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Program Status Report

September—December 1976

by

**Nuclear Safeguards Research Groups R-1 and R-2
Nuclear Safeguards, Reactor Safety and Technology Division**

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ABSTRACT

This report presents the status of the Nondestructive Assay Research and Development programs pursued by the LASL Nuclear Safeguards R&D Groups R-1 and R-2 during the period September-December 1976. Salient topics of the programs are summarized in the table of contents.

CONTENTS

PART 1. Nuclear Material Safeguards Program—Nondestructive Assay Techniques	1
I. Nondestructive Assay Applications and Results	1
A. Application of the Shuffler System with Shielding for Small Hot Samples	1
B. Experience with the Barrel Handler on the CMB-11 Segmented Gamma Scanner	2
C. Improvements in the Van de Graaff Small Sample Assay Station (SSAS)	2
II. Instrument Development and Measurement Controls	4
A. New Shift Register for High Count Rate Coincidence Applications	4
B. Effects of Self-Multiplication on Coincidence Counts of PuO ₂ Cans	6
C. Dual Range Thermal-Neutron Coincidence Counter	8
D. Portable Neutron Assay System for Light Water Reactor (LWR) Fuel Assemblies	10
E. Upgrading of the Segmented Gamma Scanner Software to Measure ²⁴¹ Pu and ²³⁹ Pu Simultaneously	11
F. Evaluation of the IAEA Gamma NP361 Five-Channel Analyzer	12
G. Prototype for Neutron Interrogation of 55-gal Barrels	12
H. Calculational Support for the Assay of Fast Flux Test Facility (FFTF) Fuel Bundles	15
I. Photoneutron Assay of Bulk PuO ₂ Samples	18
J. Bremsstrahlung Photoneutron Sources	22
K. Upgrading of the MEGAS Box Counter	25
L. Adding the DYMAC Data Link to the CMB-11 Segmented Gamma Scanner (SGS)	27
M. Determination of the Total Uranium Content in Materials Test Reactor (MTR) Fuel by K-Edge Densitometry	27
N. The ERDA Safeguards Technology Training Program	29

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MASTER

PART 2. Development and Demonstration of Dynamic Materials Control—	
DYMAC Program	34
I. Concepts and Subsystem Development	34
A. NDA Instrumentation	34
B. Data Acquisition	37
C. Data Base Management	37
D. Real-Time Accountability	39
II. DYMAC Implementation	40
A. DP-Site Test and Evaluation Phase	40
B. DYMAC for New LASL Plutonium Facility	45
III. Technology Transfer	49
PART 3. Perimeter Safeguards and Material Verification	50
I. Verification Signature Calculations	50
II. Real-Time Inventory	50
III. Verification Station for the 280-Can Sandia	
Demonstration System	51
IV. Hand-Held Personnel and Vehicle Monitors	51
References	52
Publications and Patents	53
Glossary	54

PART 1

NUCLEAR MATERIAL SAFEGUARDS PROGRAM— NONDESTRUCTIVE ASSAY TECHNIQUES

I. NONDESTRUCTIVE ASSAY APPLICATIONS AND RESULTS

A. Application of the Shuffler System with Shielding for Small Hot Samples (T. W. Crane and H. O. Menlove)

The assay of material having a high gamma-ray activity (>1 R/h) will be an important problem in the control of hot nuclear fuel from safeguards, process control, and waste categorization viewpoints. In addition, material containing ^{233}U -Th mixtures presents special measurement problems because of the high gamma-ray activity from the daughters of ^{232}U . The high gamma-ray background in these materials generally precludes the use of passive gamma-ray methods for assay applications.

To help in evaluating the method of neutron interrogation with delayed-neutron counting, a cylindrical lead shield was fabricated for the Shuffler system (see Ref. 1, p. 25). The shield was 152 mm in diameter, 305 mm long, and had a 25-mm-diam axial hole for sample insertion. The shield size was determined by the usable area within the present Shuffler assay chamber; in most irradiated fuel applications, a shield with thicker lead and a larger sample region will be required.

To evaluate the effect of the lead shielding on the measurements, the response of the Shuffler system was observed both with and without the lead shield, using an 18.4-g uranium sample (93% ^{235}U) and a 0.76-mm-thick cadmium cover for the sample. The data for these measurements are presented in Table I. The operating parameters were: 5 s irradiation time, 5 s counting, and 20 cycles total. The results given are in some cases an average of several runs and the error corresponds to 1σ . The data in Table I show that the lead slightly increases the observed delayed neutron signal from the sample, especially in the case of the cadmium-covered fast-neutron interrogation. This enhancement is probably due to

TABLE I
DATA TAKEN WITH THE
SHUFFLER SYSTEM

	No Lead Shield (counts)	With Lead Shield (counts)
Background	$1\,052 \pm 23$	$1\,017 \pm 19$
^{235}U sample ^a	$38\,844 \pm 197$	$39\,466 \pm 115$
Cadmium-covered ^b ^{235}U sample	$6\,788 \pm 59$	$7\,834 \pm 51$

^a18.4 g, 93% ^{235}U metal foil.

