system is housed (Building 7831 at Oak Ridge) generally consists of several drums that emit 10^5 to 10^6 n/s as well as

several drums that emit less than 10^4 n/s. Detailed high sensitivity passive neutron assay of the drums with less than 10^4 n/s is difficult in the presence of sources that emit more than 10^5 n/s.

We devised a simple "pre-screening" station to sort drums quickly and routinely at the approximate emission level of 5×10^4 n/s. This consists of a simple well-shielded slab counter that measures each drum in less than 1 min. Drums that emit less than 5×10^4 n/s are set aside after the prescreening. Drums that fall into the greater than 5×10^4 n/s category are then assayed immediately in the high sensitivity passive/active unit. Background perturbations are inconsequential for the drums in this high-emission category.

When all the drums that emit more than 5×10^4 n/s have been assayed, 1 cy are removed from the building, with a consequent return of normal background conditions. Then the drums that emit less than 5×10^4 n/s are assayed with high-sensitivity passive and active measurements. The nominal background for system totals ranges from 14 to 17 counts/s. With this background a routine 500-s data acquisition run results in a passive detection sensitivity of about 0.3 counts/s, equivalent to a neutron source of 2 n/s. This is an important consideration since our estimate of total alpha activity is based on the experimental observation that each milligram of alpha activity gives rise to about 2 n/s from (α,n) reactions in oxygen. (The dominant chemical form of TRU isotopes in waste is that of an oxide.)

A waste drum can thus be shown to contain less than 100 nCi/g of TRU isotopes if it produces a neutron source of less than 20 n/s (100 kg of waste) or a net detected signal less than 3 counts/s. The room background must be stable to accomplish such a low-level measurement.

DATA SUMMARY

Table I summarizes the passive and active neutron measurements for the 114 Oak Ridge drums. In this summary, the total fissile mass is expressed in milligrams equivalent of ^{235}U. Conversion of a value to milligrams of ^{239}Pu is obtained by multiplying it by 0.67. For example, a 15-mg ^{235}U mass produces the same active neutron signal as a 10-mg ^{239}Pu mass.

Considerably more information exists than can be expressed in Table I. The passive neutron data are summarized in terms of a total neutron source in units of neutrons/s. If the source intensity is sufficiently low (20 neutrons/s or lower) and if, at the same time, the fissile inventory is less than 100 mg, these data suffice to qualify the drum for the defined non-TRU category (i.e., less than 100 nCi/g). On the basis of this simplified interpretation of the passive and active neutron data, about 10% of the drums summarized in Table I can be certified for the non-TRU category.

Table I indicates that over 50% of the drums are potential candidates for the non-TRU category, based on a fissile inventory of less than 100 mg. (This presumes the fissile signal is due entirely to ^{239}Pu.) Most of these potential candidates contain sizable passive neutron sources, however, and unraveling the non-fissile TRU isotopic content in these drums is not an easy task. The Oak Ridge waste is fortunately not typical of most TRU waste in the DOE inventory, which instead contains principally plutonium isotopes.

For one category of the Oak Ridge waste we have been able to certify some additional drums as non-TRU by the use of our neutron multiplicity data. These are drums that contain ^{252}Cf and/or ^{244}Cm with negligible amounts of other alpha emitters. We use an analysis in which the measured net multiplicity rates for singles,

TABLE I

DATA SUMMARY FOR INITIAL SET OF WASTE DRUMS FROM OAK RIDGE NATIONAL LABORATORY

Passive Neutron, n/s	Fissile Mass, ^{235}U Equivalent (mg)					
	0 to 15	15 to 50	50 to 100	100 to 200	200 to 10^3	$>10^3$
$<10^1$	8		1		1	
10^1 to 10^2				1		1
10^2 to 10^3	1		1	1	2	2
10^3 to 10^4	3	1	3		1	2
10^4 to 10^5	6	3	1	6	3	2
10^5 to 10^6	1	9	1	3	1	1
$>10^6$	1	1	5	1		
Total	20	24	11	12	10	8

doubles, triples, and quadruples (P1, P2, P3, P4) are analyzed in a model composed of an arbitrary mixture of ^{252}Cf, ^{244}Cm, and a pure singles source of (α,n) neutrons. The model is conservative. Using this analysis, it is easily shown that any source of (α,n) neutrons actually present (and thus indicative of other alpha emitters) is over-represented.

