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CONTENTS

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ABSTRACT

We describe in a comprehensive fashion the Los Alamos second-generation system for passive and active neutron assays of drum-size containers. The developmental history of this 7-year project is presented with emphasis on the pulsed active neutron technique (differential dieaway), which has achieved milligram levels of assay sensitivity for both plutonium and uranium wastes. We describe in detail the matrix effects for both passive and active neutron assays. We present in a thorough fashion our novel approach to achieving comprehensive corrections for these matrix effects using measurements made during the assays. We develop a matrix correction formalism based on separate neutron absorption and moderator indices determined from these measurements. These are presented as a series of analytic functions fitted to the data. Absolute calibrations and calibration standards are discussed, as is a practical means (pink drum measurements) of achieving routine calibration verification at all implementation sites. We present our overall assay algorithm, integrating absolute calibrations with matrix corrections. We also present a systematic error formalism that is based on the matrix response data. Finally, we outline a strategy for the verification of our entire assay formalism. This is based on measurements with a set of salted waste matrix drums combined with systematic assay intercomparisons of well-characterized transuranic wastes.

I. INTRODUCTION

This report is intended to serve a variety of purposes, among them the documentation of Los Alamos efforts to date in certifying its drum-size second-generation system for combined passive and active neutron assays that are in routine operation at several sites in the United States. In 1983, we began a great deal of research and development aimed at improving both the assay technique and the hardware in which it is embodied. We also performed a large number of calibration and matrix response measurements and developed systematic assay algorithms based on these data. Some of these developments were reported in recent publications and workshops. This report, however, constitutes the first comprehensive publication on the drum-size second-generation system for combined passive and active neutron assays.

In this report we present eight major topics.

1. The theoretical and experimental basis for the systematic neutron absorption and moderator matrix corrections.

2. Absolute system response measurements for various fissile and other transuranic (TRU) isotopes.

3. The second-generation assay algorithms for both passive and active measurements.

4. The system verification strategy being pursued by Los Alamos and the various site organizations.

5. System intercomparison measurements for the Idaho, Rockwell-Hanford, and Savannah River second-generation units now in operation.

6. Initial field experiences for the two units now in the test and evaluation phase [Stored Waste Examination Pilot Plant (SWEPP) and Rockwell-Hanford units] including assay results.

7. Intercomparison studies with high-resolution segmented gamma scanners.

8. Analysis of systematic measurement errors for the combined passive-active neutron assay system.

II. HISTORICAL PERSPECTIVE

In 1978, staff in the Advanced Nuclear Technology group at the Los Alamos National Laboratory initiated development work on a novel technique for pulsed active neutron assays.[1-3] In 1980, the nascent development project was brought into the TRU waste program because of its potential for 10-nCi/g level of sensitivity assay.[4-6]

At about this time, the developers were able to demonstrate a routine assay sensitivity of 1-mg quantities of either ^{235}U or ^{239}Pu placed anywhere within a standard 208-ℓ waste drum.[4-6] The active technique was then named the "differential dieaway" technique by the Los Alamos developers, a name that has

some into general usage both in the United States and internationally. The name derives from the relative neutron lifetimes of the two key elements of the active assay system: the assay chamber itself, which has a thermal neutron lifetime of about 0.5 ms, and the cadmium-wrapped, fast-neutron detection units, which have a much shorter characteristic lifetime of about 0.015 ms. It is, in fact, this large difference in characteristic dieaway times that makes possible the high-sensitivity fissile assay measurement.

In early 1981, motivated by the DOE to demonstrate a working prototype of the TRU waste assay unit, we successfully integrated a high-sensitivity passive neutron assay system within the body of the active unit so that two independent assays could be performed sequentially within the same chamber.[7] The utility of the unit increased because nonfissile TRU isotopes could now be sensed. Our concept and design for a combined active-passive neutron assay system received an Industrial Research Council IR-100 award as one of the 100 outstanding inventions of 1983. Recently a patent was awarded to the US DOE based on this design and its successful embodiment, US Patent No. 4,483,816.

The first practical prototype of the combined active and passive neutron assay system was installed at Oak Ridge National Laboratory in April 1982.[7-9] This prototype is still in routine operation at Oak Ridge where it assays an inventory of stored TRU waste drums.[10] (See also Appendix A.)

In 1982, the Transuranic Lead Office of the DOE tasked Los Alamos to develop a second-generation version of the combined assay system that would incorporate improved waste matrix corrections, would provide for the assay of drums weighing more than 1500 lbs, would improve assay algorithms and software, and would incorporate other engineering improvements. This effort was intended to provide the DOE and its contractors with a design that could be implemented successfully at the large-volume TRU waste repositories in the United States. The assay system was to measure wastes rapidly and routinely with a sensitivity such that the bulk wastes could be definitively segregated into those meeting the criteria for low-level classification and those that would be TRU waste. In addition, the measurement would be used to determine whether the container and its contents were in compliance with the Waste Isolation Pilot Plant (WIPP) regulations regarding maximum fissile loading and total alpha activity.

To shorten overall development time, Los Alamos agreed to design and construct three versions of the second-generation concept. All three would embody the same hardware design from a neutronic point of view, but would differ in mechanical design, primarily in how the required large entrance door operated and whether drums were loaded manually on dollies or semi-automatically from a fork lift onto a moving load platform. We recognized that an additional advantage would result from having three nearly identical units in the field, as the three sites where the units would be placed all have very different TRU waste characteristics. Thus, we felt that system experience with this larger variety of waste would result in a correspondingly more rapid understanding and evaluation of performance than could be achieved by implementation at a single site.

The SWEPP unit was installed in the new SWEPP building at Idaho National Engineering Laboratory in early October 1984 and is currently in an intensive test and evaluation phase. As of June 1985, some three hundred drum assays were completed. A preliminary analysis of a number of these drums appears in Sec. X-E-3 and Appendix C.

The Rockwell-Hanford unit was delivered and installed in Building 224-T at the Hanford site in February 1985. (See Appendices B and C.) This unit is also being used in an intensive test and evaluation program; the results of its first assays also appear in Sec. X-E-2. In addition, we are including the cross comparisons available from segmented gamma scanner assays of many of these same drums (Sec. X).

The Savannah River unit is completed and was in use at Los Alamos for extensive matrix response studies, as well as for assay of a few Los Alamos waste drums with low plutonium content and for cross-comparison measurements with the SWEPP and Hanford units. This unit, delivered to Savannah River in May 1985, is in a test and evaluation phase of at least 6 months duration with emphasis on wastes containing heat-source plutonium in addition to standard weapons-grade plutonium.

The DOE asked Los Alamos (FY 1986 project listing) to prepare a technology transfer document that summarizes all experiences at Los Alamos and the various sites with the combined active-passive neutron assay units. That document, due at the end of FY 86, will summarize several years of field experience with six or seven combined passive and active neutron assay systems, including both drum and crate sizes. This report describes the project status for our second-generation drum units as of June 1985 in addition to providing a detailed analysis of matrix effects, matrix corrections, and calibration procedures.

III. NEUTRONICS DESIGN FOR THE SECOND-GENERATION DRUM COUNTER

The development work that preceded the second-generation system is discussed in Refs. 1-9 and in Appendix A. Briefly, the neutron detection system consists of two types of detector packages: cadmium-shielded and bare ^{3}He detectors. The assay chamber has one shielded and one bare package in each of the six modules: the four vertical sides and the top and bottom (Fig. 1). All vertical side detectors are 91-cm-long by 5-cm-diam proportional counters filled to a 2-atm pressure with ^{3}He. The top and bottom modules contain a similar layout of intermixed shielded and bare detector packages 61 cm long by 5 cm in diameter that are filled to a 2-atm pressure with ^{3}He. Individual counting electronics sets (preamp, amplifier, discriminator) are provided for each detector package in the vertical, top, and bottom modules, for a total of 12 electronics sets.

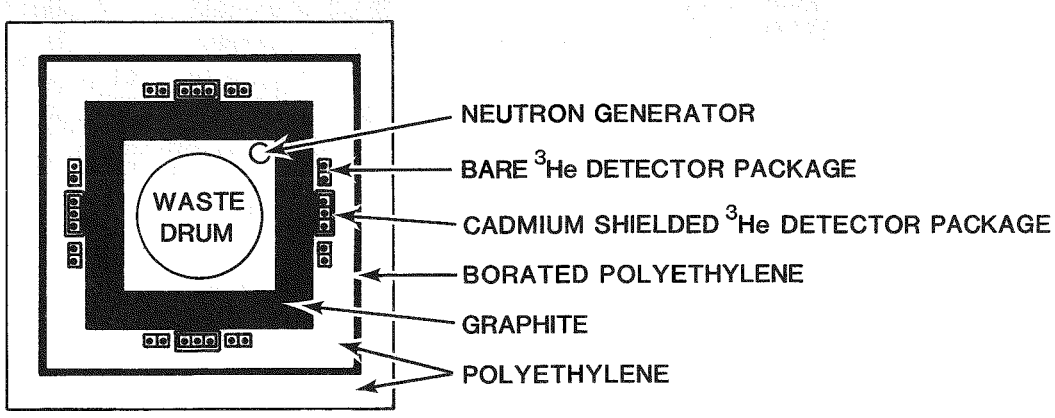

Fig. 1.
Cross-sectional views of the second-generation assay chamber show the layout and relative positions of both shielded and bare ^{3}He detector packages.

The separate neutron counts from the 12 individual detector packages are a routine portion of the system printout for each measurement (Fig. 2). For assay purposes, the six shielded packages are summed to form the "4π" shielded totals count rate. Similarly, the sum of all 12 packages is called the systems totals count rate. The systems totals rate is used for the most sensitive passive counting mode and has a nominal 4π detection efficiency for ^{252}Cf spontaneous fission neutrons of about 12.5%. The shielded totals rate is used for the active measurement and has a nominal 4π detection efficiency for fission neutrons of about 2.9%. The shielded totals rate is also used for the passive measurement.

Three passive coincidence quantities are used in this system: (a) 250-μs system totals coincidence rate, the most sensitive of the time-correlation measurements and the basis for passive plutonium assays for low count-rate situations; (b) 70-μs shielded system coincidence rate, used for passive plutonium assay at high count rates; and (c) reduced variance, a backup coincidence measurement not currently incorporated in the assay algorithm.