^b0.76-mm-thick cadmium.

the increased flux from neutron scattering within the lead shield.

Also of interest is the lack of an increase in the number of background counts with the insertion of the 61.4-kg lead shield. The previous progress report (see Ref. 2, p. 3) noted that at the 2200-m altitude of Los Alamos a substantial background could be attributed to cosmic-ray-initiated neutrons in a massive lead shield. Recently, it has been observed that a 0.9-m-thick concrete ceiling reduces the cosmic-ray-initiated neutron background by a factor of 20. The test measurements with the Shuffler system were made in an area with a total ceiling thickness of 2.3 m, which will be sufficient to reduce the cosmic-ray-initiated neutrons in the lead shield to negligible levels. Because highly radioactive material will be

assayed in shielded hot cells, the cosmic-ray-initiated background is not expected to limit the assay sensitivity for either a large sample Shuffler system or a passive neutron coincidence system.

These measurements indicate that it should be possible to assay very hot or irradiated samples by using lead shielding combined with neutron interrogation and neutron counting. Additional tests are needed to evaluate the neutron background levels from different types of irradiated fuel materials.

B. Experience with the Barrel Handler on the CMB-11 Segmented Gamma Scanner (SGS) (E. R. Martin and D. F. Jones)

The barrel handler fabricated to our custom specifications by Eberline Instrument Corp. was made operational during this reporting period by exchanging stepping motors, adding external mechanical resonance dampers, changing the optical encoders, and carefully adjusting the mechanical system. The unit can now lift 205 kg a vertical distance of 812 mm in just over 1 min.

After the unit was installed at the present SGS in CMB-11, it was set to operate at an assay speed of one 5-cm segment per 40 s. For many barrels, two passes were necessary because the software was set to require a two-pass assay if the assay peak counts did not exceed the measured background by a factor of at least 2. As a result, the total barrel assay time was about 30 min and had an accuracy in the measured values of $\pm 3\%$. This long assay time was seriously impeding production throughput, so after the system had been installed steps were taken to increase its speed. First, the software was modified to require two passes only if the small table was in use, so that the barrel is now assayed in a single pass, regardless of the resulting spread in the standard deviation estimates. Then the hardware was modified to drive the stepping motor at four times the previous speed, resulting in a net time savings of a factor of 8. We now find that the estimated error has risen to about 10% for an assay time of about 4 min.

The system is in routine use, and the operators have no trouble in understanding system operation.

C. Improvements in the Van de Graaff Small Sample Assay Station (SSAS) (J. D. Brandenberger, M. S. Krick, and E. L. Adams)

Recently, the work at the Van de Graaff SSAS has been directed toward improving the precision of the assay results. The LASL delayed neutron assay (DNA) results in the New Brunswick Laboratory (NBL) "Round Robin II"⁸ (RR II) for UO_2/ThO_2 powder mixtures showed standard deviations of 1% from the NBL prepared values, as indicated in Table II. These large standard deviations (larger than those expected from statistical considerations) masked a ThO_2 -content-related bias in the uranium content assay of the NBL samples that varied up to almost 1% for samples with Th/U ratios of 16 and 25. After the NBL results⁸ were released to LASL, Evans and Malanify,⁴ Krick,⁵ and Krick and Evans⁹ were able to derive correction factors empirically for the combined effects of attenuation, multiple scattering, and neutron multiplication of the thorium content of the samples. Krick^{5,6} was able to correct the "thorium bias" to within a standard deviation of

TABLE II
STANDARD DEVIATIONS IN LASL DNA OF URANIUM CONTENT IN ROUND ROBIN II NBL SAMPLES*

<u>Th/U Ratio</u>	<u>Number of Samples</u>	<u>Relative Standard Deviation (%)</u>
0	35	0.79
10	35	1.17
16	53	1.19
25	65	1.19
All 88 samples		1.16

*Values from Ref. 3.

0.24% relative to standards containing no thorium. The limit on the precision of this correction factor was the large standard deviations, which were derived from multiple runs of many samples and which were excessive compared to statistical-related uncertainties.