It appears that about 10% of the Oak Ridge waste may be certifiable in this second non-TRU category. We use standard well-characterized ^{252}Cf and ^{244}Cm sources for proper calibration. Table II shows some representative calibration and waste drum data for this category.

A third category of Oak Ridge waste is certifiable by a simple combination of passive and active neutron data with the gamma-ray spectral data discussed in Ref. 6. In this special case only a qualitative use is made of the gamma-ray data: to identify the dominant presence of ^{233}U. (The minor isotope ^{232}U and its daughter emissions provide a suitably strong passive gamma-ray signature.) For this case a small passive neutron signal in combination with a fairly large fissile active neutron signal (1000 mg of ^{233}U in a drum is still generally an amount consistent with non-TRU status) is the indicative signature. We have certified a few such drums to date—the overall fraction of these drums in the Oak Ridge inventory is likely to be greater than we encountered in our small, 1-year sampling.

We anticipate certification of additional drums when we complete the combined passive gamma-ray, passive neutron, and active neutron analysis loop. As discussed in Ref. 6, many of the Oak Ridge TRU isotopes are strong gamma emitters that can be quantified with the segmented gamma-ray scanning system.

SYSTEM PERFORMANCE

All hardware has performed well during the 15 months to date (July 1983) of our test-and-evaluation program at Oak Ridge. The original neutron generator is still in operation; it has provided steady service with neutron output levels constant to about ±10% of an average value. We estimate that the neutron generator has produced the equivalent of about 5000 pulsed active assay runs (2000 pulses each) to date, including operation at Los Alamos for several months before it was shipped to Oak Ridge.

All 11 neutron detection modules currently display the same detection efficiency to within ±1% of the original (April 1982) values. System electronics stability and reproducibility of both passive and active assay results are impressive. Repeat assays of a drum after several months elapsed time generally agree with original results at the ±3% level or better.

The LeCroy 3500 data acquisition system is providing excellent service after repair of an apparently defective power supply.

Although detailed records of system availability were not kept, we estimate that the assay system provided better than 95% up time during the 15-month test-and-evaluation period. Almost the entire down time was due to the problem with the data acquisition system power supply.

Users and potential users of assay instruments invariably ask, "what is your assay accuracy?" A forthright answer, as any assay instrument developer will reply, is that "it depends." This answer is particularly appropriate for the assay of bulk TRU wastes.

TABLE II

PASSIVE NEUTRON MULTIPLICITY DATA AND INTERPRETATION USING THE ^{252}Cf, ^{244}Cm, SINGLES (α,n) MODEL

Drum ID	Multiplicity Rates (counts/s) P1	P2	P3	P4	Analysis Results ^{252}Cf	^{244}Cm	(α,n) Emitters	[illegible]
Background	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]			
^{252}Cf[a]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		
^{244}Cm[a]	[illegible]	[illegible]	[illegible]	[illegible]		[illegible]		
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]		[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

[a] Calibration standard.

We have demonstrated with this neutron assay system that, for more than 50% of the waste matrices encountered at Oak Ridge, our basic pulsed active and passive measurements can be performed at the ±20% level of accuracy without the use of matrix compensation. The ±20% value is determined from analysis of a considerable amount of detailed mockup waste matrix data and a neutronic comparison of the mockups with actual waste matrices. The ±20% value also assumes that the TRU isotopic content is either fairly well known or can be determined from our measurements. At present, studies are in progress[4,5] to implement routine matrix corrections based on the thermal neutron flux monitor measurement (Fig. 1a).

Nevertheless, it will always be possible to postulate circumstances where the assay accuracy will be poor. Examples of extreme cases are fissile TRU isotopes placed at the center of a water-filled 55-gal. drum or wrapped in cadmium sheeting. On a probabilistic basis, these cases will rarely occur in TRU wastes.

SUMMARY

We have successfully completed a 15-month test and evaluation of a high-sensitivity assay system for bulk transuranic waste at Oak Ridge. All hardware performed satisfactorily and reproducibly, with an overall uptime for the assay system of 95%. We measured the content of 114 waste drums with passive and pulsed active neutron techniques. About 20% of those drums can be certified in the legal non-TRU category, based on our measurements. Additional drums are likely to be certifiable when analysis of the passive gamma-ray assay data[6] is fully integrated with the neutron results.

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