Two thermal neutron flux monitors are an integral part of the second-generation system. They are ^{3}He proportional counters--one shielded, the other bare--placed inside the assay chamber at different levels above the drum turntable (Fig. 3). The upper flux monitor is a 15-cm-long by 2.5-cm-diam counter filled to a pressure of 0.1 atm. The lower cadmium-shielded and collimated unit, called a barrel flux monitor, is a 15-cm-long by 2.5-cm-diam counter filled to 4-atm pressure; it is located at the rear focal line of the collimated assembly about 36 cm above the turntable. A Zetatron® 14-MeV neutron generator is also placed inside the assay chamber; its effective neutron source position is about 36 cm above the plane of the turntable.

```
PNEUT OF 4-22-85--ROCKWELL HANFORD
RUN      1   DRUM OF U          9: 7:57    4/ 1/85
GATE CORRECTION FACTORS  1.001620 (70 USEC GATES)     .996384 (250 USEC GATES) SHL
D GATE COR TIMES 1ST RAN ARE          .4100      .2969
SYST GATE COR TIMES 1ST RAN ARE        1.5800    2.4900
FOLLOWING DATA HAS BEEN BACKGROUND CORRECTED BY BACKGROU      8:52:47   4/ 1/85
 RUN      0

COUNTING TIME IS      319.41 SECONDS

     DETECTOR       COUNT      RATE          DETECTOR       COUNT      RATE

BARE DOOR 500       8430.     26.39       SHLD DOOR 501     2350.      7.36
BARE RGHT 502       7963.     24.93       SHLD RGHT 503     2425.      7.59
BARE BACK 504       7875.     24.65       SHLD BACK 505     2271.      7.11
BARE LEFT 506       7925.     24.81       SHLD LEFT 507     2365.      7.40
BARE TOP  508       4056.     12.70       SHLD TOP  509     1048.      3.28
BARE BOTM           3564.     11.16       SHLD BOTM 511     1351.      4.23
FLUX MONITOR           4.       .01       2ND FLUX MONI       20.       .06
SYSTEM TOTALS RATE       161.62       SHIELDED TOTALS RATE          36.97  (FROM PART
)
NEUTRON COINCIDENCE

SHIELDED TOTALS                    12242.+/-      110.64
SYSTEM TOTALS                      52845.+/-      229.88
1ST N 250 USEC GATES               43918.
1ST N 70 USEC GATES                11704.
RANDOM 70 USEC GATES             3194096.
RANDOM 250 USEC GATES             319410.
1ST N GATED 70 USEC TOTALS           511.
RANDOM GATED 70 USEC TOTALS         8658.
1ST N GATED 250 USEC TOTALS         8759.
RANDOM GATED 250 USEC TOTALS       13301.

RANDOM COINCIDENT NEUTRONS/250 USEC GATE     .41492E-01
RANDOM COINCIDENT NEUTRONS/70 USEC GATE     .27150E-02

250 USEC GATE LIVE TIME          308.34 SEC
70 USEC GATE LIVE TIME        318.57 SEC
NET COINCIDENT NEUTRONS/250 USEC GATE    .15795    +/-    .21613E-02
NET COINCIDENT NEUTRONS/70 USEC GATE    .40945E-01+/-    .19316E-02

SYSTEM TOTALS RATE      161.62     +/-    .72798
SHIELDED TOTALS RATE    36.974     +/-    .35246

NET COINCIDENT 250 USEC GATE NEUTRONS/LIVE TIME    22.391     +/-     .30889
NET COINCIDENT 70 USEC GATE NEUTRONS/LIVE TIME    1.4857     +/-     .71381E-01


REDUCED VARIANCE
Y =   .58536E-01     q=    .15306E-03
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Fig. 2.

Passive system printout of the count rates from the 12 detector packages and the 2 flux monitors. The printout also shows primary coincidence count quantities.

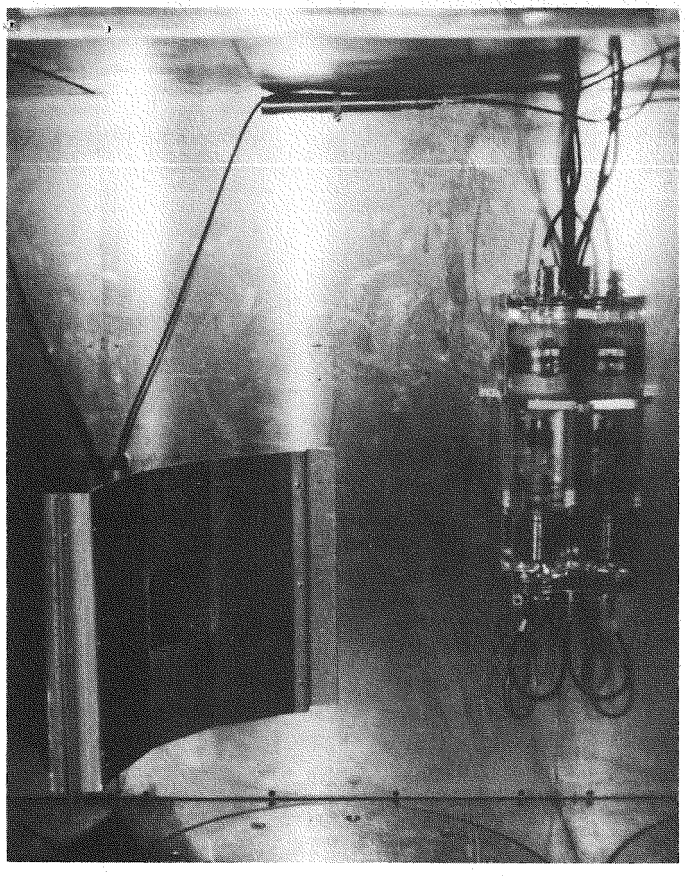

Fig. 3.
The interior of the assay chamber. Two flux monitors--one at the top of the chamber, the other on the lower left--and a neutron generator at the right are positioned above the heavy-duty turntable.

The disposition of graphite (11 cm thick) and polyethylene (25 cm thick) in the assay chamber walls [7-9] is similar to that used in the original Oak Ridge prototype and discussed in the indicated references. In the second-generation units, a 2.5-cm-thick borated polyethylene layer is used to separate the moderation and shielding regions, as indicated in Fig. 1.

IV. MATRIX EFFECTS AND MATRIX CORRECTIONS

A. General and Historical Discussion

For the active neutron measurements, there are two separable types of matrix effects: absorption and moderation.[11] The absorption effects occur almost entirely as an attenuation of the interrogating thermal neutrons, caused by the presence of various neutron poisons within the waste matrix. Moderation effects occur at two stages of the measurement. The original burst of 14-MeV neutrons can be moderated to a considerable extent during passage through the waste matrix. Generally this results in a larger thermal neutron interrogation flux than would have been produced in the absence of matrix.

After the interrogation flux has produced fission reactions within the waste matrix, the same moderating materials can attenuate the prompt fission-signal neutrons resulting in a decrease in observed response relative to the no-matrix case. This attenuation of fission-signal neutrons also is the primary matrix effect for the passive measurement.

Our approach to matrix corrections has been to base corrections on measured quantities determined as adjuncts to the primary active and passive TRU assay measurements.[11] In effect, this approach amounts to an assay of the waste matrix itself, at least in regard to its neutronics properties. Our first effort using this approach, which we implemented at Oak Ridge in 1982, was based on a measurement of the interrogating flux-time history (see Appendix A). We had long since observed a strong dependence of both flux intensity and lifetime on matrix type. Figure 4 shows a set of these time histories that illustrate how strongly different matrices affect this measurement. Note that these time histories were measured with the flux monitor located at the top of the assay chamber. In this location, it senses all thermal neutrons produced during an interrogation, both those associated with the cavity walls and the waste matrix.

The analytic procedure we developed to use this measurement for matrix corrections consisted of fitting a simple exponential function to the dieaway data of the type shown in Fig. 4. We found that an excellent fit (judged by the χ^2 values) was obtained for all types of matrices if this fit was always performed over the region 1.5 to 3.0 ms following the initial 14-MeV pulse. This fit generated two parameters: A_0, which is an extrapolated time zero amplitude, and $T_{1/2}$, the thermal neutron lifetime.

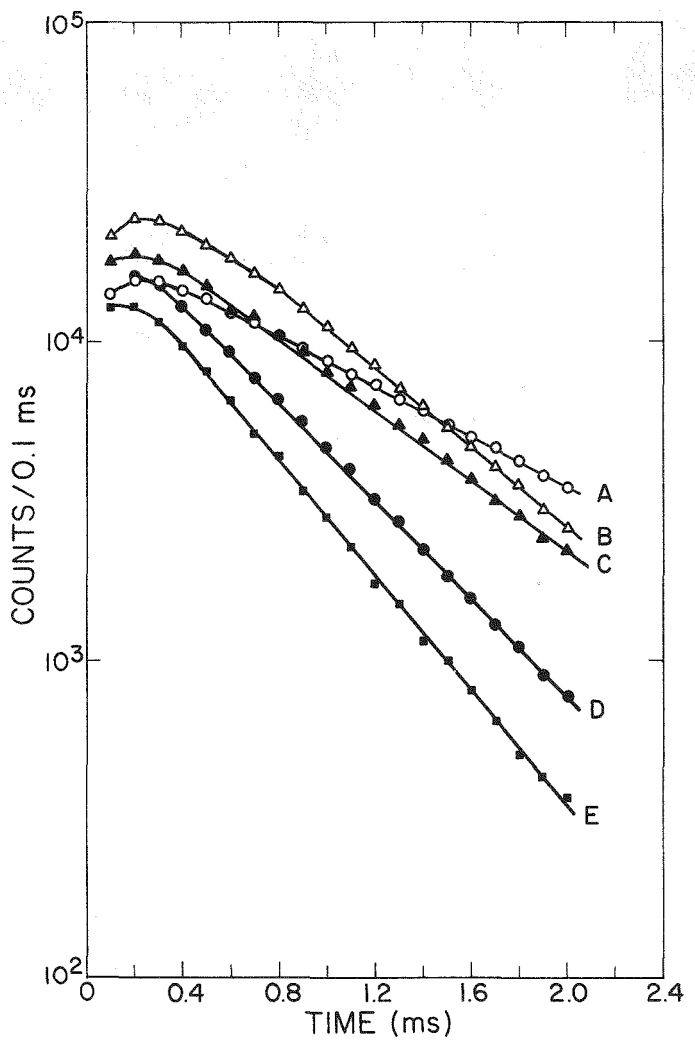

Fig. 4.
Plots of five time-history curves for matrix calibration drums measured with the flux monitor. Note the wide range of amplitudes and dieaway times for the different waste matrices.

Elementary neutron diffusion theory[12] can be used to relate $T_{1/2}$ to the thermal neutron absorption properties of the average system (cavity plus waste matrix). This relationship is

$$T_{1/2} = \frac{1}{v\Sigma_a} \quad , \tag{1}$$

where v is the thermal neutron velocity, measured to be 220,000 cm/s, and Σ_a is the system total macroscopic absorption cross section measured at thermal energies. That is, the measured $T_{1/2}$ value is inversely related to the total absorption properties of the waste matrix.

The A_0 parameter we interpreted as an extrapolated interrogation flux at time zero. Because the matrix moderator content is the primary system variable affecting this parameter, we used it to estimate a moderator correction to the assay data. Our final year-end (FY 83) report on the Oak Ridge system contains detailed information on the calibrations done to quantify this method as well as examples of its application to actual Oak Ridge waste (see Appendix A). More recently in its FY 84 end-of-year report, Oak Ridge published an even larger volume of assayed drum data using this matrix correction approach and our original calibration values.[8]

We performed a considerable amount of mockup matrix studies following our original implementation of the algorithm described in Ref. 7--as the initial portion of our second-generation assay unit research and development effort. We concluded that the A_0, $T_{1/2}$ approach to matrix corrections would be very difficult to apply to the much denser matrices with higher hydrogen content that we knew (see Refs. 13 and 14 on characterization of typical Rocky Flats sludges) would be typical of many of the major defense site wastes. In substance, the $T_{1/2}$ value, once a certain level of neutron poison is reached in the waste drum, tends to "bottom out," thereby leading to an underestimation of the required matrix correction. Analogously, the A_0 value appears to be double valued as a function of matrix hydrogen density. That is, above a certain hydrogen density, the A_0 values measured actually decrease as a function of increasing hydrogen density. As a matter of practicality, neither of these effects produced any significant difficulties for the assay of Oak Ridge wastes because the range of absorber and moderator values at Oak Ridge generally was well within the applicable region for this algorithm. Clearly, however, a more generally applicable approach was called for in the second-generation system.

B. The Second-Generation Approach to Absorption Corrections

As in the foregoing discussion, the matrix correction approach we implemented at Oak Ridge did not appear adequate for many of the waste matrices in the DOE inventory. Therefore, we took an entirely different route for our second-generation systems. Our detailed studies of the A_0, $T_{1/2}$ approach led us to conclude that we needed to monitor the flux exiting the matrix within a drum more directly than was possible using only the bare cavity flux monitor. After consultations with several Los Alamos experts in neutron

physics and a few measurements with cadmium-collimated detectors, it became apparent that a barrel flux monitor was indeed feasible. The original flux monitor (or cavity flux monitor) was retained as well for normalization purposes.

Several versions of barrel flux monitors were tried; the ultimate unit selected for our second-generation implementation program appears in Fig. 5. Although the details of the interactions between the interrogation neutrons and the waste matrix are extremely complicated and can only be studied analytically using large time- and energy-dependent Monte Carlo neutron transport codes, a great simplification results when extraneous neutrons (that part of the thermal neutron flux that has not interacted strongly with the waste matrix) are excluded. The cadmium-shielded collimator achieves this decoupling by allowing only those thermal neutrons that exit perpendicularly from the drum's surface to be sensed by the barrel flux monitor.

This "normal" flux consists of neutrons that have undergone strong drum matrix interactions and thereby reflect strongly the neutronic properties of the matrix. That this monitor does not bottom out as a function of matrix absorber is shown in Fig. 6. This plot shows the ratio of the flux monitor response to the barrel flux monitor response as a function of boron loading in a combustibles mockup matrix. (The ratio of the flux monitor response to the barrel flux monitor response is our second-generation absorption index.) As

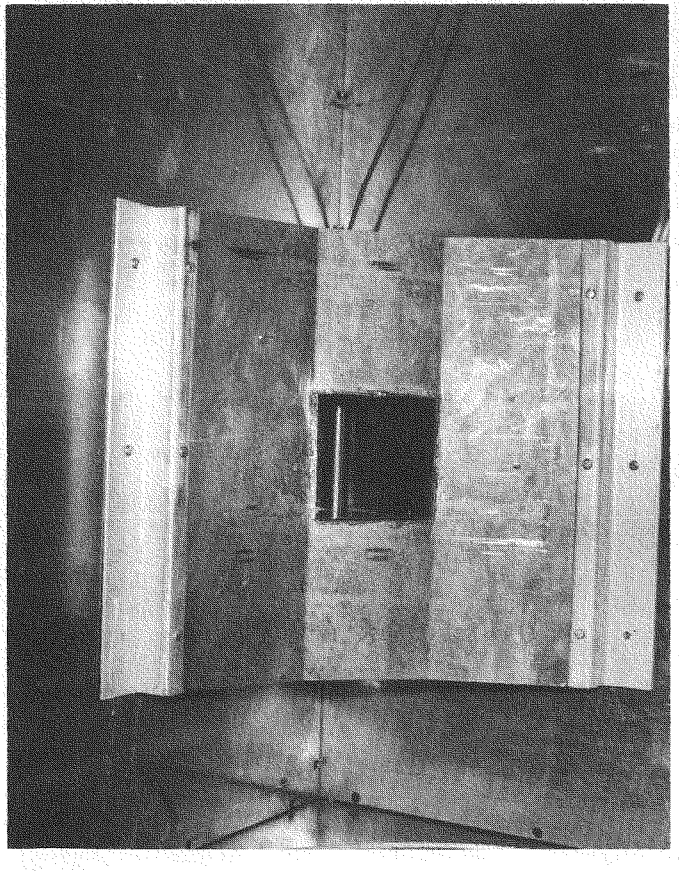

Fig. 5.
The barrel flux monitor is positioned behind a collimated cadmium shield that excludes extraneous thermal neutrons.

with the $T_{1/2}$ parameter, one expects an inverse relationship between the barrel flux monitor and the total neutron macroscopic absorption cross section. This can readily be understood because strong neutron interactions with increasing amounts of neutron poisons will clearly lead to a decrease in the exiting normal flux. Unfortunately, it is not possible to deduce from first principles the exact analytic dependence, and hence the experimental relationships must be used.

As can be seen in Fig. 6, the change in the absorption index, where

$$\text{absorption index} = \frac{\text{flux monitor response (0.7 - 4.7 ms)}}{\text{barrel flux monitor response (0.7 - 4.7 ms)}} \quad , \qquad (2)$$

with changes in the amount of neutron poison present in the drum matrix is quite dramatic and does not bottom out for high neutron poison values. For example, from the data shown in Fig. 6, the highest two boron loadings (0.6 kg of borax and 1.8 kg of borax) differ by a factor of 3. The corresponding absorption index values differ by a factor of almost 4. This is in sharp contrast to the inverse $T_{1/2}$ values for these two boron loadings, which differ only by 10%. It is thus clear that this approach will have the required sensitivity and dynamic range to provide absorption corrections for matrix neutrons in the second-generation systems.

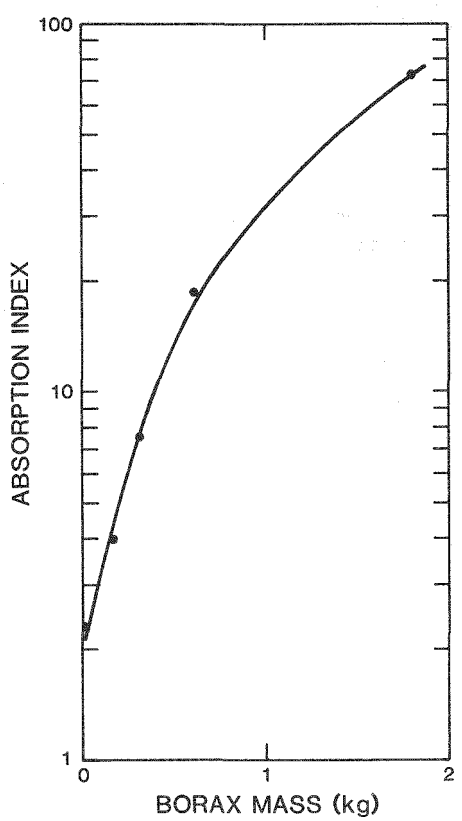

Fig. 6.
Plot of the ratio of count rates for the flux monitor to barrel flux monitor as a function of borax loading in a peat moss matrix.

C. The Second-Generation Approach to Neutron Moderator Corrections

In effect, the Oak Ridge assay algorithm makes no provision for moderator matrix corrections to passive neutrons. This presented no great difficulties for most of the Oak Ridge assays because only a small portion of the Oak Ridge waste had any significant amounts of moderator. We then used a calibration based on what we judged to be the average Oak Ridge matrix and made no attempt at an explicit moderator correction. This approach is entirely justified on the grounds that our neutron detection system design is tailored for independence of moderator content, as are all comparable 4π neutron detection systems currently in use for other applications. In fact, the common practice is to design detectors to be "under moderated," which means that, for modest amounts of moderator present in a waste matrix, the observed response actually is greater than for the empty-drum case. The observed response ultimately decreases, however, when very large amounts of moderator are present.

It became clear that this simplified approach would not be adequate for the second-generation systems. Our study of typical Rocky Flats waste revealed that sludges containing as much as 70% bound water content were common. We thus investigated the means with which appropriate corrections for high hydrogen density could be made to our routine assay measurements. The magnitude of this task was considerable: no established routine moderator correction algorithms or techniques exist for neutron detection systems such as ours.

We looked at three independent means for determining matrix moderator content, using only apparatus that could be implemented within the assay chamber using either the passive or active neutron measurements. Two of these involved the active measurement and depended on (a) the asymmetric interrogation flux and (b) the asymmetric fission signal rates observed between the portions of a matrix drum that are relatively close to the neutron generator, as compared to relatively far from it. Both of these front-to-rear asymmetries are fairly strong functions of matrix moderator density and are essentially independent of absorber density. However, both techniques proved difficult to implement for the routine production assay environment required for the second-generation installations.

These conclusions were reached following a one-week field trial of both techniques at SWEPP in December 1984, during which measurements were made of some 30 waste drums. At some later time it may be possible to apply one or both of these techniques with improved hardware. However, for the present, we selected a more readily implementable technique to develop.

D. Neutron Spectral Measurement and the Moderator Index

The third technique, which depends on the passive measurement, consists of making a crude neutron energy spectral measurement of the passive signal. The "spectrometer" is the ratio of the shielded totals detector count rate to that of the bare plus shielded detectors. These two detector systems have different responses as a function of neutron energy, differing especially in their responses to moderated neutrons. The shielded detectors are encased in cadmium

and thus show no response at all to the thermalized portion of any spectrum whereas the bare detectors respond strongly. Thus, the ratio of shielded-to-bare (or shielded-to-totals) count rates will be a function of the fraction of any given spectrum that is thermalized. In turn, the thermalized fraction depends very strongly on the moderator density of the matrix.

This type of neutron spectrometer has been used many times in various nuclear physics applications. A variation of it has been in use at Lawrence Livermore National Laboratory[15-17] for over 20 years with a particle-accelerator-based, large 4π neutron detection system to determine average spectral energies for various photonuclear reactions. One of the authors of this report (JTC) used this as part of his thesis work on photonuclear reactions.[18] Another variation of the method has been used on occasion at Los Alamos to determine such things as the average thickness of high-explosive regions in large assemblies.[19] In other words, the methodology is not new although the application is.

Figure 7 shows our experimental data for this approach as a function of moderator content in 208-ℓ drums filled with mockup waste matrices. The x axis is the water density equivalent in the drum matrix, and the y axis is the moderator index we derived from the basic measurement using the passive neutron spectrometer discussed previously. In essence, the ratio of shielded totals count rates to system totals count rates has been normalized so that a value of zero is obtained when no moderator is present. In addition, a small correction has been made to account for absorption effects. If neutron absorbers as well as moderators are present in the waste matrix, then a portion of the neutrons thermalized by the moderators will be absorbed before exiting the matrix. This effect must be accounted for to obtain a reliable measurement of thermalized fractions. We accomplish this correction by using the independent matrix absorption index determined during the active assay (see Sec. IV-B and Eq. 2). The magnitude of the correction is obtained empirically by measuring mockup matrix drums that have a constant moderator content but have varying amounts of absorber. Some of the data shown in Fig. 7 were obtained with mockup matrices that have the same moderator content but widely differing absorber content. How well the systematics account for the absorber effects may be judged by how well these data follow a single response curve as a function of moderator content. The actual moderator index we used is as follows:

$$\text{moderator index} = \left[1 - \left(\frac{\text{shielded totals}}{\text{system totals}}\right) / A_0\right] \cdot \left[A_1 + A_2 \cdot \ln(\text{absorption index})\right], \qquad (3)$$

where shielded totals and system totals are, respectively, the net shielded totals and system totals count rates obtained during the passive neutron portion of the measurement; A_0 is the normalizing constant determined from passive neutron calibration data; and A_1, A_2 are parameters determined from combined passive and active calibration data. The term within the first set of brackets is the basic raw spectral data and the term within the second set of brackets is the correction term for matrix absorption effects.

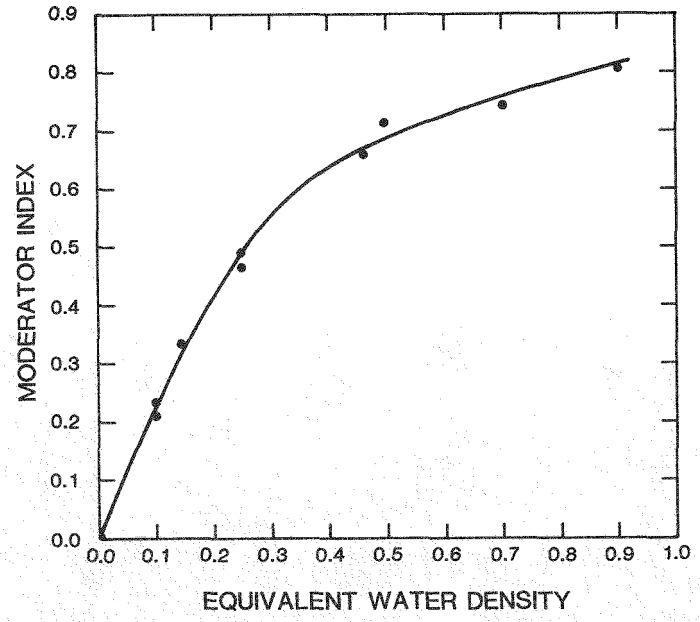

Fig. 7.
This plot of moderator index as a function of water density illustrates the sensitivity of the index to large moderator masses in waste matrices.

As can be seen in Fig. 7, the moderator index we have determined is indeed a sensitive function of moderator content over the entire region from "empty drum" to "barrel full of water." (The largest moderator index was that measured with a barrel of water.) One of the most attractive features of this moderator index is that it is based on a measurement of the actual passive-neutron signal being used for the assay. Thus, spatial distribution effects tend to be automatically taken into account. As we show in Sec. VI-B, moderator effects are generally more important for the passive assay than for the active. Operationally it is thus appropriate to use a moderator correction based on passive neutrons. It is important to always bear in mind, however, that the passive and active assays are co-analyzed. When required (as shown in Sec. VI-A), a moderator correction is also made to the active assay results.

V. DEVELOPMENTAL STUDIES AND SYSTEMATIC MATRIX MEASUREMENTS

A. Background and Approach

The first step in our developmental work was to obtain a comprehensive set of matrix response data from a set of mockup matrices having known absorbing and moderating properties. These data were obtained with our original developmental prototype at Los Alamos. An early report on this work appears as Ref. 11.

To facilitate a systematic understanding of matrix effects as a function of position within a matrix drum, we devised a method of obtaining quantitative passive and active responses that could be associated with a fixed coordinate system within the matrix material. Our r, θ, z coordinate system is referenced to a standard cylindrical 208-ℓ drum in Fig. 8.

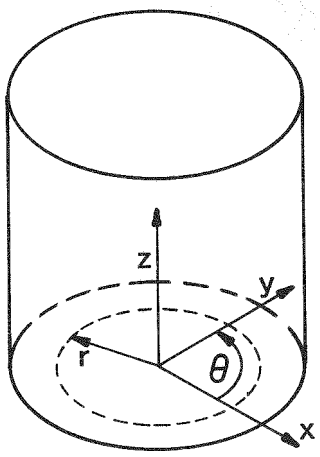

Fig. 8.
The r, θ, z coordinate system within a 208-ℓ drum.

Because one of the standard features of all Los Alamos neutron assay systems is drum rotation within the assay chamber during both passive and active measurements, the θ variation is in essence automatically averaged over angle during each measurement. The only requirement is that either an integral number of rotations occur during the measurement or that the total number of rotations be so large that any effect of a partial rotation is negligible. To ensure that these conditions will apply, Los Alamos standards require the drum rotation unit to revolve once every 10.0 s. The MA-165C second-generation pulsed neutron source is also set up to provide a standard 1000 interrogating pulses per 20.00 s. Thus, if interrogations are always performed in increments of 1000, an integral number of drum rotations will occur during an active interrogation. With the usual standard 2000-pulse interrogation, a total of four rotations occurs.

The usual passive assay standard run time is 400 s, which results in 40 total revolutions. This also is such a large total number that any partial rotations will generally not affect the averaging process.