Because future assays at the SSAS will face the same limitations in accuracy imposed by large standard deviations and the precision of correction factors to account for matrix-related effects, an effort is being made to make the precision of SSAS measurements approach those due to statistical considerations. To determine the extent of the problem, a statistical analysis was made of all UO_2/ThO_2 powder samples used as standards for assays of RR II. Consecutive measurements made on each of these samples for RR II were grouped into sets of measurements that resulted in statistical uncertainties of about 0.5%. The results of this restudy of the standard sample data are summarized in Table III. The results are that the standard deviations exceed the statistical uncertainties by more than a factor of 3.

To help correct this situation, the old stainless steel ^3He neutron counters were replaced with more

stable aluminum detectors with carbon inner linings. However, a recent test indicated that for a set of 20 runs, each with a statistical uncertainty of 0.40%, the standard deviation was over twice this amount.

Several possible causes were considered for the discrepancy between statistical uncertainties and the standard deviations, but one obvious way of improving the system quickly appeared. In the SSAS, the sample is bombarded for about 40 ms with the counting system gated off. About 7 ms after bombardment, the delayed neutrons emitted by fission fragments from the sample are counted for a period about equal to the bombardment time. Figure 1a shows one such neutron-detector output just before entering the pulse discriminator, which eliminates low-level pulses and noise. This figure shows approximately one complete bombarding and counting cycle with the counting portion appearing in the first half of the oscilloscope photograph. The start of the trace of Fig. 1a shows the transient caused by the gating-on of the pulse preamplifier. This is followed by 60 Hz and other noise superimposed on the much faster signals from neutron

TABLE III
STANDARD DEVIATIONS IN DNA MEASUREMENTS
ON STANDARD SAMPLES USED IN ROUND ROBIN II

Sample Number	Number of Measurements	Approximate Statistical Uncertainty of Each Measurement (%)	Relative Standard Deviation (%)
74	15	0.46	1.36
75	14	0.48	1.46
75	7	0.51	0.66
1058	20	0.44	3.70
1059	3	0.51	1.95
1051	3	0.44	1.78
1065	3	0.44	1.03
75	5	0.43	0.91
99	10	0.44	0.62
Average weighted by the number of measurements		0.46	1.75

(a)

(b)

Fig. 1.

Oscilloscope photographs of the SSAS counting cycle.

pulses that are approximately 2.5 cm high and barely visible in the photograph. Since these neutron pulses are randomly superimposed on a very high level of noise, some counting problems can be expected. This problem was largely solved by replacing the pulse amplifier with one whose pulse-shaping circuits passed few of the lower frequencies. This modification, when the above test was repeated, resulted in a statistical uncertainty of 0.44% and a standard deviation of 0.55%. Thus for statistical uncertainties of about 0.5%, the standard

deviations are not much larger. Whether or not the improvement will extend to higher statistical precisions has not yet been determined.

A further improvement was made in reducing low frequency noise by introducing a high-pass filter in front of the pulse amplifier. The resulting pulses at the output of the amplifier and the input to the discriminator are shown in Fig. 1b, where the low-frequency noise is essentially eliminated. This system has not yet been tested for counting precision.

II. INSTRUMENT DEVELOPMENT AND MEASUREMENT CONTROLS

A. New Shift Register for High Count Rate Coincidence Applications (J. E. Swansen, N. Ensslin, M. S. Krick, and H. O. Menlove)

Three electronic circuits² have been under consideration for use with thermal-neutron coincidence counters. With any of the three circuits large samples of fissionable material can be assayed in the presence of random background rates of at least 30 000 counts/s. However, the variable deadtime counter circuit was vulnerable to changes occurring in the room background during the assay. The updating one-shot circuit required large deadtime cor-

rections and had not been used much. Consequently, a new shift register circuit capable of handling very high rates was built.

A block diagram of the new shift register is shown in Fig. 2. Some of the features that distinguish it from the previous LASL shift register⁷ are:

(1) The new circuit has only one shift register gate and two triggers (one prompt and one delayed), thereby eliminating several integrated circuits. In principle it is identical to the earlier shift register, which had two gates and one trigger, although the timing is reversed in the accidental gate.

Fig. 2.
Block diagram of new shift register.

(2) The delayed trigger is $1000 \mu\text{s}$ long compared with a typical value of $100 \mu\text{s}$ for the older circuit, hence no real coincidences will be present in the accidental gate.

(3) A fixed clock frequency of 2 MHz is used for all timing applications; thus the timing is less critical and does not vary with choice of gate length (8 to $128 \mu\text{s}$).

(4) The predelay can be varied from 1 to $33 \mu\text{s}$, in $1/2\text{-}\mu\text{s}$ increments, independent of gate length. At high count rates it is important to have a long enough predelay to override all amplifier deadtime and pileup effects, as described below.