Usually, three or four hollow vertical tubes are positioned in a 208-ℓ drum to obtain (r,z) matrix responses (Fig. 9). The central three tubes are normally placed at drum radius values of 0, 10, and 20 cm, respectively. They are secured and maintained in a vertical orientation by two thin perpendicular stabilizing aluminum bars at z values of about 25 and 65 cm. These stabilizing bars have holes punched in them to correspond to r values of 0, 10, and 20 cm, and the nominally 2.5-cm-diam aluminum tubes are fed through the holes in the secured stabilizing bars. For some matrices, namely those having extremely large moderator quantities, we also add a fourth vertical tube, usually secured to the drum wall. This produces response data for a fourth r value of about 26 cm. Note that in most 208-ℓ drums the radius value for the drum wall is about 28.6 cm. For drums with thick polyethylene inserts, the fit appears to be sufficiently tight so as not to affect the use of the 28.6-cm outermost radius value. The walls of the polyethylene liner may be regarded as part of the overall matrix. The liner does narrow near the drum top and may not always extend to the drum bottom. These perturbations can be regarded as second-order effects.

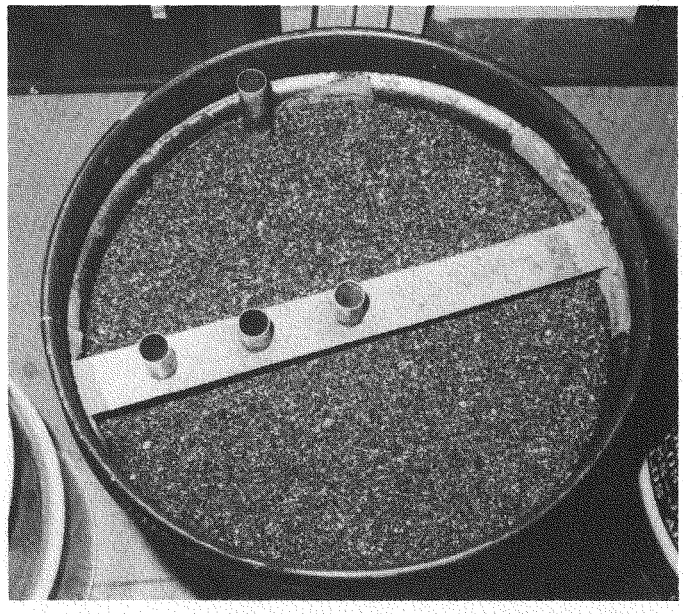

Fig. 9.
Hollow source tubes within a matrix drum allow source measurements that provide (r,z) matrix responses.

Our standard measurement set consists of data taken at 10-cm z increments over the entire z range of the matrix within the drum. Many of the matrix drums used were not completely filled to the top; measurements for these matrices were carried out only to a z value consistent with the actual fill height. Measurements over the range of z were carried out for all (three or four) r values.

A typical set of such matrix response data for one active measurement and two passive measurement sets appears in Table I. These are data for our combustibles mockup, a drum filled with 34 kg of peat moss. One can easily observe the systematic behavior of both passive and active quantities as a function of r and z. For our early developmental studies, we used data of this type for about 15 different mockup matrices ranging from a drum filled with vermiculite to drums filled with Rashig rings and very wet rags. These early studies led to the development of an extremely effective sensor for absorbing materials (see Sec. IV-B and Fig. 5). These early studies also revealed that systematic effects are due only to gross neutron absorption and moderator amounts and are independent of the actual nature of the materials themselves. That is, a drum filled with Rashig rings produces the same responses as a drum filled with vermiculite mixed with an equally absorbing amount of borax. Or, a drum filled with peat moss produces the same effects as one filled with rags, Kimwipes, or waste paper. This observation makes possible relatively simplified systematic studies because one can use mixtures of such materials as peat moss, vermiculite, borax, and water to obtain the complete range of responses to be found in real wastes.

TABLE I

MATRIX RESPONSE DATA FOR A COMBUSTIBLES MOCKUP

z(cm)	4 = 0 cm	4 = 10 cm	r = 20 cm	Volume average
Active: Shielded Totals/Flux Monitor for a 2.0-g Plutonium-Equivalent Source				
0	0.3979	0.3744	0.3285	0.3315
10	0.5994	0.5505	0.4710	0.4777
20	0.7032	0.6533	0.5347	0.5399
30	0.7355	0.6876	0.5674	0.5719
40	0.7034	0.6489	0.5412	0.5482
50	0.6154	0.5570	0.4861	0.4902
60	0.4943	0.4739	0.4176	0.4191
Passive: System Totals Detection Efficiency Measured with a ^{252}Cf Source				
0	0.1149	0.1161	0.1162	0.1162
10	0.1179	0.1161	0.1193	0.1195
20	0.1213	0.1208	0.1238	0.1240
30	0.1228	0.1238	0.1244	0.1243
40	0.1255	0.1256	0.1274	0.1274
50	0.1236	0.1235	0.1259	0.1260
60	0.1229	0.1229	0.1236	0.1237
Passive: 250-μs Gated System Totals Coincident, ^{252}Cf Source Efficiency Per Fission Event				
0	0.0440	0.0453	0.0453	0.0453
10	0.0453	0.0475	0.0473	0.0475
20	0.0478	0.0475	0.0511	0.0512
30	0.0486	0.0486	0.0528	0.0529
40	0.0503	0.0514	0.0539	0.0538
50	0.0509	0.0503	0.0539	0.0540
60	0.0503	0.0517	0.0525	0.0524

An additional advantage is that these simple materials can readily be used to produce uniform mockup matrices, which generally lead to smoothly varying response curves. Of course, real wastes will have nonuniform and heterogeneous material distributions--this is not disputed. However, the simple fact is that nonuniform and heterogeneous matrix materials do not add to one's understanding of systematic behavior; they only confuse the issue by producing random anomalies. The study of the effects of nonuniformities and heterogeneities can only proceed after one understands the systematic behavior patterns from uniform and homogeneous matrices.

B. Examples of Matrix Response Data

As can be seen in Table I, the various passive and active assay matrix response values do vary in a smooth fashion as a function of r and z. The fifth column shows the volume-weighted average value for each vertical slice. The volume-weighted average is defined as the average value found by dividing the given vertical slice up into several equal volume elements, determining the matrix response for each element, and averaging these values.

For instance, if one divides a vertical slice of total radius 28.6 cm and thickness 10 cm into five equal volume elements, one obtains a set of annular cylindrical solids all of which are 10 cm thick. The innermost volume element would be a cylindrical solid of outer radius 12.8 cm, the next a cylindrical annulus with an inside radius of 12.8 cm and outside radius of 18.1 cm, the next a cylindrical annulus with an inside radius of 18.1 cm and outside radius of 22.1 cm, the next with an inside radius of 22.1 cm and outside radius of 25.6 cm, and the last with an inside radius of 25.6 cm and outside radius of 28.6 cm.

C. Analytic Fits to Matrix Response Data and Volume-Weighted Averages

If one can fit a simple analytic function to the measured data, the volume-weighted average response can be calculated analytically. We have found that most of the observed distributions can be fitted to a power law

$$y = A + Br^N, \tag{4}$$

where A, B, and N are the fit parameters and r is the drum radius (see Figs. 8 and 9).

One other observation can be made: the volume-weighted average quantity is usually nearly equal to the measured quantity at r = 20 cm. This happens because 20 cm is very nearly the volume-weighted mean value of the radius.

The significance of volume-weighted average values is that they represent the most probable measurement result for either a totally uniform or a totally random distribution of source material within the matrix. Thus, in determining calibrations, they are the appropriate weighted value to use. Because each of the volume slices has the same thickness (10 cm), one determines an overall most-probable response value by merely averaging linearly over the volume-weighted averages for an individual slice. For example, in Table I the overall most-probable response value for the active data is 0.4826.

D. Systematic Distribution Studies and Average Assay Errors

An additional advantage accrues to determining an analytic fit to each of the vertical slices: one has a complete "analytic roadmap" of the matrix responses within that drum. If one wishes to divide the entire drum into 25 or so equal volume elements, it is a very simple exercise to calculate the matrix response values for corresponding individual elements. With this set of 25 or so response values, one may play the "distribution" game and calculate mean standard deviations for various assumptions about source material distributions. In principle, these distribution mean standard deviations can be related to expected assay error distributions for large numbers of assays.

For instance, if one assumes that the source material is uniformly distributed throughout the drum volume (in the real world, sludge wastes might approximate this situation), then a calculation of the mean standard deviation leads to a value of zero. This happens because the measured value for this assumption is exactly the calculated volume-weighted average.

If, at the other extreme of assumptions, one assumes that the total source material is confined to a single volume element in each drum, but this element is distributed randomly in a large collection of drums, then one can show from the data in Table I that a mean standard deviation of 21% is obtained.

For distributions between these assumptions, it is reasonable to expect intermediate values of mean standard deviations. In all likelihood, the real-world wastes will group themselves somewhere between these two extreme distributions. The important point is that a complete analytic description of the matrix response provides an extremely powerful means with which to calculate and study the expected error distribution patterns for large quantities of drum assay measurements. Furthermore, this information is readily obtained for any absorber and moderator combination, which is to say, for any waste matrix.

VI. ANALYTIC FITS TO SYSTEMATIC MEASUREMENTS OF MATRIX CORRECTION FACTORS

A. Active Neutron Measurement

1. General Discussion. As discussed in Sec. IV, the active neutron measurement is affected by both matrix absorbers and moderators. These effects are separable in the systematic Los Alamos approach. The first step in this approach is to determine the volume-weighted average response for a standard

fissile source in an empty drum. Data of the type shown in Table I (first data block) are used. We have used the same fissile standard for all active matrix response data, a 2-g plutonium-equivalent sample 2.5 cm in length and 2.5 cm in external diameter.

Plutonium equivalence is determined by comparison with neutronically thin plutonium samples in an open geometry. For the actual response measurements, we used a uranium sample to obtain relative responses between various matrices and the empty drum. (Absolute fissile standard measurements are discussed in Sec. VII.)

We define the total active matrix correction factor (MCFA) as follows:

$$\text{MCFA} = \frac{\text{Volume-weighted average response measured in an empty drum}}{\text{Volume-weighted average response measured in the matrix drum}} . \quad (5)$$

With this definition, the MCFA values will generally be numbers greater than 1. That is, generally, the effect of the matrix will require a correction factor greater than 1. An MCFA = 1.00 indicates that no correction is necessary.

Using the ideas developed in Secs. IV and V, we have determined that the MCFA is a function of the two indices, one for absorption and one for moderator, which have been defined in Eqs. 2 and 3. The overall MCFA is then separated into two independent matrix correction factors:

$$\text{MCFA} = \text{MCFA (absorption index)} \cdot \text{MCFA (moderator index)} . \quad (6)$$

Based on the experience we gained in 7 years of work on this assay technique, we know that there is likely to be a threshold of moderator content below which no moderator correction is required. To first order, an addition of moderator will result in an increased interrogation flux within the drum, which over a limited region compensates for the accompanying loss of shielded detector system efficiency.

Eventually a large enough moderator amount produces such a large decrease in the shielded detector response that the interrogation flux increase does not fully compensate for it and the overall fissile response drops off. However, it is possible to identify a large number of matrices for which the compensating condition is valid for moderator content. Identifying the threshold for the effect requires a simple iterative approach.

Table II shows our current experimental MCFA (volume-weighted average) data base, which consists of complete measurements with 19 mockup matrices ranging from vermiculite to pure water. The measured absorption index and moderator index values are tallied for each matrix as are the MCFA values. These MCFA values were measured in the three finished second-generation units: Rockwell-Hanford, Idaho (SWEPP), and Savannah River. These units were designed to be identical neutronically; a study of Table II reveals how well this has been accomplished. Over one-half of the mockup matrices have been measured in at least two of the three units, and four matrices have been measured with all three units. All intercomparison values agree to within $\pm$ 5%.

TABLE II

SUMMARY OF ACTIVE CALIBRATION DATA FOR
SECOND-GENERATION ASSAY SYSTEMS

Matrix	Absorption Index	Moderator Index	Volume Average Matrix Correction Factor		
			Hanford	SWEPP	Savannah River
Vermiculite	1.52	0.000	1.05	1.08	1.08
35 kg peat moss	2.32	0.234	0.95	0.96	---
34 kg peat moss + 8 kg water	2.50	0.336	0.97	---	0.93
205 kg sand + vermiculite	3.05	0.000	1.23	---	---
34 kg peat moss + 0.16 kg borax	3.96	0.231	1.28	---	---
50 kg water + vermiculite	3.88	0.491	1.46	---	1.39
200 kg alumina	4.35	0.139	1.61	1.54	---
200 kg iron	5.58	0.000	1.64	1.68	---
323 kg iron	10.4	0.000	---	---	1.93
34 kg peat moss + 0.3 kg borax	7.55	0.214	2.05	---	---
Vermiculite + 1.8 kg borax	10.9	0.000	2.11	2.13	---
Vermiculite + 20 kg Pyrex glass	12.9	0.000	2.24	2.41	2.31
Vermiculite + 50 kg water + 0.3 kg borax	9.17	0.466	---	---	2.80
34 kg peak moss + 0.6 kg borax	18.8	0.207	3.31	3.48	3.19
92 kg water + vermiculite	8.62	0.654	3.46	---	---
34 kg peat moss + 77 kg water	12.1	0.708	3.90	---	---
PREPP concrete + iron	34.9	0.568	6.85	7.04	---
34 kg peat moss + 1.8 kg borax	70.7	0.214	7.64	8.27	8.08
200 kg water	31.4	0.801	---	---	9.62

2. Absorption Correction Factor. The first step in the iterative procedure is to plot the 19 MCFA values as a function of their absorption index values (Fig. 10). For reasons that will be apparent, we have used a natural logarithm scale for both x and z axes. As can be seen, these data fall into three categories: (a) three matrices having absorption index values less than 3.0 that are consistent with unity MCFA to ± 8%, (b) ten matrices that are consistent with a linear relationship between MCFA and absorption index plotted on a log-log basis, and (c) six matrices that clearly depart from the linear relationship.

Comparison of the latter six matrices with Table II reveals that all six have moderator index values in excess of 0.4. The same comparison shows that all 10 of the "linear" matrices have moderator index values less than 0.4. We interpret this result to mean that the threshold moderator index value is in the neighborhood of 0.4, and thus only the 10 matrices having a moderator index value less than 0.4 will be used to determine the linear fit region parameters.

The next step is to fit the 10 matrices with the function

$$\text{MCFA (absorption index)} = A \cdot (\text{absorption index})^{N} \quad . \tag{7}$$

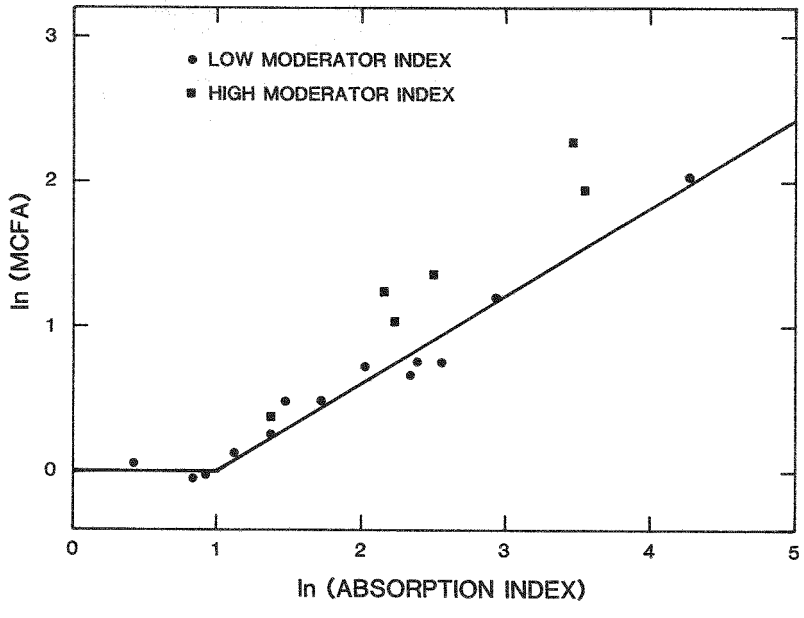

Fig. 10.
Plot of total active matrix correction factor as a function of the absorption index. Points that fall above the systematic straight line (indicated by squares) require a moderator correction.

Applying a standard, unweighted least-squares fitting procedure results in values A = 0.540 and N = 0.612. In order to avoid systematic bias in the fit, only one value of MCFA was used for each matrix. Using the analytic fit to predict the actual MCFA values for these 10 matrices results in a mean standard deviation of about ± 10%.

The least squares fit is shown as a straight line in Fig. 10. This straight line intercepts an MCFA value of 1.00 (ln MCFA = 0.00) at an absorption index = 2.72. In other words, there is a threshold value of the absorption index below which MCFA = 1.00, and this threshold value occurs for the absorption index = 2.72.

Putting all this together, we can now state that

$$\text{MCFA (absorption index)} = 1.00, \tag{8}$$

for the absorption index $\leq$ 2.72, and

$$\text{MCFA (absorption index)} = 0.54\ (\text{absorption index})^{0.612}, \tag{9}$$

for the absorption index > 2.72. This is the analytic representation of the absorption portion of the active assay matrix correction factor.

3. Moderator Correction Factor. The six nonlinear matrices from Table II require the combined absorption and moderator correction indicated in Eq. 6. The MCFA (absorption index) portion is obtained by calculation from Eq. 9 using the appropriate absorption index value for each matrix. As indicated in Eq. 6, the moderator portion, MCFA (moderator index) is obtained by dividing the total measured MCFA values shown in Table II by the calculated MCFA (absorption index) values. These separated MCFA (moderator index) values are shown in Fig. 11a.

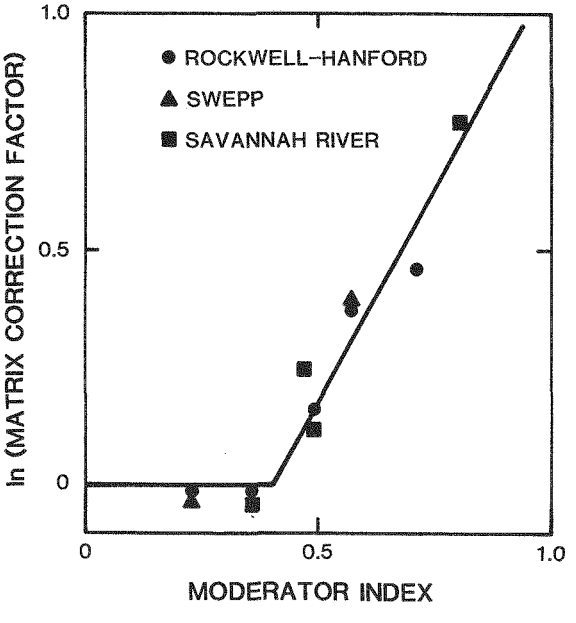

Fig. 11a.
Plot of the separated moderator portion of the total active matrix correction factor for matrices having a moderator index in excess of 0.40.

For simplification of data presentation, the matrix correction factor values in Fig. 11a are plotted on a semi-log scale as a function of the moderator index. As can be seen, there is definitely a threshold as a function of the moderator index, below which no moderator correction is required. Above this threshold the natural log of the correction factor varies linearly with the moderator index. The largest correction factor shown in Fig. 11a was that obtained with a barrel full of water. This point represents the upper extreme in hydrogen density that could be expected to occur in real world wastes.

The analytic representation of these data is thus of the form

$$\text{MCFA(moderator index)} = 1.00, \tag{10}$$

for the moderator index ≤ 0.40,

$$\text{MCFA(moderator index)} = 0.483\ e^{1.817 \cdot \text{moderator index}}, \tag{11}$$

for the moderator index > 0.40.

The combined absorption and moderator corrections for the data base of Table II are shown in Fig.11b, for which the six nonlinear matrices have had moderator corrections made by using Eqs. 10 and 11 and the experimental moderator index values. The overall mean standard deviation for this data set is about $\pm$ 10%. The overall active matrix correction factor for an unknown waste drum is calculated from the measured absorption and moderator indices using Eq. 6, with separate absorption and moderator portions calculated from the set of Eqs. 8-11.

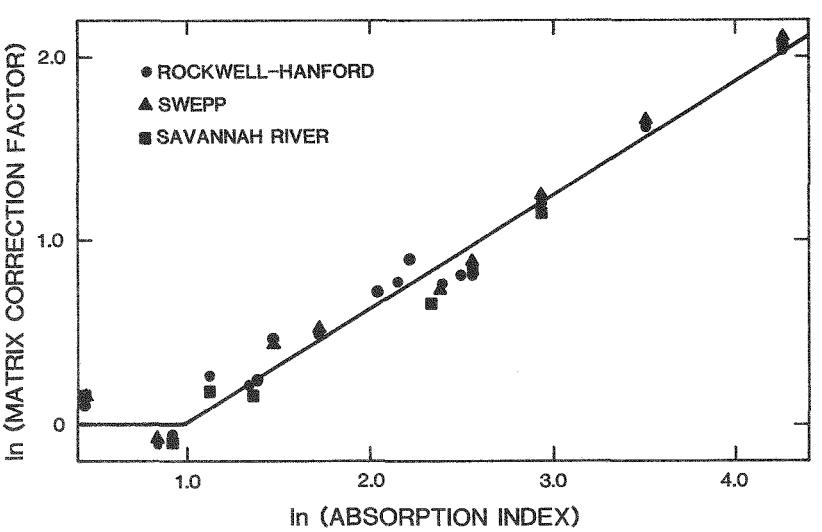

Fig. 11b.
Plot of the entire data set of active matrix correction factors with the high hydrogen density matrices corrected using the systematic moderator corrections shown in Fig. 11a.

B. Passive Neutron Measurement

1. General Discussion. The passive neutron matrix corrections are determined by systematic drum matrix measurements in a manner similar to the active measurements discussed in earlier sections. For these, a standard ^{252}Cf spontaneous fission neutron source is positioned at the various (r,z) locations within the hollow tubes in the matrix drum and a passive response measurement is performed similar to that used during actual drum assay measurements. As the ^{252}Cf neutron sources used at Los Alamos all have been calibrated relative to a single National Bureau of Standards (NBS) calibrated standard, these data are readily analyzed into absolute passive detection efficiencies for a ^{252}Cf fission spectrum source. Typical (r,z) matrix response data of this type are shown in Table I. In analogy with the active measurements, only the responses relative to the empty drum case are required for the matrix correction factor data. Thus, the ^{252}Cf spontaneous fission source is appropriate to use because its relative responses are essentially the same as for plutonium or other TRU spontaneous fission sources. Absolute calibration responses are determined in open geometry using plutonium standards having known isotopic composition; these are discussed in Sec. VII.

Again, in analogy with the active matrix correction data, one defines the matrix correction factors in terms of the response observed with an empty drum in the assay chamber (Eq. 5). The principal difference between passive and active matrix correction factors is that the passive matrix correction factors depend only on the moderator index. The data analysis is thus simplified relative to the active case. It is significant that the same moderator index values are used for both active and passive matrix corrections.

There are currently five separate and independent quantities measured in the passive assay:

(a) system totals,
(b) shielded totals,
(c) system totals 250-μs coincidence,
(d) shielded totals 70-μs coincidence, and
(e) shielded totals 70-μs reduced variance.

The first four of these quantities require independent matrix correction factors whereas the fifth uses the same correction factor as the shielded totals 70-μs coincidence.

Because the procedure for determining volume-weighted averages is the same for passive and active matrix correction factors, we will not discuss it again. Similarly, the analytic fit functions (Eq. 4) used to determine a systematic (r,z) dependence are the same, although the fit parameters differ significantly.

The essence of determining the passive matrix correction factor is dealing with a massive amount of calibration data, preceded by the equally laborious task of acquiring the data. Over a man-year of work has been expended in this effort to date.

2. Analytic Fits to Systematic Volume-Weighted Passive Data. Figures 12 and 13 show the systematic measurements of passive matrix correction factors for the systems totals (Fig. 12) and for the systems totals coincidence (Fig. 13) plotted as a function of the moderator index. The corresponding data for the shielded totals matrix correction factor display a similar behavior. We have determined that the four independent quantities for the passive matrix correction factor can be fitted with the following equations:

$$\text{MCFP(system totals)} = 1.00, \tag{12a}$$

for the moderator index ≤ 0.355,

$$\text{MCFP(system totals)} = -0.16 + 3.28 \cdot \text{moderator index}, \tag{12b}$$

for the moderator index > 0.355,

$$\text{MCFP(shielded totals)} = \frac{1}{1 - \text{moderator index}}\ , \tag{13}$$

$$\text{MCFP(system coincidence)} = \left(\frac{0.5967}{1 - \text{moderator index}} + 0.4187\right)^2\ , \tag{14}$$

$$\text{MCFP(shielded coincidence)} = \left(\frac{0.8092}{1 - \text{moderator index}} + 0.2337\right)^2\ . \tag{15}$$

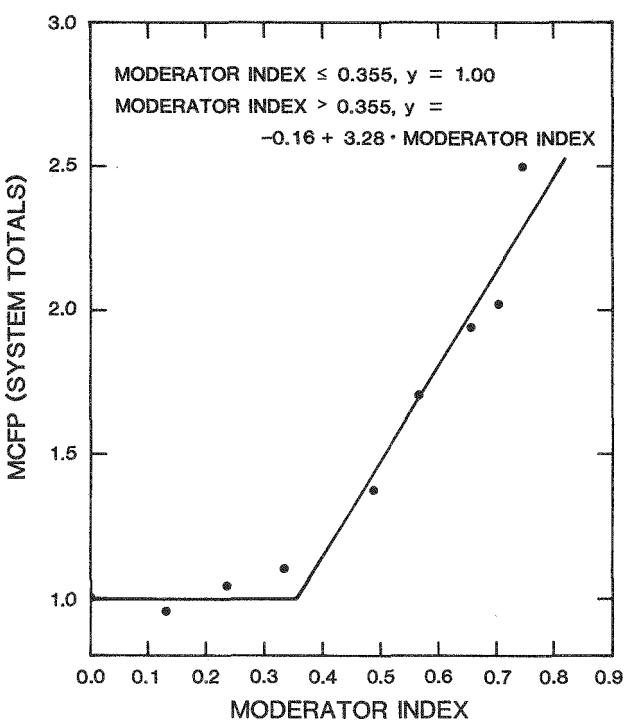

Fig. 12.
Plot of systematic measurements of matrix correction factors for the passive system totals as a function of the moderator index.

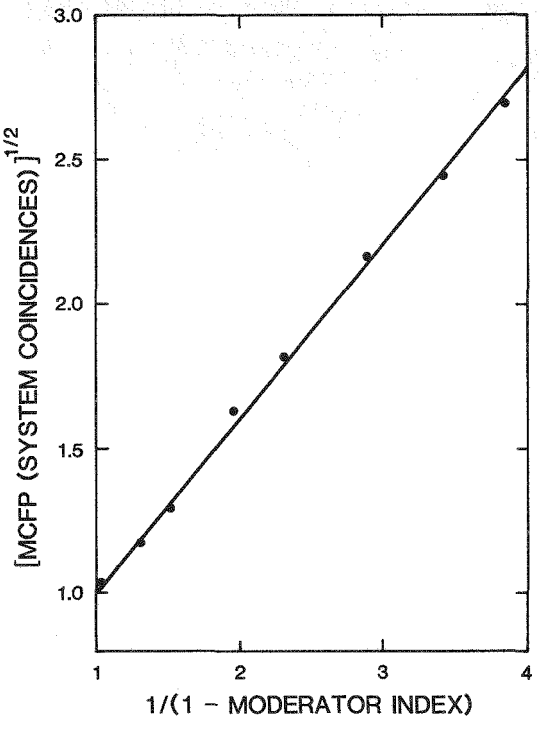

Fig. 13.
Plot of systematic measurements of the matrix correction factors for the passive system totals 250-μs coincidence as a function of the moderator index.

C. Use of Systematic Matrix Correction Factor Data in Other Combined Passive-Active Systems

Note that the same passive matrix correction factors apply to all three Los Alamos second-generation systems. The passive data presented in Figs. 12 and 13 were obtained from a mixture of measurements performed with all three units. The corresponding information was noted previously for the active data (Fig. 11 and Table II).

However, it is unlikely that any system built with different geometric configurations of ^{3}He counters or different graphite and polyethylene layer thicknesses can use directly either the passive or active matrix correction factor quantities discussed above. For instance, systems containing a 15-cm-thick graphite layer (such as that used at the Atomic Energy Research Establishment at Harwell and by a prototype under development at Rocky Flats) instead of the 10-cm layer used in the Los Alamos systems will have considerably different passive and active responses as a function of both matrix absorber and moderator content. We would expect the systematic description we have developed to be valid; however, a detailed matrix response calibration effort similar to the one done at Los Alamos will be required to determine the appropriate fit parameters.

Because the Los Alamos/Oak Ridge prototype unit and the Los Alamos mobile assay drum counter under development both have a different ^{3}He counter layout as well as different dimensions for the assay chamber, separate detailed calibration efforts are required. Los Alamos has already designed, built, and installed at Oak Ridge a barrel flux monitor appropriate for matrix correction data for the absorption portion of the active matrix correction factor. Los

Alamos also outlined for Oak Ridge a program of systematic moderator matrix calibrations required to determine parameters in the combined passive-active matrix correction formalism presented here. When this is done, the Los Alamos/Oak Ridge prototype will also qualify as a second-generation unit and can be used for systematic general assay studies such as those presented in this report.

The Los Alamos mobile nondestructive assay (NDA) unit has also been similarly equipped, and systematic second generation calibrations have been performed. This unit has been used in an extensive field test campaign at the Nevada Test Site.

VII. ABSOLUTE CALIBRATION STANDARDS AND CALIBRATION MEASUREMENTS

A. Calibration Standards

Table III lists some of the many uranium, plutonium, americium, curium, and californium isotopic standards used by Los Alamos for determining the absolute passive and active calibration. These standards have been accumulated over a 20-year period associated with a great variety of projects in nuclear research, safeguards, national defense, and waste assay. Virtually all isotopic mixtures of interest are represented and virtually all have isotopic abundances and absolute mass values that are traceable to accepted national standards laboratories such as the National Bureau of Standards or New Brunswick.

In addition, calibration materials prepared at other laboratories have also been used as standards. These will be discussed in later sections where appropriate.

B. Active Calibration Measurements

Because the active measurement depends on thermal neutron interrogation, some precautions must be taken to assure a quantitative calibration. Principally, one must use calibration materials that do not display self-absorption or for which self-absorption corrections are calculable.

The basic active response to fissile material is given by

$$\text{Net signal} = \sigma_F \cdot \upsilon_p \cdot \epsilon \cdot \Phi \cdot K \ , \tag{16}$$

where σ_F is the isotopically weighted fission cross section for thermal neutrons, υ_p is the isotopically weighted average number of prompt neutrons emitted per fission, ϵ is the neutron detection system efficiency, Φ is the thermal neutron interrogating flux, and K is the self-absorption factor. Ideally one would choose K to be unity; however, most practical calibration sources will require a small self-absorption correction. These factors are readily determined using elementary neutron diffusion theory such as that discussed in Ref. 12.

TABLE III

ISOTOPIC STANDARDS AT PAJARITO SITE

Identification Number	Description	Origin/Calibration	Characteristics
W25243-1	^{252}Cf (and several other ^{252}Cf sources)	Amersham-Searle calibrated relative to an NBS standard	Neutron source strength (6/12/84) 1.62 x 10^5 n/s (± 5%)
CVN-2	^{252}Cf	Amersham-Searle, calibrated relative to an NBS standard	Neutron source strength (6/12/84) 1.57 x 10^5 n/s (± 5%)
15268	^{244}Cm	Oak Ridge Special Isotopes Division provided calibration also	18.6 mCi (230μg) in oxide form, 100% isotope
15267	^{240}Pu	Oak Ridge Special Isotopes Division provided calibration also	117 mCi (1242 mg) in oxide form, > 98.5% isotopic purity
UN2982	^{241}Am	Oak Ridge Special Isotopes Division provided calibration also	10.29 mCi (3.0 μg) in oxide form, 100% isotope
CMB-11	^{239}Pu	Los Alamos CMB Division did preparation and calibration	93.70% - ^{239}Pu, 5.93% - ^{240}Pu, 0.31% - ^{241}Pu, 0.06% - others, Seven separate encapsulated standards (1% mass accuracy) having elemental plutonium masses in mg of 0.11, 1.2, 11.2 51.0, 102.0, 202.0, 1001.0
CMB	^{238}Pu	Los Alamos CMB Division did preparation and calibration	4.4 g ^{238}Pu 80% isotope purity, in oxide form
Zero-Power Plutonium Reactor	^{239}Pu fuel plates	Idaho National Engineering Laboratory/Zero-Power Plutonium Reactor did preparation and calibration	Several individual fuel "coupons" ranging in ^{240}Pu content from 4.5% to 27%
Los Alamos	Depleted uranium	Los Alamos	100.0-g cylinder: 99.8 g - ^{238}U, 0.20% - ^{235}U
Los Alamos	Enriched uranium	Los Alamos	Thin foils: 93.5% - ^{235}U, 6.5% - ^{238}U
Los Alamos	Natural uranium	Los Alamos	51.7-g foil
Los Alamos Group Q-2	Depleted uranium (multiple standards)	Los Alamos CHM Division and Goodyear Atomic Corp. at Portsmouth (did high-precision mass spectral measurements)	Large number of metal pieces cut from same 0.25-in.-thick plate: ^{235}U = 0.2004%, ^{238}U = 99.80%
Los Alamos	^{233}U	Los Alamos	10-g disk

Equation 16 illustrates a very important feature of this active neutron technique: the basic active signal is proportional to the quantity $\sigma_F \cdot \upsilon_p$. For the principal fissile isotopes encountered in TRU wastes (^{239}Pu and ^{235}U), these basic nuclear physics parameters have been measured with great accuracy. This has been done because these same parameters are fundamental to the operation of all nuclear power and isotope production reactors. Practically speaking, this means that uranium and plutonium standards may be used interchangeably because the measured active response can be scaled accurately by the factor $\sigma_F \cdot \upsilon_p$. Using the most recent nuclear data compilation, one determines that the active response per gram of ^{239}Pu is 1.50 times that of ^{235}U per gram.[20] The experimental uncertainty in this ratio is less than 1%.

Figure 14 shows the active response measured with the Los Alamos second-generation assay units for several of the uranium and plutonium standards described in Table III. These measurements were made in one of the combustible mockup matrices to illustrate that the relationship between ^{239}Pu and ^{235}U responses is also independent of matrix.

For these measurements, we determined a ratio between ^{239}Pu and ^{235}U responses of 1.48 ± 0.05, in excellent agreement with the expected value of 1.50. Note also from Fig. 14 that the active response per gram is constant as a function of fissile mass. This, of course, is strictly true only when self-absorption effects are properly taken into account. Note as well the measured response for a 1-mg plutonium standard shown in Fig. 14, indicative of the very high system sensitivity for fissile assay.

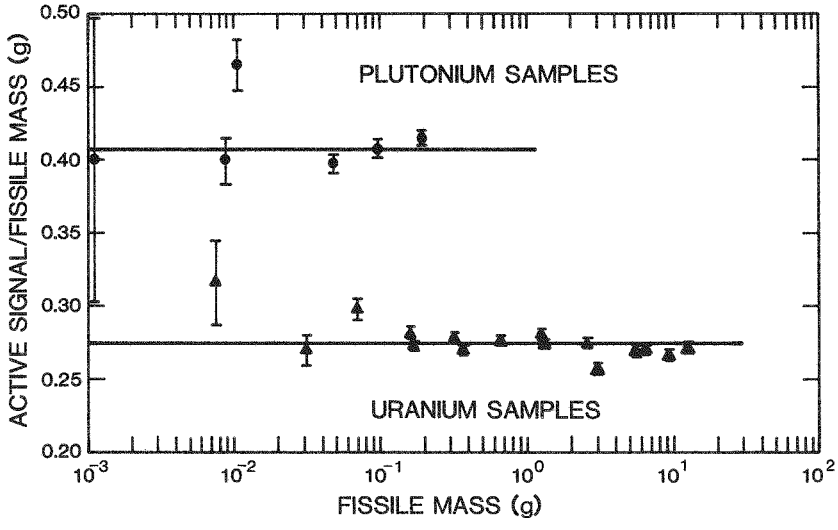

Fig. 14.
Plot of measured active response per gram for a large number of ^{239}Pu and ^{235}U standards.

To provide active system calibration standards for all the second-generation units, we produced a large number of ^{235}U metal standards at Los Alamos, all cut from the same 0.63-cm-thick sheet of depleted uranium metal. These 40 standards vary in length and width (widths between 0.63 and 1.9 cm and lengths between 0.63 and 15 cm) so that a range of ^{235}U mass is provided for studies such as that illustrated in Fig. 14.

This uranium metal sheet was randomly sampled over its entire volume, and the random samples were subjected to independent double-precision mass spectrometry, which was done at the Goodyear Atomic facility in Portsmouth, Ohio, with intercomparison during the mass spectrometry measurements to NBS-traceable uranium standards. This determination established that all portions of the original uranium metal sheet have the same isotopic composition. The ^{235}U isotopic content was measured to be 0.2004 wt% with an estimated uncertainty of 0.0009% (95% confidence level).

The ^{235}U content was also determined independently at Los Alamos in CHM Division by means of a standard mass spectrometer. The result obtained was a ^{235}U content of 0.20 ± 0.002 wt%, in excellent agreement with the more accurate double-precision spectrometer results of Goodyear Atomic.

At least one of these ^{235}U standards will be provided on a permanent basis to each of the second-generation units. These standards are not accountable in a special nuclear materials sense so that they may be kept in nonsecure areas, a considerable advantage over ^{239}Pu standards, which are accountable and must be protected with appropriate safeguards. We recommend that a ^{235}U standard assay measurement be made each day that waste assay measurements are made. The absolute calibration can thus be checked routinely, without having to follow an elaborate, inconvenient, and costly procedure of calling up security guards to bring the standards.

C. Passive Calibration Measurements

The dominant neutron sources in plutonium-contaminated wastes are the even isotopes of plutonium. In typical weapons- and reactor-grade materials (over 98% of all defense wastes by volume), it is the isotope ^{240}Pu that provides the spontaneous fission neutron signal used to quantify plutonium. Given the relatively weak emission rates for usual weapons-grade materials (about 30 spontaneous fissions per second per gram of total plutonium) and the relatively low coincidence neutron detection efficiency (about 2% per fission in the most sensitive mode), fairly large plutonium mass is required for absolute standard measurements.

Because such large plutonium standards must be safeguarded, and thus require elaborate physical security procedures, we have followed the strategy of providing the initial and primary absolute calibration at Los Alamos followed by cross calibrations with ^{252}Cf spontaneous fission sources. These NBS-referenced ^{252}Cf sources are very convenient to use and provide a direct and strong coincident neutron signal. Once the cross calibration is established the ^{252}Cf serves as a relative monitor of the system coincidence efficiency for plutonium neutrons. In addition, because the ^{252}Cf

sources are absolute standards in themselves, the system neutron detection efficiency is determined with each use of the ^{252}Cf source, which serves as a certifiable calibration within itself.

It must also be emphasized that absolute plutonium standards may be used at any time to verify the original absolute calibration. We would, in fact, recommend that this be done periodically. The point is, however, for a routine verification, the ^{252}Cf source is more than adequate. In actual fact, most precise passive neutron measurements carried out these days specify an NBS-certified ^{252}Cf source as the absolute standard.

The passive neutron calibration responses obtained with a set of NBS-referenced cylindrical plutonium standards are shown in Table IV. These plutonium standards are of typical weapons-grade isotopic composition; the actual isotopic composition is included in the table. In contrast to the active measurement, increasingly large plutonium samples undergo a signal enhancement relative to small plutonium samples, a classic sample multiplication effect. The data shown in Table IV have been corrected for this effect, about 4% for the smallest cylinder and 13% for the largest cylinder.

The last column in Table IV shows the measured ratio between a ^{252}Cf standard and the four plutonium cylinders. The quantity tabulated is the ratio of coincidence signals per spontaneous fission. As can be seen, this averages to about 2.27. The reason ^{252}Cf produces a greater coincidence signal per fission than ^{240}Pu is that it emits more neutrons on the average per fission (3.76 compared with 2.15).

D. The Pink Drum Measurements

To provide a complete active and passive certification measurement for each of the second-generation systems, one that can be carried out daily, we have packaged one of the uranium standards discussed above and one of the ^{252}Cf sources discussed in the previous section in a pink drum. These highly conspicuous drums (that have been painted pink so they will not be confused with actual waste drums) have been provided to the three second-generation systems in operable status, one drum to each site. The pink drum is assayed each day as the first data acquisition; the data are recorded on magnetic disk in the same fashion as all waste assay data are recorded. The active portion of the pink drum assay provides the certification of the active system through its known ^{235}U content. The passive portion provides its certification through the measurement of the ^{252}Cf response, which, after accounting for source decay (^{252}Cf has a 2.64-year spontaneous fission half-life), provides both a direct detector efficiency value and a coincidence signal relatable to the original absolute plutonium measurement.

TABLE IV

CALIBRATION MEASUREMENTS OF PASSIVE NEUTRON STANDARDS IN OPEN GEOMETRY

Source Description	Calculated Source Multiplication[a]	System Coincidences[b] (counts/s/g of plutonium)	Shielded Coincidences[b] (counts/s/g of plutonium)	^{252}Cf Coincidence Signal ÷ Plutonium Coincidence Signal
W25193-2 ^{252}Cf neutron source	1.00	0.0639 ± 0.008	0.0039 ± 0.002	---
2.94-g weapons-grade plutonium cylinder	1.039	0.79 ± 0.004	0.050 ± 0.011	2.31 ± 0.12
9.82-g weapons-grade plutonium cylinder	1.059	0.80 ± 0.02	0.052 ± 0.005	2.23 ± 0.06
29.7-g weapons-grade plutonium cylinder	1.085	0.78 ± 0.01	0.052 ± 0.002	2.29 ± 0.04
98.9-g weapons-grade plutonium cylinder	1.127	0.79 ± 0.01	0.054 ± 0.002	2.26 ± 0.04

[a]These calculations were performed by Glenn Brunson of The Advanced Nuclear Technology Group at Los Alamos.

[b]Multiplication-corrected values. These are expressed as counts per second per ^{252}Cf fission and as counts/s/g of plutonium for the plutonium samples. Plutonium isotopics: ^{238}Pu = 0.012%, ^{239}Pu = 93.81%, ^{240}Pu = 5.81%, ^{241}Pu = 0.35%, ^{242}Pu = 0.002%.

We are assembling a working history of these pink drum calibration/certification measurements at SWEPP and Rockwell-Hanford. Figure 15 shows the pink drum assay data taken at SWEPP over a 6-month period ending in early May 1985. As can be seen, both passive and active measurements fall generally within a ±5% measurement error band spanning the time duration of the measurements. These data agree with original measurements at Los Alamos made before shipment to SWEPP. We do not believe the error band is indicative of instrumental drift; the detectors themselves have a long and well-documented history at Los Alamos of ±1% stability or better. We believe, rather, that the band is caused by a combination of statistical and positional measurement errors. At any rate, a ±5% reproducible system calibration, made each day that assays are carried out and that is repeatable over a time period of months and eventually years, is a satisfactory result.

VIII. SYSTEMATIC BACKGROUND DETERMINATIONS

A. Active Assay

We have observed over many years of development that there is a small and persistent active signal background (residual after all usual cosmic ray and drum passive source backgrounds have been removed) characteristically found in different dieaway measurements. This can be minimized through careful design and construction of shielded detector packages. However, some background remains even after this effort.

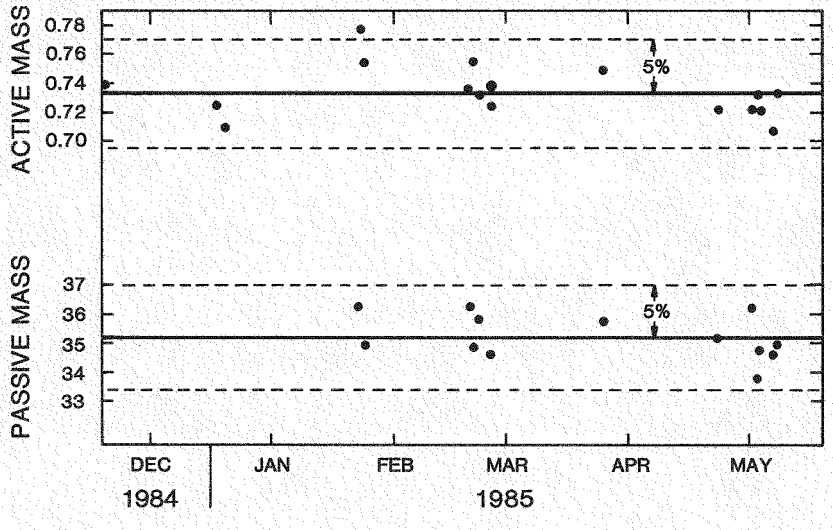

Fig. 15.
Pink drum measurements at SWEPP. The passive and active standards are assayed and the data analyzed as if the standards were actual waste.

Our studies indicate that the principal source of this background is interrogation thermal neutron leakage into the detector packages. Because a thermal neutron flux is described by a Maxwellian distribution in energy, one can calculate that perhaps as much as one part in 10^7 of the total thermal flux exists at energies of 0.6 eV or greater at any given time. This relatively energetic fraction can penetrate the cadmium shielding layer and produce counts indistinguishable from a fission signal. The calculated magnitude of this effect is of the same order as the observed true quiescent background.

An additional contribution to the background is produced by the two-stage effect of high-energy neutron-capture gamma rays from aluminum, cadmium, iron, or other structural or matrix materials that subsequently produce photoneutrons (threshold 2.23 MeV) in the natural deuterium fraction of the polyethylene surrounding the ^{3}He neutron detectors. This is a small effect, but it has been observed and is definitely contributory.

Regardless of the ultimate origin of background, it must be accounted for properly in order to perform accurate low-level fissile assays (i.e., milligram quantities of ^{239}Pu or ^{235}U). Our systematic studies (i.e., compilation of backgrounds found with the 20 or more matrices used for matrix response studies) indicate that both the matrix absorber and moderator contents affect the background. Our most successful absorption-moderator index correlation function used to describe the background in a systematic fashion is

$$\left(\frac{\text{shielded totals count}}{\text{flux monitor count}}\right)_{\text{background}} = \frac{[0.0036 + 0.00139\ \ln(\text{absorption index})]}{[(1 - \text{moderator index})^{-1} + 1]} \quad . \qquad (17)$$

A plot of the quantity

$$\left(\frac{\text{shielded totals count}}{\text{flux monitor count}}\right)_{\text{background}} \cdot [(1 - \text{moderator index})^{-1} + 1]$$

appears in Fig. 16 to illustrate the quality of the correlation. (The correlation coefficient for the 16 matrices measured in the Rockwell-Hanford system is 0.816.)

B. Passive Assay

The passive backgrounds are produced by cosmic ray interactions within the assay chamber and matrix materials. The principal variable here is that of height above sea level: the observed backgrounds at Los Alamos and the SWEPP facility are 5 to 10 times larger than those observed at Rockwell-Hanford under

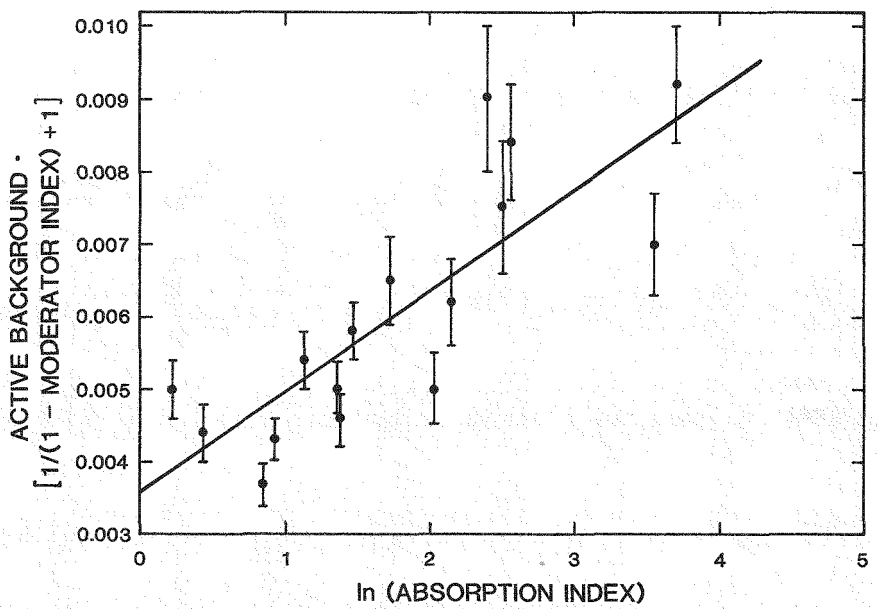

Fig. 16.
The active background is a strong function of both absorption index and moderator index. Note that the moderator index portion of this dependence has been removed from this plot by the indicated factor.

otherwise identical conditions. Therefore, it is necessary to use measured basic passive backgrounds at each site. Because atmospheric pressure changes alter the incident cosmic ray flux, it is also necessary to make daily passive background measurements at each site. These facts have all long since been recognized, and the standard operating procedure and program require a new background to be acquired for each data disk.

In addition to these variations, one also observes a change in background due to matrix, principally due to matrix moderator content. We believe this is caused by changes in detection efficiency (that is, changes in the observed efficiency for a neutron source placed within the matrix) that appear to be almost entirely a function of matrix moderator.

Systematic measurements of matrix-dependent cosmic ray backgrounds have to be performed carefully because other variables such as atmospheric pressure changes can easily distort apparent matrix effects. To date our analysis of existing data suggests that at the elevation of Los Alamos and SWEPP the following functional forms are appropriate.

$$(\text{System totals})_{\text{background}} = (\text{System totals})_0[1 - K_1 \cdot \text{moderator index}], \quad (18)$$

$$(\text{System totals coincidence})_{\text{background}} = (\text{System totals coincidence})_0[1 - K_2 \cdot \text{moderator index}]^2, \quad (19)$$

where the quantities with the zero subscript are the measured backgrounds with an empty drum in the assay chamber. The functional forms for all shielded quantity backgrounds would be similar. We are still determining optimum values for these parameters at the time of writing. Furthermore, these background variations are small and only of great consequence for weak passive neutron signals in high hydrogen-content matrices such as sludges. In these cases the passive matrix corrections can be large; hence, it is important to use a very accurate passive background. Preliminary values for both K_1 and K_2 are 0.35 ± 0.10. At the sea level installations (such as Oak Ridge, Rockwell-Hanford, and Savannah River) these effects are extremely small and will almost never be of practical concern.

IX. ASSAY ALGORITHM

A. General Discussion

We have now developed all the separate factors required to determine a complete assay algorithm--or rather, two complete assay algorithms because the passive and active portions are separate and independent. The basic approach we used in developing the assay algorithm is to isolate each major factor entering into the assay and to determine a separate formalism for each factor. The overall algorithm is thus a product function of several factors. Where the factor amounts to a correction for a measurement defect (i.e., matrix absorption or moderation, fissile self-absorption), we have taken the approach of using a normalized factor such that when no correction is required the factor is unity, when the correction is 25%, the factor is 1.25, and so forth. We believe this approach, especially when all factors are also printed out as part of the routine output, maximizes the user's awareness of data quality and also allows him to determine in a simple fashion how much each factor in the algorithm contributes to the assay result.

Because this report covers the second-generation assay units, which will be dedicated to plutonium-contaminated waste assay, we will specialize this algorithm for plutonium waste. (We estimate that over 98% of the assay work at the three sites will be composed of weapons-grade, reactor-grade, and heat-source plutonium measurements.) Note that all three isotopic mixtures (in fact, any plutonium isotopic mixture) are accommodated. An estimate of the total alpha emission inventory is also included so that, in effect, the presence of ^{241}Am is determined as well.

B. Active Neutron Assay Algorithm

The total plutonium mass for the active assay is given by

$$\text{plutonium (g)} = \text{SIG} \cdot \text{F1} \cdot \text{F2} \cdot \text{F3} \cdot \text{ISO} \cdot \text{CALIB}, \quad (20)$$

where SIG is the net active signal corrected for all backgrounds; F1 is the matrix correction factor for absorption of the interrogating flux, Eqs. 8 and 9; F2 is the matrix correction factor for neutron moderator effects, Eqs. 10 and 11; F3 is the fissile material self-absorption correction factor; ISO is the plutonium isotopic correction factor, the inverse of the ^{239}Pu isotopic abundance fraction; and CALIB is the open geometry (empty drum, neutronically thin) calibration in units of grams of ^{239}Pu per net unit signal count (the current best value is 4.35 for the three second-generation units).

This formalism presumes that the plutonium isotopic percentages are known; the operating program allows the operator to supply this information. In the absence of specific isotopic information, usual weapons-grade material (94% ^{239}Pu) is assumed to be present.

The F3 fissile self-absorption factor has not been discussed in detail in previous sections. In fact it is a difficult correction to make because it presupposes knowledge of how the fissile material is distributed within the waste matrix. Currently we have developed a very simple exponential one-parameter self-absorption model (the one parameter is the "many drum" average of the plutonium effective volume based on preliminary Rockwell-Hanford assay data). In this model, all measured masses below 10 g of plutonium would have negligible self-absorption corrections; sizable corrections occur for indicated large plutonium masses. If the indicated plutonium mass is 50 g, the self-absorption correction factor is 36%; if the indicated mass is 100 g, the correction factor is 85%, and so forth.

The approach is inherently conservative in that all indicated large signals are presumed to come from a self-absorbing source. The present simple model should be regarded as a first step; we are developing this concept and anticipate a more sophisticated correction will be introduced at a later time after more comparison data from waste drums have been studied.

In the case of heat-source plutonium, the active signal will generally be small. Isotopic percentages typical of heat-source material are assumed: 20% ^{239}Pu and 80% ^{238}Pu. Minor isotopes are neglected in this formalism.

For reactor-grade plutonium, especially for recycled and breeder reactor material, some contribution to the active signal is expected from ^{241}Pu. In this case, a ^{239}Pu-equivalent isotopic fraction should be used in the isotopic information input phase.

C. Passive Neutron Assay Algorithm

The total plutonium mass for the passive assay is given by

$$\text{plutonium (g)} = \text{SIGP} \cdot \text{F4} \cdot \text{ISOP} \cdot \text{CALIBP} \quad , \quad (21)$$

where SIGP is the net passive coincidence signal corrected for all backgrounds; F4 is the appropriate passive coincidence matrix correction, Eqs. 14 or 15; ISOP is the passive plutonium isotopic factor; and CALIBP is the passive coincidence calibration factor in open geometry in units of plutonium grams per net coincidence count (currently used values are 1.28 for system coincidences and 18.9 for shielded coincidences, assuming standard weapons-grade isotopics).

The input quantity asked for in the operating program is ^{240}Pu%. The algorithm internally ratios the input value to standard weapons-grade plutonium to obtain the appropriate calibration. In the case of heat-source plutonium, the passive input will be a ^{238}Pu "key" so that the operating program will branch to a separate analysis in which the system totals will be used to determine an appropriate ^{238}Pu mass. This formalism will be developed in detail during the test and evaluation work at Savannah River.

Earlier we noted the appropriate quantity--system coincidence or shielded coincidence. In principle, both of these coincidence count rates can be used to determine a total plutonium mass. In fact, system coincidence is appropriate from a statistical accuracy basis for low and moderate count rates. For high count rates, shielded coincidence will almost always produce the statistically more accurate value. An internal operating program key determines in each case which of the two will be used for mass calculations. Currently, this key is system totals. For a system totals count rate below 2000 counts per second, system coincidence is used; shielded coincidence is used above 2000 counts per second.

The system totals value is also used to estimate the total alpha activity in the waste packages. This estimate is based on the fact that alpha particles bombarding light matrix materials such as oxygen, aluminum, magnesium, boron, and fluorine undergo (α,n) reactions with consequent neutron emissions. Historically, a value of two neutrons per second per millicurie of alpha emitter has been used. This value is being re-examined. Our initial measurements at SWEPP appear to support an average emission rate of 5 to 10 neutrons per second per millicurie. Theoretically, even fewer than 2 neutrons per second could be emitted.

The matter will be resolved in the near future; the important fact is that the system totals value does provide a very sensitive if somewhat inaccurate estimate of the internal alpha activity in a waste package. This estimate can be used to determine upper limit bounds on the corresponding heat-source values required by the Waste Isolation Pilot Plant. These estimates are generally conservative, so the function of screening out potentially hazardous high-alpha-activity drums is served admirably. In those cases where independent information confirms that "extra" ^{241}Am is contained in a given package, such as is often given in "tag value" information sheets, one can readily convert the total alpha-activity estimate into an ^{241}Am mass estimate. This can be pursued one step further by using matrix-specific (e.g., sludges where ^{241}Am is found frequently) calibration factors. This should improve those particular alpha-activity estimates. We plan to pursue auxiliary (α,n) measurements at SWEPP in late FY 85 to determine appropriate factors. These measurements will make use of quantitative reaction gamma-ray information.[21]

D. Error Analysis

In most NDA measurements being carried out, little effort has been put into estimating systematic errors. The common practice is to use the measurement statistical error as the basis of the total assay error. This is true for all segmented gamma scanners now in use. Only under the most ideal of circumstances will this procedure result in a realistic total error estimate. We have attempted to improve upon this situation by using our positional matrix response data (see Sec. V) to estimate in a systematic fashion the average error associated with a general matrix.

As we discussed in Sec. V, our matrix response measurements determine the assay system response in each volume element of a matrix drum. If one then models the distribution of TRU isotopes within a waste drum, the volume element response data can be used to estimate total measurement errors. Because we have determined the volume element response for a large number of mockup matrices and have also related these in a systematic manner to the measured absorption and moderator indices, we can apply the modeling process to a general matrix in a systematic fashion. As an initial step, we have calculated the expected average measurement error for a distribution in which all fissile isotopes are assumed to occur within a 4% volume element that can appear anywhere with equal probability throughout the waste drum. Figure 17 is a plot of the calculated average fractional error associated with this distribution for 16 mockup matrices spanning the complete range of absorption and moderator index values encountered to date in actual wastes. These calculated distributional errors have been plotted as a function of the total matrix correction factor.

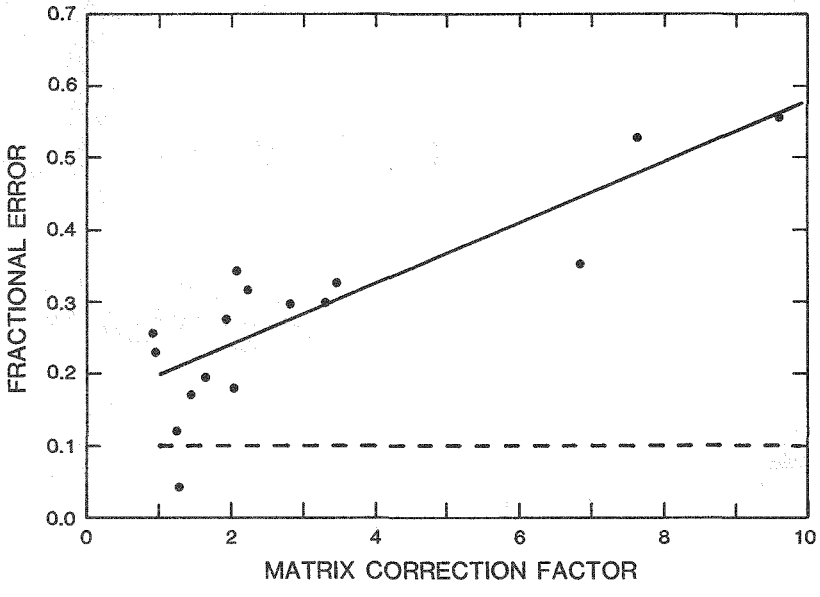

Fig. 17.
Fractional error as a function of the active matrix correction factor. The solid straight line fit is 0.158 + 0.042 × matrix correction factor. The dashed line represents an estimate of the minimum overall systematic error.

As would be expected, the fractional error increases generally as the total matrix correction factor increases. That is, large correction factors are associated with large fractional errors.

If the assumed distribution volume element increases, the fractional error associated with each matrix decreases. We have calculated these errors for a few matrices to get an indication of the effect. For instance, if the volume element increases to 20%, the calculated errors are reduced to about one-half of the 4% volume element values. At the extreme of this effect, a totally uniform distribution throughout the matrix leads to a zero calculated error because the volume-weighted average response has been used to determine the matrix correction factor.

The systematic analytic fits to the matrix correction factors also have an error associated with them [the mean standard deviation of the overall calculated responses (Eqs. 8-11) compared to the measured values (Table II) is 10%]. One does not really have a zero measurement error associated with even a uniform distribution. The dashed line in Fig. 17 represents this minimum 10% error for all matrices on the average. The solid line in Fig. 17 is a fit to the 4% volume element error calculations. This fit predicts a 20% error for the nominal "no matrix correction" case, about a 30% error for a matrix correction of 4, and a 50% error when the matrix correction is as large as 8. In all likelihood the real world wastes will have measurement errors that fall somewhere in between the 4% volume element predictions and the flat minimum error estimate ($\pm$10%) for a uniform TRU isotope distribution. This error calculation procedure can easily be performed for more complicated distribution models than the very simple single-volume element distributions presented here. When more assays are performed with the second-generation assay systems, we will be obtaining information on what the TRU isotope distributions are. These data can then be used to refine the error estimates presented here.

As an initial working estimate of total error based on the two extreme limits of Fig. 17 and an initial study of real wastes, we have used the following relation for the active assay error estimate:

$$\text{Fractional error} = 0.05 + 0.05 \cdot \text{MCFA} \quad . \tag{22}$$

This estimate produces a generally conservative value for the total error that increases linearly with the total matrix correction factor. Statistical errors are compounded within this formalism as well. Generally, statistical errors are wholly negligible for the active neutron measurements with the exception of very low level fissile values--those below 10 mg ^{239}Pu. Statistical errors generally are important for the passive measurement. For most matrices the passive measurement error will be dominated by the statistical component. (Remember that only moderator amounts affect the passive measurement so that generally systematic passive assay errors will be smaller than predicted in Eq. 22.)

X. VERIFICATION STRATEGY

A. General Approach

In the preceding sections we have discussed in detail our combined passive and active neutron assay system. We believe that the calibration, matrix corrections, isotopic effects, and overall assay algorithm we have developed will provide accurate results in routine use. The purpose of this section, however, is to discuss the several ways that the second-generation system's performance can be verified independently and to provide the overall strategy for carrying out these verification activities.

The verification approach we are pursuing is based on using the fielded second-generation assay units in a systematic campaign of measuring TRU waste and "salted" (i.e., known amounts of TRU waste added) waste drums. The strategy is simple: to establish, using multiple independent and comprehensive assay measurements of well-characterized waste and salted waste drums, that this second-generation system and its associated algorithm do indeed provide credible and accurate results.

B. Cross Comparisons of Second-Generation Units

The first step required to carry out this strategy is to ensure that the various Los Alamos second-generation units provide consistent assay results. That is, a waste drum measured with one unit will yield the same result as the same waste drum measured with another unit. We have established this consistency with the passive and active matrix response data and absolute calibration data presented in the foregoing sections. However, we believe it is important to demonstrate the consistency more directly by comparison assays of salted waste drums and well-characterized actual waste drums.

Once the consistency of all Los Alamos second-generation units is established, verification measurements done with any one of the units provide generally valid data applicable to all units. At the date of this writing (June 1985), there are four second-generation units available to carry out the verification strategy--the SWEPP, Rockwell-Hanford, Savannah River, and Mobile units. All four units have been calibrated, and matrix response corrections developed using the standards and mockup matrices as discussed in this report. A fifth second-generation unit, scheduled to be completed by mid-FY 86 and installed at Rocky Flats, will be available later. It, too, will be calibrated and matrix corrections will be determined using the set of standards and mockup matrices discussed in this report.

Some cross-comparison measurements have already been carried out and will be discussed later in this section. However, the overall plan is to greatly enlarge upon this by preparing a set of about 20 well-characterized salted waste drums that can be shipped to all the sites for a set of round-robin measurements. This set of drums will not only provide a definitive cross comparison for all five Los Alamos second-generation units, but the set can also be maintained as long as is required for similar measurements with subsequent Los Alamos neutron units and alternative NDA systems such as segmented gamma scanners.

C. Salted Waste Drums

1. General Discussion. The cornerstone of any verification project must be a set of standards that are well characterized. We believe that for waste assay systems, these standards must themselves be credible representatives of actual waste matrices and also must contain known amounts of specific TRU isotopes. Because the verification program at Los Alamos and the various sites at which the second-generation units have been (or soon will be) implemented is not a specifically funded effort, the preparation of these salted waste drum standards is of necessity a low-budget endeavor.

2. Rockwell-Hanford Salted Drums. The first increment of these low-budget standards was prepared by Rockwell-Hanford personnel under the direction of Randy Roberts and Steve Norton. This is a set of five actual waste drums prepared from "cold" Rockwell-Hanford waste and salted with 97% enriched ^{239}Pu material that was available. These drums were prepared by Rockwell-Hanford (with no involvement from either Los Alamos or other contractor laboratories) strictly for their internal system verification purposes and as part of the acceptance tests performed at Los Alamos before shipment of the Rockwell-Hanford system. These five drums are described in Table V, which includes a brief description of the waste matrix contents of each drum, total mass of the waste matrix, the ^{239}Pu mass added to the matrix, and the results of assay measurements using both the Rockwell-Hanford and Savannah River units. (Both units were functional at Los Alamos at the time of the acceptance tests.)

As can be seen, two of the drums contain typical laboratory wastes and three contain typical decontamination and decommissioning wastes. The ^{239}Pu amounts added were intended to test the assay system's ability to perform at or below the 100-nCi/g level. The first laboratory waste drum contains 10 mg of ^{239}Pu (the plutonium used to salt all drums was prepared in a large number of approximately 10-mg increments; no particular placement sequence was used within any drum so as to simulate actual waste). For this matrix the 10 mg of ^{239}Pu represents about a 20-nCi/g loading, or well below the 100-nCi/g limit. As can be seen, not only did both assay units obtain the same ^{239}Pu mass within experimental errors, both measurements also agree reasonably well with the expected 10-mg value. The agreement is particularly satisfying considering the small net active signal involved.

The second laboratory waste drum matrix is similar in content and mass to the first, but contains 82.7 mg of ^{239}Pu, roughly a 160-nCi/g loading. As can be seen, the cross comparison of the two units is excellent, and both assay values agree well within 1σ of the estimated total experimental error.

The third and fourth drums contain very heavy loadings of typical decontamination and decommissioning waste, with the third containing 57.9 mg of ^{239}Pu (9 nCi/g) and the fourth containing 554.9 mg of ^{239}Pu (90 nCi/g). Again, the cross comparisons are excellent for both drums. A detailed examination of Drum 3 revealed that it contained 150 kg of lead. Our experience has been that most commercial lead contains trace amounts of natural uranium; we estimated the ^{235}U portion of this natural uranium probably contributed an equivalent ^{239}Pu signal to the measurement of about 10 to 15 mg. This is a very small amount

TABLE V

ACTIVE ASSAY RESULTS WITH THE HANFORD PLUTONIUM SALTED WASTE DRUMS

Drum Identification	Waste Matrix Description	Matrix Mass (kg)	Measured Matrix Correction Factor	^{239}Pu Mass Added to Matrix (mg)	Hanford System Assay ^{239}Pu (mg)	Savannah River System Assay ^{239}Pu
84-9-26-1	Laboratory waste: tape, gloves, shoe covers, polyethylene bottles, metal cans	32	1.00	10.0	7.9 ± 1.8	6.5 ± 1.6
84-9-26-2	Laboratory waste: tape, gloves, shoe covers, polyethylene bottles, metal cans	32	1.00	82.7	82 ± 9	85 ± 9
84-9-26-3	Decontamination and decommissioning waste: rebar, steel, lead, concrete	381	1.39	57.9	83 ± 11	81 ± 11
84-9-26-4	Decontamination and decommissioning waste: steel pipe, concrete, sand	375	1.87	554.9	506 ± 73	492 ± 72
84-9-26-5	Decontamination and decommissioning waste: Pyrex glass concrete, steel	178	1.99	272.1	220 ± 35	230 ± 34

(2 nCi/g), but considering the small amount of ^{239}Pu added to this drum originally (58 mg), the effect must be calculated for the purposes of considering how well the assay measurement duplicated the expected total fissile inventory. When this is done, both units produce assay values within experimental errors that agree with the expected response.

The fourth drum did not have a significant lead inventory; instead it had a ^{239}Pu loading 10 times larger. For this drum, as can be seen in Table V, both cross comparisons and absolute assay values are in excellent agreement. It should also be noted that the matrix correction factor for Drum 4 was significant, 1.87. The agreement with expectations is thus an excellent test of our matrix correction formalism.

The fifth and last drum is also a decontamination and decommissioning drum, but contains a different mix of matrix materials, notably a large amount of Pyrex glass, which is a considerable source of boron, a strong neutron absorber. As a result, as can be seen in Table V, the measured matrix correction factor is substantial, 1.99. Again, however, the experimental measurements are in good agreement with the expected value of 272 mg ^{239}Pu (94 nCi/g) as well as with each other in the cross comparison.

To summarize the measurements of this initial set of five salted waste drums: the two second-generation units produced excellent assay results both in regard to unit cross comparison and to agreement with the absolute assay results for the salted fissile inventories. These five drums were a considerable test of the system matrix correction formalism as well, with the measured matrix correction factor being 0% for two drums and 39%, 87%, and 99% for the other three, respectively. Clearly, more such definitive experimental tests will be required before an overall verification result can be declared; however, the performance of the system in this 9- to 160-nCi/g range is excellent. It would appear that the active system performance in the 100-nCi/g and below regime is rather well verified.

These five salted waste drums, with the permission of Rockwell-Hanford, will form a portion of the round-robin set of 20 or so salted waste drums that will be sent to the various sites during the balance of FY 85 and FY 86 to provide detailed cross comparisons and absolute assay results for all the Los Alamos second-generation units.

3. Lynchburg Salted Drums. About a year ago personnel from the Lynchburg Research Center of the Babcock and Wilcox Company contacted Los Alamos. This nondefense contractor has a small but active NDA group using a high-resolution segmented gamma scanner to perform low-level assays of ^{235}U, ^{239}Pu, and ^{241}Am in contaminated soils, laboratory wastes, and decontamination and decommissioning wastes. They had heard of the Los Alamos high-sensitivity neutron-based assay work and expressed an interest in comparing the segmented gamma scanner with the passive-active neutron studies of mutual interest in the low-level $^{235}U/^{239}Pu$ contamination area.

This mutual interest effort has now developed into a small, low-budget project in which Lynchburg has agreed to prepare a set of 15 salted TRU waste drum standards that can be used to cross-compare their segmented gamma scanner

to the Los Alamos passive-active neutron units. The TRU materials to be used for the salting will consist primarily of 12% ^{240}Pu isotopic enrichment material that contains an almost equilibrium ^{241}Am grow-in concentration (20-year grow-in time). This material is ideal for the cross comparisons, as the relatively high ^{240}Pu concentration will produce easily detectable passive neutron signals with low total plutonium masses in the matrix. The presence of the relatively high ^{241}Am concentrations will, in addition, provide valuable and quantitative (α,n) yield information in a variety of waste matrices typical of much of the defense DOE inventory.

The waste matrices will be selected from separated general lab wastes, decontamination and decommissioning wastes, and soils. Lynchburg will select the specific TRU loadings for each drum. Los Alamos has requested that the general TRU loading for each drum be in the area of 1 to 20 g of plutonium, but that no particular care or effort be exercised in TRU isotope placement. This procedure, which will maximize the "actual waste" simulation for these drums, is the same procedure followed by the Rockwell-Hanford team preparing its salted waste drums.

In addition, Lynchburg has agreed to provide two or three drums of local uncontaminated dirt to be taken from the vicinity of a natural uranium ore body outcropping. The area from which this dirt will be taken is now being used for agricultural purposes. Lynchburg estimates from its measurements with the segmented gamma scanner that this soil contains 100 to 200 ppm of natural uranium. We estimate a drum of this dirt could contain 100 mg or more of ^{235}U in the natural uranium fraction (^{239}Pu equivalent signal of 67 mg). The ^{235}U can readily be quantified with the segmented gamma scanner and, of course, a drum of this dirt will provide an easily measured active neutron signal as well. This will be an interesting cross comparison and an absolute active assay verification item.

The schedule calls for some 15 salted waste drums to be prepared by Lynchburg. They will also perform quantitative assay measurements with their segmented gamma scanning system. After this the drums will be shipped to Los Alamos or the Nevada Test Site, with subsequent trans-shipment to the various sites as arrangements can be made. We anticipate these drums could be used to provide verification measurements for the segmented gamma scanners in operation at many of the DOE defense sites. Our primary interest is, of course, in providing verification data for the Los Alamos neutron units. If so directed, we are willing to arrange for segmented gamma scanner verification as well. See Ref. 22 for a characterization of the salted drums.

D. Comparison of Segmented Gamma Scanner and Neutron Assays

1. General Concept. At many DOE sites it is routine practice to assay TRU wastes with commercial-grade, high-resolution segmented gamma scanners. The exact method of use varies from site to site, but generally the wastes so assayed are those having total plutonium loadings greater than 10 g and waste matrices having low gamma-ray absorptions. These matrices are usually thus confined to typical low-density laboratory wastes and some other combustible types. High-density wastes such as soils, concrete, metals, and sludges cannot be assayed with segmented gamma scanners because of the very high gamma-ray

absorption within the matrix. The reason plutonium loadings greater than 10 g are required is because the gamma ray used to quantify ^{239}Pu--414 keV--does not produce a strong enough signal to provide a statistically valid assay at lower loadings for the 10- to 15-minute assay times used routinely at most sites. Note, however, that the intrinsic detection limit for plutonium in low-density matrices is close to 1 g. Much longer count times are required to quantify 1-g amounts, however, and most facilities do not routinely attempt to assay amounts this small.

Nonetheless, in the plutonium loading range above 10 g and for sufficiently low-density matrices, there is a large amount of plutonium-contaminated waste that can, in principle, be accurately assayed with both segmented gamma scanners and the Los Alamos passive-active units on a routine basis. Furthermore, as the segmented gamma scanner assays are usually performed routinely for various waste streams at a given site, and the same drums of waste are then subsequently sent to a central certification facility containing a Los Alamos neutron unit, the segmented gamma scanner and neutron assay comparisons are readily obtained without any extraordinary effort other than verifying paperwork.

Thus, a very large volume of potential verification data of this type is available with little extra effort. The cautions involved here are only those common to any NDA effort--making sure that the paper values are correct, estimating systematic errors, and ensuring that the assays have been performed correctly and in a plutonium and matrix regime where accurate assay values are expected.

2. Rockwell-Hanford Comparisons. Because the Rockwell-Hanford team was very interested in the overall verification effort and was willing to exert the considerable extra effort required to obtain early local operational approvals, we were able to include as part of this report a significant amount of segmented gamma scanner to neutron assay comparisons only a few weeks after the Rockwell-Hanford neutron assay unit was delivered and set up.

As it turned out, because the current waste generations at Rockwell-Hanford include two distinct ^{240}Pu isotopic enrichments--usual 6% weapons grade and a 12% reactor grade--both passive and active neutron assays for what amounts to both weapons grade and reactor grade can be compared with the segmented gamma scanner values. It also appears these ^{240}Pu isotopic values are well characterized. To ensure that this is the case, and in any event to obtain more accurate ^{240}Pu isotopic values for a more meaningful verification effort, the Rockwell-Hanford team has also agreed to carry out a detailed plutonium isotopic analysis for several of these comparison drums. This will be done nondestructively via gamma-ray spectroscopy using a well-established and routine formalism in which the relative intensities of several adjacent gamma-ray lines associated with each of the major plutonium isotopes are determined and the isotopic abundances of each isotope are calculated from the set of observed gamma-ray line intensity ratios.

A plot of this early comparison data is shown in Figs. 18 and 19. Figure 18 shows the experimental ratio of passive neutron and segmented gamma scanner assays of the same drums. The plutonium mass range covered is roughly 10 to 200 g and, as can be seen, a systematic ratio of or close to 1.0 is obtained, with an indicated mean standard deviation of about 15% for this data set. No attempt to separate statistical from positional errors has been made.

Figure 19 shows the same type of comparison for the active neutron and segmented gamma scanner assays for the same set of waste drums. In fact, the active assay values have been corrected for self-absorption (see Sec. IX) using a preliminary semi-empirical model, the single parameter of which has been evaluated with this data set. As can be seen, the observed assay ratio appears to be near 1.0 for this data set as well, with an indicated mean standard deviation of about 20%.

We will obtain a great deal of this type of comparison data at Rockwell-Hanford during the fall of 1985, as well as the experimental ^{240}Pu isotopic values for several waste drums. We expect this data to be of high quality so that credence can be given to this very extensive joint Rockwell-Hanford/Los Alamos verification effort. This single item in the overall verification strategy should be very strong. It should be emphasized that the logic of these comparisons is such that the segmented gamma scanner systematics are being verified at the same time that both passive and active neutron assay systematics are verified. The nuclear physics of the detected events for all three assays are independent, and the matrix effects for each assay technique are also quite different. In short, if the same total plutonium mass is obtained for a given waste drum using each of the three assay techniques, the only reasonable conclusion to draw is that this mass value estimate is accurate; the probability of obtaining the same result independently with each technique without it being correct is virtually nil.

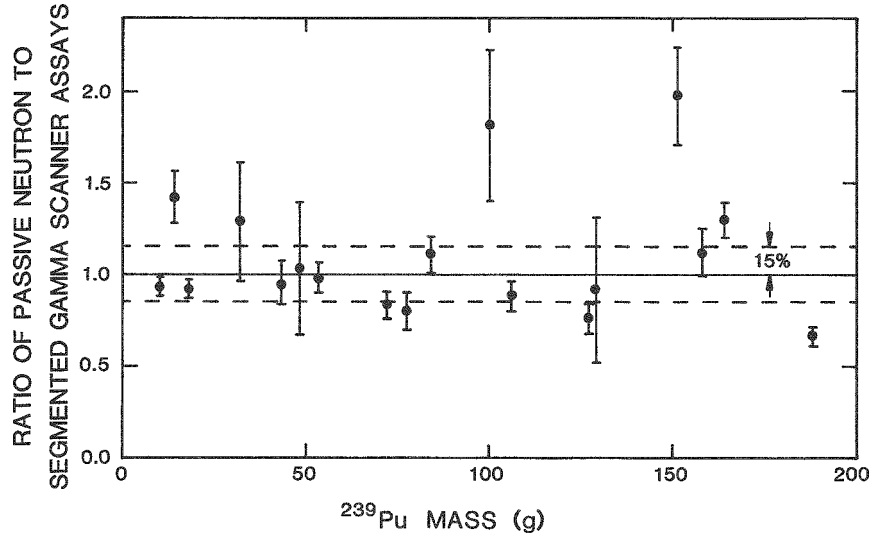

Fig. 18.
Ratio of passive neutron and segmented gamma scanner assays for the initial set of Rockwell-Hanford data.

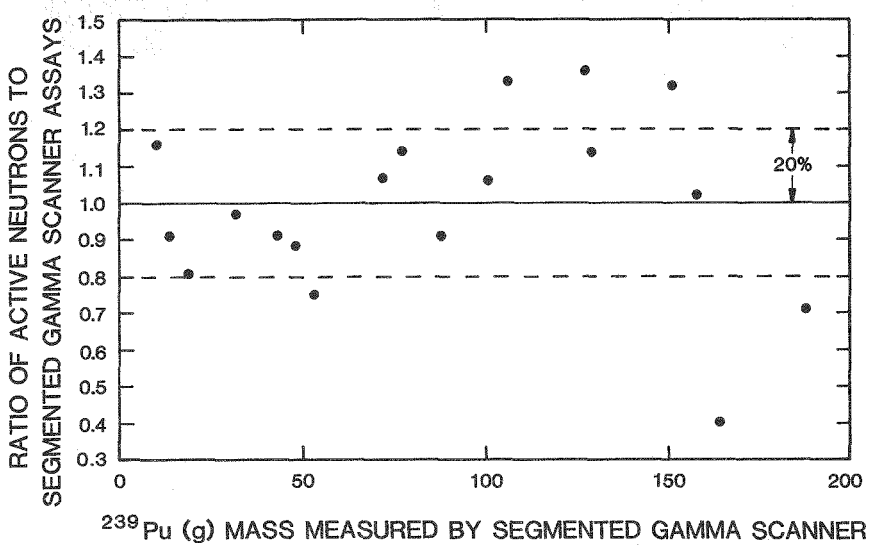

Fig. 19.
Ratio of active neutron and segmented gamma scanner assays for the initial set of Rockwell-Hanford data.

3. Comparisons at Other Sites. We expect the Rockwell-Hanford comparisons to provide the cornerstone for the segmented gamma scanner and neutron assay results. However, similar comparison data will be obtained at other sites as well, using the other Los Alamos second-generation neutron units. For example, in late FY 85 (probably extending into FY 86) Los Alamos will be taking its mobile drum counter to the Nevada Test Site. There it is scheduled to perform an extensive assay campaign on suspect TRU waste drums that are currently in interim storage. These waste drums were generated originally at the Lawrence Livermore National Laboratory. We recently determined that many of these drums (likely to be several hundred total) were assayed at Livermore using a commercial-grade segmented gamma scanner. We contacted the appropriate Livermore personnel (Steve Chen and others) who agreed to provide us with copies of their segmented gamma scanner data for the purpose of comparing it with neutron assays.

We believe that some of the newly generated wastes of Rocky Flats are also now being assayed with a segmented gamma scanner. It appears that some of these drums are or will soon be in the current interim storage inventory at SWEPP. Thus, with a bit of paper-trail sleuthing it should also be possible to compare the segmented gamma scanner with the neutron assay results for the SWEPP unit. It will also be possible to make comparisons directly at Rocky Flats beginning in late FY 86 after we deliver and set up the unit.

Mound Laboratories is currently shipping its waste (principally ^{238}Pu heat-source waste that has been measured by a segmented gamma scanner) to SWEPP. Thus, the possibility of comparisons with the Mound segmented gamma scanner results also exists. This comparison would be most interesting as the isotope quantified with its segmented gamma scanner is ^{238}Pu. We hope to pursue these comparisons later in FY 86 or whenever the time can be devoted to the effort.

Finally, some currently generated Savannah River plutonium wastes, both weapons-grade and heat-source, are now being measured with segmented gamma scanners and passive neutron units. We plan, as part of the test and evaluation program at Savannah River with its passive-active neutron unit, to perform comparison studies with these systems as well.

Thus, it appears that all five second-generation neutron units will ultimately be compared with at least one segmented gamma scanner by the end of FY 86. This should add considerably to the overall verification effort.

E. Comparison of Passive and Active Neutron Assays

1. Conceptual Basis. An additional means of verifying the passive and active assay formalism we have developed is simply to compare as independent assay quantities the separate passive and active values obtained from a given drum. That is, for well-characterized waste--waste for which the plutonium isotopics are known--the passive and active assays can be compared as independent quantities. The passive value is actually based on determination of the ^{240}Pu mass and the active value on determination of the ^{239}Pu mass. The isotopic percentages allow one to derive from each independent assay an estimate of the total plutonium mass.

Thus, for a large number of cases of real TRU wastes, one can, in principle, compare passive and active assay values and determine with some confidence how well the combined system formalism is performing. In short, one obtains valid verification information. The only caveat involved is common to comparison of any two sets of experimental data: one must be careful to select for comparison only those cases for which both passive and active measurements are valid. For instance, if the plutonium within the drum is know to occur in lumps, then obviously the active measurement will not provide a valid result. Another example of an invalid measurement occurs if the passive coincidence signal is masked by an extremely large uncorrelated (α,n) background, a real-world case that occurs on occasion when a large amount of extra ^{241}Am has been placed in a waste drum or when the waste is chemically in the form of a fluoride or other high-yield (α,n) material. In both cases illustrated, one of the two required measurements is rendered invalid for reasons not associated with the assay formalism. Practically speaking, these exceptions are not frequent; most assays of drums containing plutonium of known isotopic composition will be available for the passive-active comparison.

2. Rockwell-Hanford Data. We have already discussed the initial set of Rockwell-Hanford assay data and have shown (Figs. 18 and 19) a comparison of assays, both passive and active, to the segmented gamma scanner. It remains only to show the same set of data as a comparison of passive-to-active neutron assays. This comparison is shown in Fig. 20. As can be seen, the ratio is near 1.0 on a systematic basis, with a mean standard deviation of about 25%. Thus, this initial set of passive-active assay comparisons for Hanford data is quite favorable for verification. A considerably larger data set (hundreds of comparisons) will be available in the near future.

3. SWEPP Data. There is also a large set of passive-active comparisons possible at SWEPP. To date, over 100 waste assays have been carried out; of these we will focus on a smaller subset, the sludges. (The data we will use in this report were acquired during the period November 1984 to early May 1985. Appendix C contains additional assay results obtained in late FY 85.)

The sludges are an extremely interesting case; we found that not only are the average matrix corrections for both passive and active assays quite large, but there are also at least four distinct sludge categories recognized and routinely assigned to different content code values. (The sludges themselves are generated and categorized at Rocky Flats, with subsequent shipment and interim storage at Idaho.) To date, we have analyzed assays of 17 drums of content code 3 sludges, the so-called grease-type sludges. The absorption index varies from 14.2 to 63.1 for this set, and the moderator index varies from 0.322 to 0.598. The overall average active matrix correction factor for this set is 5.91, and the overall average passive matrix correction factor is 2.41. Although individual assay comparisons for this set have a large average measurement error because of these large matrix corrections coupled with plutonium masses that averaged about 1.0 g, if one takes all 17 content code 3

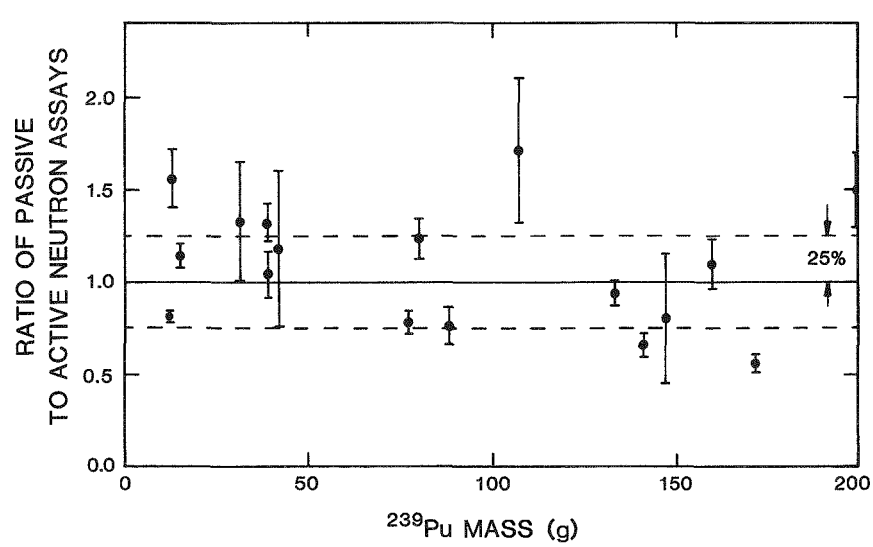

Fig. 20.
Ratio of passive to active neutron assays for the initial set of Rockwell-Hanford data.

passive-active assay comparisons and calculates the set average ratio, a value of 1.06 is obtained with an associated uncertainty of 0.11. This clearly is a favorable result, particularly considering the very large matrix corrections.

Some 11 assay comparisons of content code 7 sludges have also been compiled. For this set the absorption index values range between 10.1 and 30.6, indicating a considerably less-absorbing matrix on the average than is found for the content code 3 wastes. The moderator index values range between 0.429 and 0.768, however, indicating a far more moderating matrix than content code 3. Overall the average active matrix correction factor for this set is 5.48, and the average passive matrix correction factor is 3.81. The average ratio of assayed total plutonium mass from the passive measurement compared to the active is 0.89, with a measurement uncertainty of 0.14. The average plutonium mass for this data set is 1.6 g. Again, this is a favorable result from a verification point of view.

Finally, we have a set of 11 content code 4 sludges for passive-active comparisons. For this set the range of absorption index values is 33.6 to 45.0, and the range of moderator index values is 0.436 to 0.680. The overall average active matrix correction factor is 4.48, and the average passive matrix correction factor is 4.03. For this data set the average plutonium mass obtained is 29 g, and the error for individual assay comparisons is reasonably small. The average passive-active ratio for this set is 0.84, with an uncertainty of 0.11.

Taking the three sets of sludge data collectively, we have a total of 39 individual sludge drum assays for which the overall average passive-active assay ratio is 0.95, with an estimated uncertainty of 0.08. There is no question that this preliminary set of comparison data is encouraging. A considerably larger volume of comparisons at SWEPP will be available in FY 86, including a wide range of content codes in addition to the sludges reported here.

We include in Appendix C additional SWEPP assay data taken in late FY 85. This covers several other matrices: graphite moldings, cemented sludges, ferrous metals, and other metals. The text in Appendix C compares passive and active neutron assays with segmented gamma scanning assays.

4. Additional Comparisons. We have just discussed at some length the passive-active comparisons available at both Rockwell-Hanford and SWEPP, and we have given the results of a fairly extensive initial set of comparison data. (See also Appendix C.) In addition to these two assay units and sites, the Savannah River and Mobile units will be available for similar comparisons at Savannah River and the Nevada Test Site, respectively, during FY 86. We anticipate that by sometime in mid-FY 86, similar comparisons will be generated at Rocky Flats using its soon-to-be-built second-generation unit. In sum, this comparison data, if it continues to be as favorable as the initial results reported here, will be a very strong set of evidence that the systematics of our formalism are correct.

F. Additional Verification Activities

There are additional means available for low-budget verification activities. For instance, all sludge drums are assayed by batch grab sampling techniques. Although this approach generates large uncertainties for the indicated plutonium and americium mass contents for individual drums, if the collective drum assays for an entire batch of sludge are compared with the grab sample assay, one should obtain favorable comparisons. Given the nature of the grab sampling routine, one would expect unbiased comparison results. This verification stratagem may be employed on a long-term basis at the cost of no more than a bit of leg work to obtain the required batch numbers and associated set of waste drums.

We have purposefully considered only the passive-active neutron and segmented gamma scanner techniques as worthy of systematic and quantitative assays of waste drums. This is not to eliminate other NDA measurements completely from consideration. We would propose that other assay means in use at the various sites be considered on an individual basis as well. The odds are that at least some additional valid comparisons may be found. And, in the spirit of the other activities documented here, these verification activities will intrinsically also be low budget.

XI. SUMMARY AND CONCLUSIONS

In this report we have briefly traced out the historical development of the Los Alamos combined passive and active neutron assay system. We discuss in detail the theoretical and experimental physics basis for the assay techniques and their implemented hardware. We develop in great detail the basis for matrix effects on both the active and passive assay measurements. We then describe in a systematic fashion the means we have developed to correct for these matrix effects, including simple analytic fits to a large volume of spatially dependent matrix absorption and moderation response data. These matrix response data are thoroughly described and presented in tables and figures. We describe absolute calibration standards, and our calibration measurements--passive and active--are presented in a complete fashion. We describe our approach to systematic passive and active background determinations and present the analytic fits to a large volume of specific matrix background data. Finally, we present our integrated assay algorithm that brings together all the separate factors required to convert our raw measurements into final assay values, including systematic error estimation.

After describing the complete combined passive and active assay formalism, we then outline the Los Alamos low-budget system verification strategy. This strategy assumes that no incremental funding has been allocated to the specific task of providing a comprehensive verification strategy and to the more difficult job of accomplishing the verification. Under these constraints we describe our general approach of compiling a very large volume of independent and readily available assay measurement comparisons of salted drums and well-characterized actual waste. To greatly expand this verification data base, detailed cross-comparison measurements are carried out for all the second-generation combined passive and active neutron assay units so that verification

measurements made with any one unit apply to all. The set of realistic waste salted drums that already exists for verification purposes is then described, including detailed isotopic and waste matrix characterizations. The only incremental costs involved will be those of shipping these salted drums to the various sites. We then describe the initial set of measurements with salted drums using two separate second-generation units. These measurements provide strong evidence that the system is performing properly in the 100-nCi/g and lower range.

We then describe the concept of independent NDA measurements of isotopically well-characterized waste drums and the verification provided by assay intercomparisons, specifically between segmented gamma scanner and passive neutron measurements, between segmented gamma scanner and active neutron measurements, and between passive and active neutron measurements. We present initial actual comparison data of all three types taken at both Rockwell-Hanford and SWEPP. These first NDA comparison measurements provide strong evidence that the combined passive and active neutron assay system is performing accurately in the 1- to 200-g plutonium range and over an extensive set of both highly absorptive and highly moderating matrices.

The initial measurement described will soon be followed by a large additional set of NDA intercomparisons of waste to be made with all five of the second-generation neutron units, measuring waste drums from at least six major and minor DOE waste-generating sites. Further assay intercomparisons, such as measurements of sludge grab samples, are outlined for an even greater variety of low-budget verification activities.

In conclusion, we have described the Los Alamos combined passive and active neutron assay system and documented all matrix response and absolute calibrations in an easily verified fashion. Reproducibility of both the system design and performance has been amply demonstrated with three independent units and will undoubtedly be further demonstrated by two additional second-generation units. We have also described our specific approach to a comprehensive verification strategy that can be accomplished with little or no incremental funding. The initial set of verification measurements in support of this strategy provides strong support for our contention that the Los Alamos second-generation combined passive and active neutron assay units do provide accurate and reproducible results.

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APPENDIX A

DATA SET FOR DRUMS USED IN THE TEST AND EVALUATION PHASE AND IN THE ANALYSIS OF THE DATA

A. Data Set

Table A-I presents the basic active and passive neutron data from the Oak Ridge National Laboratory for the initial set of 114 drums studied. The drums are arranged roughly in order of increasing fissile content for a passive source $<10^5$ n/s. After all these drums are listed, the remaining $>10^5$ n/s drums are listed.

The first column lists the four-digit drum identification number. The second column lists the results of the fissile mass measurement, before matrix corrections, in units of milligrams equivalent of ^{235}U. The third column lists the net total passive neutron source strength in neutrons per second. The fourth and fifth columns list the results of the least-squares fit to the flux monitor time-history data: A_0 is the normalized amplitude and $T_{1/2}$ is the thermal neutron lifetime. The sixth column lists the ratio of the passive neutron shielded totals count to the systems total count. The seventh column lists the matrix response factor (see next section), and the eighth column lists the matrix corrected fissile mass in units of ^{235}U mg equivalent.

Table A-II presents a qualitative summary of the segmented gamma scanner data results. It shows the distribution of both alpha-emitting isotopes and fission product isotopes within the same set of drums listed in Table A-1.

B. Matrix Corrections

Matrix effects generally affect all experimental data. In the case of the differential dieaway system, the effects of interrogating neutron moderation and absorption are the dominant factors, with moderation of the signal fission spectrum neutrons also being an important factor. In the differential dieaway system measurement, time history for the thermal neutron flux monitor in the cavity actually can be used, in effect, to assay the matrix. It has been found experimentally that different matrices in a 208-ℓ drum produce radically different cavity thermal flux time histories. Examples of this are shown in Refs. 5 and 6, which show normalized thermal flux time histories for five mockup matrices and five actual waste drums. The principal neutronic factors producing these effects are the matrix macroscopic thermal neutron absorption and scattering cross section (Σ_a and Σ_s). These, in turn, are unique functions of the matrix elemental composition and density.

TABLE A-I

ACTIVE AND PASSIVE NEUTRON DATA FOR 114 WASTE DRUMS AT OAK RIDGE NATIONAL LABORATORY

Drum Identification	Uncorrected Fissile Mass (mg ^{235}U equivalent)	Passive Net Neutron Source Strength (n/s)	Thermal Neutron Flux Monitor (A_0, counts/10μs)	Thermal Neutron Flux Monitor ($T_{1/2}$,μs)	(Passive Neutron Shielded Totals)/ (System Totals)	K(Matrix Response Factor)	Matrix Corrected Fissile Mass (mg ^{235}U Equivalent)
1945	3 ± 1	0 ± 4	(4,518)	503	---	1.00	3 ± 1
1921	2 ± 1	0 ± 4	3,593	484	---	1.00	2 ± 1
1924	2 ± 1	0 ± 4	(4,119)	481	---	1.00	2 ± 1
1922	5 ± 2	0 ± 2	4,337	469	---	1.00	5 ± 2
1919	6 ± 2	0 ± 2	4,403	452	---	1.00	6 ± 2
1208	2 ± 2	0 ± 2	4,678	466	---	1.00	2 ± 2
1788	(74 ± 5)	3 ± 2	3,449	551	---	1.00	74 + 5
2101	17 ± 2	0 ± 10	---	---	---	---	17 ± 2
1923	2 ± 1	0 ± 10	5,075	458	---	1.00	2 ± 1
2321	11 ± 4	6.6×10^3	3,670	410	0.266	1.54	17 ± 6
2318	19 ± 4	1.0×10^3	3,784	360	0.251	3.18	60 ± 13
2323	505 ± 6	1.1×10^3	4,319	417	0.256	1.90	960
2281	48 ± 2	2.3×10^3	4,955	527	0.229	1.00	48 ± 2
2324	13 ± 3	3.7×10^3	4,534	417	0.252	1.99	26 ± 6
2282	5 ± 14	9.9×10^4	4,725	423	0.252	2.06	10 ± 28
2312	2 ± 2	4.2×10^2	5,253	438	---	2.33	4 ± 4
2280	48 ± 6	2.5×10^4	6,411	476	0.206	1.00	48

TABLE A-I (continued)

Drum Identification	Uncorrected Fissile Mass (mg ^{235}U equivalent)	Passive Net Neutron Source Strength (n/s)	Thermal Neutron Flux Monitor (A_0, counts/10μs)	Thermal Neutron Flux Monitor ($T_{1/2}$, μs)	(Passive Neutron Shielded Totals)/ (System Totals)	K(Matrix Response Factor)	Matrix-Corrected Fissile Mass (mg ^{235}U Equivalent)
2027	13 ± 10	2.3×10^4	4,810	542	---	1.00	13 ± 10
2201	10 ± 11	1.9×10^4	5,540	418	---	2.57	26 ± 11
1946	18	1.4×10^3	4,012	501	---	1.00	18
1374	29	1.3×10^4	4,511	387	---	3.22	93
2018	7	5.4×10^4	4,960	507	0.238	1.00	7
2025	34	1.8×10^4	6,021	493	0.199	1.00	34
2030	16	1.4×10^4	6,052	489	0.211	1.00	16
2016	31	1.2×10^4	5,586	497	0.223	1.00	31
2035	47	2.3×10^4	5,330	503	0.218	1.00	47
2047	38	3.1×10^4	(5,608)	510	0.216	1.00	38
1935	22	4.8×10^4	5,764	496	0.219	1.00	22
2011	40 ± 11	8.5×10^4	5,788	446	0.221	2.69	108 ± 30
1944	1 ± 6	1.5×10^4	4,208	437	0.253	1.80	2 ± 10
2042	11 ± 4	3.0×10^4	(5,290)	522	0.186	1.00	11 ± 4
2083	70	2.2×10^4	6,374	487	0.204	1.00	70
2004	67	1.5×10^4	4,926	538	0.254	1.00	67
2017	83	2.5×10^3	5,141	525	0.227	1.00	83
1917	58	8×10^3	3,381	372	0.159	1.89	110
1925	52	1.3×10^2	3,540	352	---	3.61	188
2006	65 ± 5	3.5×10^4	6,500	491	0.210	1.00	65 ± 5
1956	55 ± 3	1.8×10^4	3,650	442	0.258	1.52	84 ± 5
1955	110 ± 8	2.2×10^4	3,938	469	0.256	1.00	110 ± 8
2014	118	7.2×10^4	5,077	530	0.228	1.00	118
2151	308 ± 4	3.6×10^1	(4,259)	532	---	1.00	308 ± 4
2029	264 ± 5	3.2×10^4	(4,866)	529	0.192	1.00	264 ± 5
2009	279	1.9×10^4	4,893	530	0.232	1.00	279
1918	200	1.9×10^4	3,419	367	0.260	1.93	386
2031	249	1.0×10^2	2,577	345	---	2.50	623

TABLE A-I (continued)

Drum Identification	Uncorrected Fissile Mass (mg ^{235}U equivalent)	Passive Net Neutron Source Strength (n/s)	Thermal Neutron Flux Monitor (A_0, counts/10 μs)	Thermal Neutron Flux Monitor ($T_{1/2}$, μs)	(Passive Neutron Shielded Totals)/ (System Totals)	K(Matrix Response Factor)	Matrix-Corrected Fissile Mass (mg ^{235}U Equivalent)
1932	181	1.2×10^4	3,728	366	0.248	2.30	416
1676	218	4.7×10^1	5,178	528	---	1.00	218
1996	185	5.4×10^2	4,806	445	---	1.10	389
2026	940	3.4×10^2	4,706	534	msg	1.00	940
1817	714	2.1×10^3	4,635	495	---	1.00	714
1947	540	2.0×10^2	4,028	528	---	1.00	540
2107	640	0 ± 5	4,781	505	---	1.00	640
1949	742	4.7×10^2	3,491	418	---	1.89	1,400
2015	591	5.7×10^2	4,193	527	---	1.00	591
2045	980	3.2×10^4	4,089	454	0.258	1.00	980
1927	550	1.6×10^2	4,778	494	---	1.00	550
2008	910	3.9×10^2	4,975	492	---	1.00	910
2021	570	1.7×10^3	4,574	534	0.256	1.00	570
1060	910	1.4×10^3	4,165	458	0.278	1.00	910
1954	477	1.5×10^2	4,300	457	0.266	1.00	477
1920	66,029	1.4×10^4	(3,727)	435	0.257	1.56	1,030
1952	408 $\pm$ 6	3.6×10^3	(4,201)	439	0.254	1.80	734
2053	1,404 $\pm$ 11	3.2×10^3	2,881	475	0.285	1.00	1,404
1772	16,800	4.9×10^2	4,613	514	0.227	1.00	16,800
1771	13,700	4.3×10^2	4,630	517	0.227	1.00	13,700
1957	2,100	3.4×10^4	3,881	345	0.255	4.12	8,650
1930	1,800	1.2×10^3	3,753	428	0.284$\pm$0.007	1.58	2,840
1977	1,280	5.6×10^2	4,558	481	0.243	1.00	1,280
1776	3,550	2.4×10^2	4,329	417	---	1.86	6,600
SNM-156	29,000	3.4×10^4	6,978	480	0.197	1.00	29,000
2153	20,300	7.4×10^3	3,864	459	0.245	1.00	20,300
2111	1,100	1.4×10^4	msg	msg	0.249	---	1,100

TABLE A-I (continued)

Drum Identification	Uncorrected Fissile Mass (mg ^{235}U equivalent)	Passive Net Neutron Source Strength (n/s)	Thermal Neutron Flux Monitor (A_0, counts/10μs)	Thermal Neutron Flux Monitor ($T_{1/2}$, μs)	(Passive Neutron Shielded Totals)/ (System Totals)	K(Matrix Response Factor)	Matrix Corrected Fissile Mass (mg ^{235}U Equivalent)
1770	13,900	3.5×10^2	4,938	524	---	1.00	13,900
1961	6,370	2.5×10^4	4,527	432	0.251	1.96	12,500
1777	3,000	8.0×10^1	4,493	428	---	1.95	5,850
2046	1,414	1.2×10^4	8,186	495	msg	1.00	1,414
1631	4,814	1.5×10^4	3,693	380	---	2.25	10,800
1798	2,700	1.7×10^3	4,315	477	0.242	1.00	2,700
1789	41,400	1.1×10^3	3,907	527	---	1.00	41,400
2059	2,600	1.4×10^3	4,487	510	0.246	1.00	2,600
2108	1,270	9.0×10^2	4,609	451	0.239	1.50	1,900
1783	1,300	5.3×10^2	5,365	464	---	1.00	1,300
1941	6,400	6.7×10^3	3,430	509	0.282	1.00	6,400
1791	40,800	3.4×10^4	4,771	501	0.211	1.00	40,800
2051	11 ± 36	3.6×10^5	5,378	518	msg	1.00	11 ± 36
2057	36 ± 21	1.2×10^5	6,115	485	msg	1.00	36 ± 21
1940	17	1.2×10^5	---	---	0.246	---	17
2081	42	4.0×10^5	5,599	496	0.230	1.00	42
2075	14	1.9×10^5	4,665	548	0.267	1.00	14
1934	16 ± 9	6.5×10^5	5,500	524	0.222	1.00	16 ± 9
1928	22 ± 13	1.8×10^5	4,825	510	0.237	1.00	22 ± 13
1929	25	1.7×10^5	6,039	479	0.217	1.00	25
2039	11 ± 11	1.0×10^5	(4,775)	472	0.247	1.00	11 ± 11
2005	37	1.5×10^5	5,994	483	0.234a	1.00	37
2112	18	3.1×10^5	5,220	472	0.265	1.00	18
1929	33	2.1×10^5	6,636	483	0.197	1.00	33
2013	7	2.3×10^5	5,064	525	msg	1.00	7
2001	12	1.1×10^5	4,708	542	0.246	1.00	12
2010	10	1.3×10^5	5,215	532	0.228	1.00	10

TABLE A-I (continued)

Drum Identification	Uncorrected Fissile Mass (mg ^{235}U equivalent)	Passive Net Neutron Source Strength (n/s)	Thermal Neutron Flux Monitor (A_0, counts/10 μs)	Thermal Neutron Flux Monitor ($T_{1/2}$, μs)	(Passive Neutron Shielded Totals)/ (System Totals)	K(Matrix Response Factor)	Matrix Corrected Fissile Mass (mg ^{235}U Equivalent)
1926	---	2.7×10^5	---	---	0.197	---	---
2003	78	5.9×10^5	2,575	354	0.286	2,50	195
1951	113	3.6×10^5	5,840	490	msg	1.00	113
2052	124 ± 44	5.5×10^5	6,389	480	msg	1.00	124
1950	674	4.0×10^5	4,891	534	msg	1.00	674
2109	4,800	8.8×10^5	3,587	402	0.276	1.49	7,150
749	45	2.5×10^6	2,910	551	---	1.00	45
2044	0	4.4×10^6	4,088	535	0.267	1.00	0
1998	0	7.6×10^6	3,478	459	---	1.00	0
2094	4 ± 103	2.0×10^6	3.318	377	0.270	1.81	7 ± 180
1933	84 ± 44	2.2×10^6	4,770	526	0.271	1.00	84 ± 44
2110	0 ± 55	2.5×10^6	4,305	476	0.200	1.00	0 ± 55
1959	---	3.0×10^6	---	---	---	---	---
1936	94	2.1×10^6	3,323	372	0.260	1.82	171
1938	75	5.7×10^6	2,617	463	---	1.00	75
1948	106	1.4×10^6	3,152	376	0.293	1.62	172
2019	61 ± 109	1.3×10^6	2,821	365	0.270	1.85	113
1939	700	6.0×10^6	3,869	545	0.133	1.00	700

TABLE A-II

DISTRIBUTION OF ISOTOPES[a]

Isotope	Frequency of Occurrence (drums)	Isotope	Frequency of Occurrence (drums)
Alpha-Emitting Isotopes			
^{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	^{245}Cm	2
^{233}U	11	^{244}Cm	54
^{235}U	9	^{250}Bk	5
$^{237}Np/^{233}Pa$	29	^{249}Cf	44
^{238}Pu	2		
Fission Products			
^{60}Co	39	^{134}Cs	60
$^{106}Ru/^{106}Rh$	43	^{137}Cs	97
^{110}Ag	13	^{144}Ce	25
^{125}Sb	25	^{154}Eu	53

[a]Qualitative analysis of 114 drums of transuranic waste from the Oak Ridge National Laboratory.

Data shown in several of the references indicate that the shape of these curves is exponential after about 1 ms. A careful examination of the Oak Ridge waste drums and a large number of mockup matrices has led us to use the standard time interval 1.5 - 3.0 ms to fit a single exponential curve of the analytic form

$$Y = A_0 \exp\left(\frac{-0.693t}{T_{1/2}}\right) .$$

This is, of course, the standard form in which A_0 is identified as the extrapolated time zero amplitude and $T_{1/2}$, the half-life. In all cases examined, the goodness of fit parameter, chi-squared per degree of freedom, has been observed to be 1.3 or lower, indicating a good fit to the data.

The two least-squares fit parameters may be interpreted as follows. The term $T_{1/2}$ is the thermal neutron half-life in the assay chamber. From diffusion theory, it can be shown to be inversely proportional to the quantity Σ_a. That is, the greater the total absorption, the lower the thermal neutron lifetime. The factor A_0 is the thermal neutron flux extrapolated to time T = 0 and is closely related to Σ_s, although the exact relationship is complex. In the simplest picture, $T_{1/2}$ is a measure of the matrix absorption characteristics and A_0 is a measure of the matrix-moderating characteristics. Empirically, the two parameters, A_0 and $T_{1/2}$, may be used to identify a particular matrix and thus to effect a matrix correction to the assay data.

We have evaluated the response of the system to some 20 individual 208-ℓ drums filled with mockup matrices covering a wide range in the parameter set (A_0, $T_{1/2}$). To assure that a proper volume-weighted response is obtained, we placed three hollow aluminum tubes vertically in each mockup matrix drum, each at a different radial location. A standard source is then measured in each of approximately 10 vertical locations in each radial tube, for a total of approximately 30 measurements with each matrix drum. The net fissile assay quantity

$$\frac{\text{Shielded totals counts}}{\text{Flux monitor counts}}$$

at each location is then volume-averaged to obtain an average response for the drum. The quantity

$$K = \left(\frac{\text{Shielded totals}}{\text{Flux monitor}}\right)_{\text{matrix}} \Big/ \left(\frac{\text{Shielded totals}}{\text{Flux monitor}}\right)_{\text{empty drum}}$$

is calculated for each matrix drum and tabulated as a function of the corresponding set of (A_0, $T_{1/2}$) values.

Finally, an analytic function of the two variables (A_0 and $T_{1/2}$) is determined from a fit to the set of 20 determinations of K as a function of A_0 and $T_{1/2}$. This function is given as follows.

(a) If $T_{1/2} > 450\ \mu s$, then $K = 1.0$.

(b) If $400\ \mu s < T_{1/2} < 450\ \mu s$, then $K = 1.1 + 0.00050(A_0 - 2800)$.

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

(d) If $T_{1/2} < 360\ \mu s$, then $K = 2.5 + 0.0015(A_0 - 2800)$.

The factor K should be regarded as a matrix response factor. Generally speaking, the larger the matrix response factor, the greater the uncertainty in an assay value. We estimate that for K = 1.0, this assay uncertainty is ±20%. For K = 4.0, we estimate the uncertainty to be ±40%. Intermediate K values have correspondingly intermediate uncertainties. Some of the uncertainty is attributable to measurement errors in the matrix identifier quantities (A_0, $T_{1/2}$). Most of the uncertainty is due to other factors such as nonuniform distribution of fissile material in the drum and nonuniform distribution of matrix in the drum. The distribution of K values for the 114 waste drums from Oak Ridge is as follows.

$K = 1.0$ (±20%) for 70% of the drums.

$1.0 < K < 1.5$ (±20%) for 2% of the drums.

$1.5 < K < 2.0$ (±20% to 25%) for 16% of the drums.

$2.0 < K < 3.0$ (±25% to 30%) for 8% of the drums.

$3.0 < K$ (±30% to 40%) for 4% of the drums.

$K_{max} = 4.12$.

A majority (70%) of all the Oak Ridge drums measured require no matrix response correction to the original assay values.

C. Data Interpretation

Table A-III summarizes the passive and active neutron measurements for the 114 Oak Ridge drums. The total fissile mass, obtained from the pulsed active neutron interrogation, is expressed in milligrams equivalent of ^{235}U. Conversion of a value to milligrams of ^{239}Pu is obtained by multiplying by 0.65. For example, a 15-mg ^{235}U mass produces the same active neutron signal as a 10-mg ^{239}Pu mass. Conversion to milligrams of ^{233}U is obtained by multiplying by 1.05. The passive neutron source strength is expressed in units of neutrons per second.

Even though considerably more information exists than is expressed in Table A-III, this simple set of data can be used to certify many waste drums in the non-TRU category. If the passive source intensity is sufficiently low (20 n/s or lower) and if 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

TABLE A-III

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

Passive Neutrons/s	Fissile Mass, ^{235}U Equivalent (mg)					
	0 to 15	15 to 50	50 to 100	100 to 300	300 to 10^3	$>10^3$
$<10^1$	8	---	1	---	1	---
10^1 to 10^2	---	---	---	3	---	1
10^2 to 10^3	1	---	1	1	7	7
10^3 to 10^4	2	3	2	---	5	7
10^4 to 10^5	6	8	4	6	2	7
10^5 to 10^6	6	9	1	2	1	1
$>10^6$	4	1	5	1	---	---
TOTALS	27	21	14	13	16	23

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 A-III can be certified for the non-TRU category. A listing of these drums is shown in Table A-IV.

Also included in Table A-IV (column 4) are the results of quantitative segmented gamma scanner measurements done on these drums. Column 4 shows the gamma-ray assay results for ^{241}Am content in each drum in units of mCi. Because these drums weigh about 50 kg each, a total alpha-emitter inventory of about 5 mCi or less is required for non-TRU certification. As can be seen in Table A-IV, the ^{241}Am contents are well below this level. In addition, other gamma-ray-emitting TRU isotopes were observed in two of the drums (^{228}Th, ^{233}U, ^{249}Cf, and ^{237}U). The activity levels of these isotopes are many orders of magnitude below the legal TRU definition threshold. In short, the comprehensive gamma-ray measurements on this set of drums are in excellent agreement with the passive and active neutron measurements.

Table A-III 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 and an average drum mass of 60 kg. (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 nonfissile TRU isotopic content in these drums is not an easy task. (The Oak Ridge waste is fortunately not typical of most defense TRU waste in the DOE inventory, which contains principally plutonium isotopes with only small amounts of nonfissile TRU isotopes.)

We have used passive neutron multiplicity data to certify another category of the Oak Ridge waste drums to be non-TRU. These are drums that contain ^{252}Cf or ^{244}Cm with negligible amounts of other alpha emitters. We use an analysis in which the measured net multiplicity rates for singles, 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 as much as 10% of the Oak Ridge waste may be certifiable as non-TRU in this category. We use standard well-characterized ^{252}Cf and ^{244}Cm sources for proper calibration. Table A-V shows some representative calibration and waste drum data for this category. This analysis can only be performed if the passive neutron source strength is sufficiently low that accurate multiplicity data may be obtained. Generally, this limits the analysis to drums with about 10^4 n/s sources or lower.

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. 23. 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 ^{233}U.) For this case, a small passive neutron signal in combination with a fairly large active neutron signal (600 mg of ^{233}U in a

TABLE A-IV

DRUMS CERTIFIED AS LESS THAN 100 nCi/g
BASED ON TOTAL FISSILE CONTENT AND TOTAL
PASSIVE NEUTRON SOURCE STRENGTH

Drum Identification	Matrix-Corrected mg ^{235}U equivalent	Total n/s	^{241}Am Gamma-Ray Analysis (mCi)
1945	3 ± 1	0 ± 4	0.59
1921	2 ± 1	0 ± 4	0.17
1924	2 ± 1	0 ± 4	0.049
1922	5 ± 2	0± 2	0.13
1919	6 ± 2	0 ± 2	0.10[a]
1208	2 ± 2	0 ± 2	0
2101	17 ± 2	0 ± 4	(not analyzed)
1923	2 ± 1	0 ± 4	0.11[b]

[a]Additional isotopes: $^{228}Th(7.6x10^{-4}mCi)$, $^{233}U(8.8x10^{-2}mCi)$, $^{237}U(7.4x10^{-4}mCi)$, $^{249}Cf(1.1x10^{-3}mCi)$.

[b]Additional isotopes: $^{228}Th(7.2x10^{-4}mCi)$, $^{249}Cf(3.0x10^{-4}mCi)$.

TABLE A-V

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 (μCi))	^{244}Cm (mCi)	Other α Emitters (mCi)	Active, ^{239}Pu Equivalent (mg)
Background	16.2 ± 0.02	0.229 ± 0.002	0.0259 ± 0.007	0.0043 ± 0.003	---	---	---	---
^{252}Cf[a]	71.2 ± 0.3	12.3 ± 0.1	1.43 ± 0.04	0.11 ± 0.02	0.19	---	---	---
^{244}Cm[a]	199.1 ± 0.5	22.7 ± 0.2	1.72 ± 0.07	0.06 ± 0.02	---	16	---	---
1925	17.0 ± 0.2	1.70 ± 0.05	0.13 ± 0.02	0.006 ± 0.006	---	1.5	17	10
2281	206 ± 1	29.7 ± 0.4	3.14 ± 0.14	0.22 ± 0.05	0.50	---	110	27
2318	107.8 ± 0.6	13.2 ± 0.2	0.92 ± 0.07	0.03 ± 0.02	---	9.0	15	16
2321	606 ± 1	90.1 ± 0.9	8.48 ± 0.42	0.57 ± 0.18	7.4	19	110	13

[a]Calibration standard.

drum is 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, one-year sampling. A listing of the ^{233}U waste drums is shown in Table A-VI. Table A-VII lists the results of the gamma-ray analysis for all alpha-emitting isotopes in the drums listed in Table A-VI.

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. 23, many of the Oak Ridge TRU isotopes are strong gamma-ray emitters that can be quantified with the segmented gamma scanner system.

D. Detailed Neutron and Gamma-Ray Data Analysis Comparison

The ^{241}Am activity in each drum listed in Table A-IV is consistent with the observed neutron count rate. Even for the two drums (1919 and 1923) that have additional isotopes, the additional isotopes do not contribute to the passive neutron signal. The gamma-ray and neutron analyses are consistent for this category of drums.

The gamma-ray analysis does not indicate any ^{244}Cm in drum 1925 (see Table A-V for neutron results). None of the gamma rays from the decay of ^{244}Cm are seen in the gamma-ray spectrum from this drum. Furthermore, neither are the plutonium K x rays. We have not determined the gamma-ray detectability limit for ^{244}Cm, hence 1.5 mCi may be below the gamma-ray detectability limit. From the gamma-ray analysis, only ^{239}Pu could give any sizable neutron signal.

Drum 2281 has been analyzed only qualitatively. Such an analysis does indicate the presence of ^{244}Cm. However, only the plutonium K x rays were observed. It is extremely difficult to unravel that particular energy region of the gamma-ray spectrum, so a quantitative analysis has not been done. A qualitative analysis of the gamma-ray spectrum also indicates the presence of ^{237}U, ^{239}Np, ^{239}Pu, ^{241}Pu, and ^{249}Cf. In addition, the following fission products are seen: $^{106}Ru/^{106}Rh$, ^{110m}Ag, ^{125}Sb, ^{137}Cs, and ^{154}Eu.

There is generally good agreement between the analysis of the neutron data and the analysis of the gamma-ray data for those drums listed in Table A-VI. For those drums where the agreement is good (1771, 1772, 1776, 1777, 1783, 1788, 1789, 1791, 1919), there is no other major potential contributor to the neutron signal (see Table A-VII).

There is potentially some discrepancy on the ^{233}U results for drum 1918; the neutrons indicate 367 mg, the direct ^{233}U gamma rays are not observed. However, ^{241}Pu, which is fissile, is observed in the gamma-ray spectrum and may account for the observed active neutron signal. In other words, drum 1918 probably does not belong in the pure ^{233}U waste category. (The ^{228}Th line indicates a small amount of ^{233}U.)

TABLE A-VI

^{233}U WASTE DRUMS

Drum Identification	Passive Neutron (n/s)	Flux Monitor A_0	Flux Monitor $T_{1/2}$ (μs)	Uncorrected ^{233}U Mass (mg)	Matrix Response K Factor	Corrected ^{233}U Mass (mg)	^{228}Th (mg)	^{233}U Gamma-Ray Assay (mg)
1770	350	4974	492	13,200	1.00	13,200	8.63×10^{-7}	---
1771	430	4630	517	13,000	1.00	13,000	2.16×10^{-3}	1.24×10^{4}
1772	490	3845	509	16,140	1.00	16,140	2.11×10^{-3}	1.32×10^{4}
1776	240	4231	419	3,370	1.86	6,270	4.02×10^{-4}	3.03×10^{3}
1777	80	2545	412	2,848	1.95	5,550	4.61×10^{-4}	2.50×10^{3}
1783	530	5363	464	1,234	1.00	1,234	4.05×10^{-3}	2.64×10^{4}
1788	0	3449	551	70	1.00	70	5.51×10^{-4}	---
1789	1.1×10^{3}	3906	527	39,300	1.00	39,300	1.02×10^{-2}	5.03×10^{4}
1791	3.4×10^{4}	4769	501	38,730	1.00	38,730	6.15×10^{-3}	1.80×10^{4}
1918	1.9×10^{4}	3419	367	190	1.93	367	3.47×10^{-7}	---
1919	0	4403	452	6 ± 2	1.00	6 ± 2	9.25×10^{-7}	9.15
1941	6.7×10^{3}	3430	509	6,080	1.00	6,080	3.20×10^{-6}	---
1957	3.4×10^{4}	3881	345	1,994	4.12	8,215	1.12×10^{-5}	---

TABLE A-VII

GAMMA-RAY ANALYSIS FOR DRUMS LISTED IN TABLE A-VI

Drum Identification	Alpha-Emitting Isotopes (mCi)							
	^{211}Pb	^{228}Th	^{233}U	^{237}U	^{239}Np	^{241}Pu	^{241}Am	^{249}Cf
1770	---	7.1×10^{-4}	0	---	---	---	4.8×10^{-3}	1.8×10^{-5}
1771	---	1.8	1.2×10^{2}	1.02×10^{-1}	---	---	---	6.0×10^{-2}
1772	---	1.7	1.3×10^{2}	---	---	---	---	---
1776	---	3.3×10^{-1}	2.9×10^{1}	---	---	---	---	---
1777	---	3.8×10^{-1}	2.4×10^{1}	---	---	---	---	---
1783	---	3.3	2.5×10^{2}	---	---	---	---	---
1788	---	4.5×10^{-1}	0	---	---	---	---	3.6×10^{-2}
1789	---	8.3	4.9×10^{2}	---	---	---	---	---
1791	---	5.0	1.7×10^{2}	---	---	---	---	---
1918	---	2.8×10^{-4}	0	1.6×10^{-1}	---	1.3×10^{4}	1.4×10^{2}	4.5×10^{-4}
1919	---	7.6×10^{-4}	8.8×10^{-2}	7.4×10^{-4}	---	---	1.0×10^{-1}	1.1×10^{-3}
1941	5.7×10^{-4}	2.6×10^{-3}	0	---	6.9×10^{-3}	---	---	5.3×10^{-4}
1957	---	9.2×10^{-3}	0	1.8	---	7.1×10^{4}	1.7×10^{3}	1.7×10^{-2}

There are three drums where there is a very large discrepancy (>10^3) on the ^{233}U mass. The gamma-ray analysis for drum 1770 is consistent with the observed low passive neutron signal. However, the gamma-ray analysis is totally inconsistent with the large ^{233}U mass determined by the neutron interrogation. This drum should probably be reassayed by both neutron and gamma-ray methods to make sure a spectrum labeling error has not been made.

The same general comments based on the gamma-ray analysis can also be applied to drums 1941 and 1957. However, in these two cases, not only is there disagreement on the ^{233}U mass, there is also disagreement on the large passive neutron signal. The gamma-ray analysis does not indicate the presence of any ^{233}U in either drum, or the presence of any spontaneous neutron emitters. The ^{228}Th lines do indicate the presence of at least some ^{233}U. However, these drums, like 1918 discussed above, appear to be blended waste. That is, they do not contain pure ^{233}U waste and thus do not really fit the analysis conditions for this category. Drum 1957 appears also to contain a large amount of ^{241}Pu. It will take further data analyses to explain the apparently large discrepancy between the neutron and gamma-ray results for these drums.

The nature of the waste at Oak Ridge dictates that fission products will also be present in the drums. The segmented gamma scanner, being sensitive to all gamma rays regardless of their origin, can quantify the fission products as well as the alpha-emitting isotopes. Table A-VIII lists the fission product activity for the drums that have been assayed with both neutrons and gamma rays. The drums in this table are a composite list of drums from Tables A-IV through A-VI.

E. TRU Classification

Most of the 114 drums listed in Table A-I clearly contain amounts of alpha-emitting isotopes in excess of the 100 nCi/g legal TRU definition. Except for the special class of ^{233}U waste (Table A-VI), all drums with a measured fissile mass inventory in excess of 100 mg (for an average Oak Ridge drum weight of 60 kg) must be presumed to contain ^{239}Pu in excess of the allowed 100 nCi/g. For the ^{233}U waste, all drums containing a fissile mass in excess of 600 mg also exceed the legal 100 nCi/g TRU definition. Table A-IX lists the drums that are classified as legal TRU waste based solely on their fissile mass inventories. Column 1 gives the drum identification, column 2 gives the principal fissile isotope, and column 3 shows the calculated fissile TRU isotope inventory in millicuries.

F. Special TRU Oak Ridge Category

As far as can be ascertained, Oak Ridge National Laboratory has a considerable amount of unique waste that no other DOE facility possesses. In the Data Summary Table (Table A-III) these are the drums that fall in the <100-mg-fissile and >10^5 n/s category. These drums do not exceed 100 nCi/g of ^{239}Pu. Also, the passive neutron coincidence measurement on these drums indicates that most of the passive neutrons are due to either ^{252}Cf or ^{244}Cm. By DOE definitions, neither ^{252}Cf nor ^{244}Cm has a sufficiently long half-life to qualify as a legal TRU isotope.

TABLE A-VIII

FISSION PRODUCT ASSAY RESULTS

Drum Identification	Fission Product Isotopes (mCi)					
	^{60}Co	^{106}Ru	^{134}Cs	^{137}Cs	^{144}Ce	^{154}Eu
1208	0	0	0	0	0	0
1770	---	---	0.007	0.032	---	---
1771	0	0	0	0	0	0
1772	0	0	0	0	0	0
1776	0	0	0	0	0	0
1777	0	0	0	0	0	0
1783	---	---	---	7.1	---	---
1788	0	0	0	0	0	0
1789	0	0	0	0	0	0
1791	---	---	---	33	---	---
1918	---	---	---	0.28	---	---
1919	0	0	0	0	0	0
1921	---	---	---	0.12	---	---
1922	---	---	---	0.029	---	---
1923	---	---	0.13	0.59	---	---
1924	0.043	---	---	0.11	---	---
1925	---	---	0.093	0.70	---	---
1941	---	0.82	0.11	0.68	---	---
1945	0.022	---	---	0.14	---	0.053
1957	---	---	---	2.2	---	---

TABLE A-IX

TRANSURANIC WASTE DRUMS BASED ON FISSILE MASS INVENTORY

Drum Identification	Principal Fissile Isotope	Fissile Isotope Inventory (mCi)
1770	^{233}U	132
1771	^{233}U	130
1772	^{233}U	161
1776	^{233}U	63
1777	^{233}U	56
1783	^{233}U	12.3
1789	^{233}U	39
1791	^{233}U	39
1941	^{233}U	61
1957	^{233}U	82
2151	^{239}Pu	15
2029	^{239}Pu	18
2009	^{239}Pu	19
1918	^{239}Pu	26
2031	^{239}Pu	42
1932	^{239}Pu	28
1676	^{239}Pu	15
1996	^{239}Pu	26
2026	^{239}Pu	63
1817	^{239}Pu	47
1947	^{239}Pu	36
2107	^{239}Pu	43
1949	^{239}Pu	93
2015	^{239}Pu	39
2045	^{239}Pu	65
1927	^{239}Pu	37
2008	^{239}Pu	61

TABLE A-IX (continued)

Drum Identification	Principal Fissile Isotope	Fissile Isotope Inventory (mCi)
2021	^{239}Pu	38
1060	^{239}Pu	61
1954	^{239}Pu	32
1920	^{239}Pu	69
1952	^{239}Pu	49
2053	^{239}Pu	94
1930	^{239}Pu	180
1977	^{239}Pu	85
SNM-156	^{239}Pu	1900
2153	^{239}Pu	1350
2111	^{239}Pu	73
1961	^{239}Pu	830
2046	^{239}Pu	94
1631	^{239}Pu	720
1798	^{239}Pu	180
2059	^{239}Pu	170
2108	^{239}Pu	127
2003	^{239}Pu	13
1951	^{239}Pu	8
2052	^{239}Pu	8
1950	^{239}Pu	45
2109	^{239}Pu	477
1936	^{239}Pu	11
1948	^{239}Pu	11
2019	^{239}Pu	8
1939	^{239}Pu	47
2323	^{239}Pu	64

However, these drums also generally contain several millicuries of ^{241}Am or ^{243}Am and occasionally other legal TRU isotopes as determined by the segmented gamma scanner measurements. The segmented gamma scanner data set is not complete for these drums, and some of the early segmented gamma scanner data are suspect from a quantitative point of view because of very high count rates and poor measurement geometry. However, the conclusion that any drum containing a passive neutron source in excess of 10^5 n/s is a de facto TRU drum is not in doubt. Table A-X lists all drums falling in this category. Column 1 shows the drum identification, column 2 shows the calculated fissile TRU isotope inventory in millicuries, and column 3 shows the passive neutron output in neutrons per second.

G. Potential Non-TRU Certifiable Category

After categorizing most of the 114 Oak Ridge waste drums as either non-TRU or definitely TRU (Tables A-IV through A-X), there still remain some drums that, at present, do not clearly fall in either category. These are drums that contain <100 mg fissile mass and a modest (<10^5 n/s) neutron source. These are the drums that, by a combination of passive neutron coincidence and multiplicity measurements with accurate segmented gamma scanner measurements, may be certifiable as non-TRU. Certification of these drums on a routine basis will require careful measurements and great attention to detail. The character of this work is quantitative rather than qualitative.

Table A-XI presents a listing of drums falling in this category. Column 1 gives the drum identification, column 2 the fissile mass in milligrams, and column 3 the passive neutron source strength in neutrons per second.

H. Summary of Data Set and Analysis

The first (and most important) conclusion is that the Oak Ridge waste is atypical. It contains substantial amounts of ^{233}U, ^{244}Cm, ^{252}Cf, ^{243}Am, ^{237}Np, ^{228}Th, ^{245}Cm, ^{250}Bk, and other exotic alpha-emitting isotopes that seldom (if ever) are encountered in waste at other DOE facilities. Thus, detailed data analyses and waste subcategories delineated for the Oak Ridge waste are not likely to be directly applicable to the waste from other DOE facilities.

For the Oak Ridge waste, in spite of its complex makeup, we have been able to identify a substantial number of waste drums that are certifiable as non-TRU. The most straightforward subcategory of this type is the "zero, zero" drum. That is, drums containing <100 mg fissile and emitting <20 n/s passively. (This is also one of the few subcategories at Oak Ridge that has a counterpart at other DOE facilities.) Other examples are (a) ^{233}U waste with <600 mg fissile and <20 n/s passive output and (b) <100 mg fissile content combined with a passive neutron component composed of ^{244}Cm, ^{252}Cf, and <100 nCi/g total other alpha emitters.

All the Oak Ridge waste drums (^{233}U waste excluded) containing >100 mg fissile are in the definite TRU category. The ^{233}U waste drums containing >600 mg fissile are also in the definite TRU category.

TABLE A-X

DE FACTO TRU DRUMS

Drum Identification	^{239}Pu Inventory (mCi)	Passive Neutron Source (n/s)
2051	<3	3.6×10^5
2057	<3	1.2×10^5
1940	<3	1.2×10^5
2081	<4	4.0×10^5
2075	<2	1.9×10^5
1934	<2	6.5×10^5
1928	<3	1.8×10^5
1929	<3	1.7×10^5
2039	<2	1.0×10^5
2005	<4	1.5×10^5
2112	<3	3.1×10^5
1929	<4	2.1×10^5
2013	<2	2.3×10^5
2001	<2	1.1×10^5
2010	<2	1.3×10^5
1926	<3	2.7×10^5
749	<4	2.5×10^6
2044	<3	4.4×10^6
1998	<3	7.6×10^6
2094	<5	2.0×10^6
1933	<5	2.2×10^6
2110	<4	2.5×10^6
1959	<5	3.0×10^6
1938	<6	5.7×10^6

TABLE A-XI

POTENTIAL NON-TRU DRUMS

Drum Identification	^{239}Pu Inventory Upper Limit (mCi)	Passive Neutron Source (n/s)
2280	3.2	2.5×10^4
2312	0.3	4.2×10^2
2282	2.5	9.9×10^4
2324	1.7	3.7×10^3
2014	7.9	7.2×10^4
1955	7.3	2.2×10^4
1956	5.6	1.8×10^4
2006	4.3	3.5×10^4
1917	7.3	8.0×10^3
2017	5.5	2.5×10^3
2004	4.5	1.5×10^4
2083	4.7	2.2×10^4
2042	0.7	3.0×10^4
1944	0.8	1.5×10^4
2011	7.2	8.5×10^4
1935	1.5	4.8×10^4
2047	2.5	3.1×10^4
2035	3.1	2.3×10^4
2016	2.1	1.2×10^4
2030	1.1	1.4×10^4
2025	2.3	1.8×10^4
2018	0.6	5.3×10^4
1374	6.2	1.3×10^4
1946	1.2	1.4×10^3
2201	2.5	1.9×10^4

Guilt by association puts all Oak Ridge waste drums emitting $>10^5$ n/s into the de facto TRU category, even if the fissile component of TRU is <100 nCi/g and most of the passive neutrons are attributable to ^{252}Cf or ^{244}Cm.

Finally, there remain a substantial number of potentially non-TRU certifiable drums that generally contain <100 mg fissile and only a modest passive neutron source ($<10^5$ n/s). The "potential" label means that a very careful passive neutron coincidence and multiplicity measurement, combined with a careful and quantitative segmented gamma scanner measurement, will be required for non-TRU certification. How many potential drums can be certified is not an answerable question at present.

APPENDIX B

ASSAY DATA FOR THE ROCKWELL-HANFORD UNIT

By September 1985, the Rockwell-Hanford unit had been used to perform over 300 assay measurements on actual Rockwell-Hanford suspect TRU waste drums. A set of five salted TRU waste drums was also prepared by the Hanford staff to test the system's performance in the 100-nCi/g range and for several typical matrices including 380 kg of high-density decontamination and decommissioning waste. The system performed exceptionally well on this set of salted waste drums.

Much of the Hanford waste is also assayed in a high-resolution segmented gamma scanner system. Cross comparisons may thus be made with the three independent assay techniques--active neutron, passive neutron, and segmented gamma scanner.

Table B-I shows some of the pink drum calibration data taken at Hanford. The data in Table B-I were taken over a one-week period to investigate the reproducibility of measurements. As can be seen, all measured quantities fall within a small error band--generally that predicted by counting statistics with a 1% to 3% positioning error superposed. Overall standard deviations for the measurements fall within 5% of the corresponding average values. This is in essential agreement with the pink drum measurements done at SWEPP covering a several-month period.

Table B-II shows a summary data sheet for those drums measured as of September 1985 at Hanford that have been determined as non-TRU based on the combined passive-active neutron measurements. These drums were in the suspect TRU category before the neutron measurements and thus represent a considerable economic savings because final disposal costs will be greatly diminished. Of the 200 actual drums measured to date at Hanford (many of these have been subjected to multiple assays), 44 were found to be non-TRU, a rate of about 22%.

We set up a systematic spread sheet analysis for all the second-generation assay system data and developed a means of reading the LeCroy 3500 floppy disks directly into an IBM personal computer and from there into the well-known spread sheet program LOTUS 1,2,3. All required hardware was installed by October 1985 as well as the first version of the required interfacing software. When this process is streamlined, we will be able to manipulate the large amounts of assay comparison data being generated now in a reasonable fashion. This will also be available to the various site contractors and to WIPP. It will greatly facilitate archiving and data certification.

TABLE B-I

SYSTEMATIC MEASUREMENTS WITH THE CALIBRATION DRUM AT ROCKWELL-HANFORD

Run	Time	Date	System Totals	Shielded Totals	System Coincidence	Shielded Coincidence	Passive Mass (g)	Active Mass (g)
1	12:36	6/6/85	154.2 ± .6	34.2 ± .3	21.1 ± 3	1.39 ± 0.6	28.5	0.478
2	12:49	6/6/85	153.7	34.5	21.3	1.44	28.4	0.476
3	13:05	6/6/85	154.7	34.2	21.5	1.28	29.1	0.453
4	13:18	6/6/85	154.4	34.1	21.0	1.30	28.6	0.473
5	13:30	6/6/85	154.2	34.5	21.0	1.27	28.1	0.471
6	13:43	6/6/85	153.9	34.3	21.3	1.28	28.6	0.447
7	13:55	6/6/85	154.0	35.0	21.3	1.42	28.0	0.482
8	14:07	6/6/85	153.8	34.1	21.3	1.29	28.8	0.464
9	12:22	6/7/85	154.7	34.9	21.1	1.34	27.9	0.471
10	12:35	6/7/85	155.3	34.8	21.2	1.39	28.2	0.469
11	12:48	6/7/85	153.6	34.8	21.1	1.36	27.7	0.468
12	13:00	6/7/85	155.3	35.3	21.6	1.40	28.3	0.490
13	13:13	6/7/85	152.7	34.1	20.8	1.30	27.9	0.487
14	13:31	6/7/85	156.4	36.1	21.5	1.33	27.6	0.468
15	13:45	6/7/85	156.0	35.3	21.4	1.45	28.2	0.516
16	13:58	6/7/85	154.4	35.0	21.4	1.29	28.2	0.467
17	12:11	6/12/85	153.7	34.0	21.2	1.33	28.8	0.450
18	12:23	6/12/85	154.5	34.7	21.5	1.39	28.7	0.471
19	12:37	6/12/85	153.3	34.0	20.9	1.33	28.2	0.468
20	12:54	6/12/85	153.0	34.1	21.1	1.20	28.3	0.464
21	13:08	6/12/85	155.5	34.7	22.0	1.35	29.5	0.466
22	13:21	6/12/85	153.1	33.6	20.8	1.36	28.6	0.461
23	13:37	6/12/85	153.5	34.2	21.2	1.34	28.5	0.459

TABLE B-II

NON-TRU DRUM MEASUREMENTS TO DATE

Drum ID	System Totals (counts/s)	Absorption Index	Passive Assay (g of Plutonium)	Active Assay (g of Plutonium)	Indicated (nCi/g)
A-12545	-0.2	1.9	-0.03 ± .03	0.001 ± .001	2 ± 2
A-12539	-0.3	2.1	-0.01 ± .02	0.001 ± .001	1 ± 2
A-12533	-0.3	1.6	-0.01 ± .02	0.001 ± .001	4 ± 4
A-12540	-0.3	1.7	-0.02 ± .02	0.002 ± .001	4 ± 2
A-12544	-0.5	4.4	-0.02 ± .02	-0.001 ± .001	-3 ± 2
A-12561	0.0	2.4	-0.02 ± .03	0.001 ± .002	1 ± 2
A-12587	0.1	2.1	-0.04 ± .03	-0.001 ± .002	-1 ± 2
A-12562	0.0	2.1	0.00 ± .03	-0.001 ± .002	-1 ± 2
A-12549	0.2	2.4	-0.01 ± .03	0.000 ± .002	0 ± 2
A-12564	0.1	1.8	-0.01 ± .03	0.001 ± .002	1 ± 2
A-12546	-0.1	4.6	0.00 ± .03	0.002 ± .002	2 ± 2
A-12560	-0.0	2.0	0.01 ± .03	-0.001 ± .002	-1 ± 2
A-12583	0.1	1.8	-0.01 ± .03	0.000 ± .002	0 ± 2
A-12588	-0.1	1.9	-0.03 ± .03	0.001 ± .002	1 ± 2
A-12590	0.1	2.5	-0.01 ± .03	0.002 ± .002	2 ± 2
A-12584	0.	2.2	0.02 ± .03	0.001 ± .002	1 ± 2
A-12581	0.	2.8	-0.04 ± .03	-0.001 ± .002	-1 ± 2
A-12589	2.1	2.1	0.01 ± .03	0.016 ± .002	15 ± 3
A-12548	-0.1	3.0	0.03 ± .03	-0.002 ± .002	-2 ± 2
A-12582	0.1	2.4	-0.01 ± .03	-0.001 ± .002	-1 ± 2
A-12563	0.1	2.4	0.00 ± .03	0.004 ± .002	6 ± 2
A-12547	0.1	5.3	-0.02 ± .03	-0.003 ± .002	-3 ± 2
A-12573	27.	3.4	0.04 ± .05	0.059 ± .006	86 ± 9
A-12662	4.0	34.8	0.00 ± .03	0.018 ± .005	27 ± 8

TABLE B-II (continued)

Drum ID	System Totals (counts/s)	Absorption Index	Passive Assay (g of Plutonium)	Active Assay (g of Plutonium)	Indicated (nCi/g)
A-12772	3.4	3.3	0.05 ± .02	0.050 ± .006	76 ± 9
A-12777	-0.6	2.6	-0.03 ± .09	-0.001 ± .002	-2 ± 4
A-12781	-0.5	15.6	0.02 ± .02	-0.014 ± .004	-22 ± 6
A-12788	-0.2	2.3	0.00 ± .01	0.001 ± .003	-1 ± 4
A-12776	-0.3	3.0	-0.02 ± .03	-0.001 ± .003	-2 ± 5
A-12779	0.0	1.7	-0.01 ± .02	-0.001 ± .002	-2 ± 5
A-12780	-0.5	2.0	-0.01 ± .03	-0.002 ± .002	-3 ± 5
A-12787	-0.2	2.4	0.00 ± .01	-0.001 ± .002	-2 ± 4
A-12782	-0.1	3.2	-0.02 ± .02	0.000 ± .003	-1 ± 3
A-12784	-0.5	2.6	-0.01 ± .01	0.001 ± .003	-2 ± 5
A-12778	-0.3	5.0	-0.02 ± .05	-0.003 ± .004	-5 ± 6
A-12785	-0.2	2.9	0.00 ± .01	-0.002 ± .003	-3 ± 4
A-12789	-0.5	3.0	-0.02 ± .02	-0.004 ± .004	-4 ± 4
A-12771	-0.4	2.4	-0.01 ± .03	0.005 ± .003	5 ± 3
A-12790	-0.2	2.4	-0.03 ± .27	0.000 ± .003	0 ± 3
A-12786	-0.2	1.8	-0.03 ± .02	0.000 ± .003	4 ± 2
85-3-004	0.1	2.4	0.04 ± .03	0.004 ± .002	4 ± 2

APPENDIX C

ADDITIONAL MEASUREMENTS AT SWEPP

Some of the comparison data taken with the SWEPP unit are shown in Tables C-I through C-III to illustrate how the assay system performs with different waste matrices. Table C-I, for instance, shows data taken with 28 drums of content code 300 matrices at SWEPP. The matrix (graphite molds) is generally both a light absorber and moderator. That is, matrix corrections to these data are generally small. As can be seen, the passive and active neutron data are in good agreement for this data set, with systematic deviation occurring only for high plutonium loadings, as expected, based on a self-absorption model. The active-passive assay ratio for the 19 assays below 20 g of plutonium is 1.07. The Rocky Flats tag values are presumed to be based on a segmented gamma scanner assay. Los Alamos will be working with both Rocky Flats and EG&G/Idaho in FY 86, as part of our technology transfer task, to determine segmented gamma scanner assay results and errors in a systematic fashion. Preliminary comparisons shown in Table C-I are reasonably favorable but must be verified and errors determined. For the 24 drums for which a Rocky Flats assay value was available to us, the average assay ratio between passive neutron and segmented gamma scanner is 0.88.

Tables C-II and C-III both show systematic results for two Rocky Flats matrices that are both highly absorbing and highly moderating. Table C-II shows the passive and active neutron assay results for 13 content code 292, or cemented sludge wastes. Nominal plutonium content ranges from about 8 to 40 g. The average ratio between active and passive assays is 0.75.

Table C-III shows the results of passive and active neutron assays of 19 content code 4 sludge drums from Rocky Flats. Nominal plutonium content ranges from 0.3 to 40 g. The average ratio between active and passive assays is 1.24.

Thus, for both these matrices (which have large matrix correction factors for both active and passive assay measurements), the basic passive and active assay algorithms yield self-consistent results over a considerable range of plutonium content.

Although it appears that the basic passive and active assay algorithms are substantially verified based on the assay comparisons and salted waste drum results just described,a much larger volume of assay comparison data and salted drum measurements will be available for verification in FY 86.

TABLE C-I

ROCKY FLATS CONTENT CODE 300 (GRAPHITE MOLDS) ASSAY DATA

Drum ID	Passive Assay (g)	Active Assay (g)	Rocky Flats Assay (g)	Active/ Passive	Passive/ Rocky Flats
3201305	3.3 ± 0.2	2.8 ± 0.3	4	0.85	0.83
1213866	4.6 ± 0.5	6.6 ± 0.7	5.6	0.7	0.82
3201278	5.2 ± 0.3	4.0 ± 0.4	7	0.77	0.74
1212907	5.8 ± 0.3	6.4 ± 0.5		1.1	
1213998	6.2 ± 0.5	7.2 ± 0.7	12	1.16	0.52
1213085	6.7 ± 0.4	6.6 ± 0.7	8	0.99	0.91
1213889	7.3 ± 0.4	7.8 ± 0.8		1.07	
3201259	7.7 ± 0.3	7.9 ± .8	7	1.03	1.1
3201317	8.0 ± 0.7	10.4 ± 1.0	13	1.3	0.62
1213993	8.1 ± 0.5	9.2 ± 0.9	10	1.13	0.81
1213885	8.3 ± 0.4	9.8 ± 1.0	6	1.18	1.38
1213906	8.4 ± 0.5	9.2 ± 0.9	14	1.09	0.6
1213674	9.4 ± 0.6	9.8 ± 1.0	15.2	1.04	0.62
1213223	11.2 ± 0.6	12.7 ± 1.3		1.13	
1213698	11.5 ± 0.7	15.1 ± 1.5	15	1.31	0.88
1213646	11.7 ± 0.8	12.7 ± 1.3	18.9	1.08	0.62
1213673	12.6 ± 0.7	15.6 ± 1.6	16	1.24	0.79
1213918	12.5 ± 0.7	13.2 ± 1.3	18	1.05	0.7
1213937	14.8 ± .8	16.7 ± 1.7	20	1.13	0.74
1213816	20 ± 1	15 ± 2	39	0.75	0.51
3201298	20 ± 1	18 ± 2	28	0.9	0.72
3201261	22 ± 1	18 ± 2	22	0.82	1
1212326	24 ± 1	24 ± 2	26	1	0.92
1213836	40 ± 3	30 ± 3	47.7	0.75	0.84
1213864	46 ± 1	25 ± 3	48.4	0.54	0.95
320221	97 ± 5	50 ± 5	101	0.52	0.96
1213819	126 ± 3	25 ± 3	48	0.2	2.63
1213917	136 ± 12	29 ± 3		0.21	

TABLE C-II

ROCKY FLATS CONTENT CODE 292 (CEMENTED SLUDGES) ASSAY DATA

Drum ID	Passive Assay (g)	Active Assay (g)	Active/ Passive
246222	8 ± 2	8 ± 1	1.02
246540	13 ± 2	8 ± 1	0.64
246083	14 ± 2	12 ± 2	0.83
2503361	16 ± 3	12 ± 1	0.775
246030	16 ± 2	12 ± 2	0.73
246313	17 ± 2	14 ± 2	0.82
245893	17 ± 3	15 ± 2	0.89
246120	20 ± 3	18 ± 3	0.78
245964	24 ± 4	15 ± 2	0.64
246310	29 ± 3	17 ± 2	0.59
245969	31 ± 5	22 ± 3	0.71
245989	42 ± 6	30 ± 4	0.71

TABLE C-III

ROCKY FLATS CONTENT CODE 4 (SLUDGE) ASSAY DATA

Drum ID	Passive Assay (g)	Active Assay (g)	Active/ Passive
2500426	0.3 ± 0.2	0.3 ± 0.1	1
74404185	3 ± 1	5 ± 2	1.67
74494194	4 ± 1	4 ± 1	0.97
74404205	4 ± 1	2 ± 1	0.54
74404083	4 ± 1	5 ± 2	1.25
74404076	5 ± 1	10 ± 4	1.95
74404201	9 ± 2	9 ± 2	0.99
74404208	9 ± 2	10 ± 4	1.11
74404151	10 ± 2	19 ± 7	1.89
74404179	12 ± 2	17 ± 7	1.42
74404181	16 ± 3	23 ± 9	1.44
74404090	16 ± 3	22 ± 10	1.37
74404197	19 ± 4	22 ± 8	1.16
74404189	20 ± 3	19 ± 8	0.95
74404184	22 ± 4	23 ± 10	1.04
74404203	26 ± 4	24 ± 9	0.92
74404198	32 ± 7	41 ± 13	1.28
74404081	37 ± 7	40 ± 14	1.08

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