(5) The up-down counter has a capacity for 99 counts, as opposed to 9 in the older circuit. This is essential for high count rate applications. For example, at 100 000 counts/s the average number of events in a $64\text{-}\mu\text{s}$ gate is 6.4, and nine counts are exceeded about 11% of the time—a large effect, which preferentially reduces the counting of real events.

(6) The scaler readout has been increased from 7 to 9 digits to accommodate larger counts without overflowing.

The first version of this new circuit has been constructed, and has been tested at high count rates using a thermal-neutron coincidence counter of 20% efficiency, four amplifiers of various designs, and four leading-edge discriminators added together. The tests consisted of assaying a ^{252}Cf fission source in the presence of a large random background from an Am-Li source. Figure 3 shows that the new circuit can be used at much higher counting rates than the old. The rates used for Fig. 3 are not unrealistic; 2 kg of PuO_2 (10% ^{240}Pu) will produce a total count rate of 75 000 counts/s at this efficiency.

The dashed line in Fig. 3 corresponds to the uncorrected data and the solid line has been corrected for a bias caused by an imbalance between the real and accidental registers. This bias was measured by counting a purely random Am-Li source separately.

Fig. 3.

Comparison of old and new shift register circuits at high rates with four bipolar amplifiers (0.2- μ s time constant).

Careful tests of a later version of the new shift register showed that it has no internal bias. The bias observed here is due to the time required by the amplifiers to return to zero baseline. At high count rates even a slight deviation from zero will affect the ability of closely following pulses to clear the discriminator threshold. Bipolar amplifiers tend to produce a negative bias; unipolar amplifiers, a positive bias. Figure 4 is an example of the bias from the same amplifiers used in Fig. 3. The amplifier bias must either be corrected for by measuring a random source or eliminated by careful choice of amplifier design or shift register predelay setting. From a study of various amplifier-discriminator combinations, we chose a fast bipolar amplifier and leading-edge discriminator that will minimize this bias. The amplifier is now being constructed and tested.

When the raw data in Fig. 3 were corrected for bias, the result was a straight line on a semilogarithmic scale (the solid line in the figure). Therefore the data can be fit with an updating deadtime correction of

true coincidence = measured coincidence $\times e^{2\tau T}$,
and

T = true totals = measured totals $\times e^{\tau T}$,

where τ is the deadtime, here about 1 μ s, and deadtime corrections of higher order have been ignored. The magnitude of this deadtime can be understood on the basis of amplifier pulse width and shift register input synchronization time.

Fig. 4.

Internal distribution of random pulse events at 120 000 counts/s taken with four bipolar amplifiers (0.2- μ s time constant). There is a well-defined deadtime of about 1 μ s and a small negative bias in expected distribution out to about 5 μ s. One heavy dot/ μ s, linear scale. Exponential baseline has been subtracted.

The new shift register circuit works correctly at high rates and experiences only small, easily understood deadtime effects. It is now being packaged for use in the IAEA high-level neutron coincidence counter (HLNCC) and the DYMAL program.

B. Effects of Self-Multiplication on Coincidence Counts of PuO₂ Cans (N. Ensslin and J. L. Sapir)

One potential application of thermal-neutron coincidence counters is the assay of large amounts of plutonium in oxide form. A typical problem facing the IAEA, the DYMAL program, and other organizations is the assay of 1 to 2 kg of PuO₂ (10-20% ²⁴⁰Pu) in cans 12 to 30 cm high and 8 to 10 cm in diameter. During the past year we have studied the problems of detecting coincidence events in the presence of the large accidental count rates produced by such samples. The new shift register that resulted from this work can operate at rates over 100 000 counts/s with reasonable deadtime corrections. Consequently, the largest remaining

problem in the assay of PuO_2 cans now appears to be self-multiplication.

The need for self-multiplication corrections in metallic plutonium is obvious, as is the need for induced-fission corrections in oxides, fluorides, etc. The need for a substantial self-multiplication correction to PuO_2 , however, has been concealed by compensating electronic deadtime effects. To minimize deadtime and isolate multiplication, we used the new shift register to measure a series of PuO_2 standards ranging in size from 20 g (6% ^{240}Pu) to 780 g (10% ^{240}Pu). We also used the new dual range coincidence counter (described in Sec. 1-II-C) in the low-efficiency mode (5.7% efficiency, 16 μs die-away time). Four fast (0.2- μs time constant) bipolar amplifiers were used in conjunction with four leading-edge discriminators and a shift register pre-delay of 6 μs . Under these conditions amplifier pulse pileup effects were negligible, and deadtime corrections for coincidence and total events were

$$C = C_{me}^{0.0028T}, T(\text{kc}) = T_{me}^{0.0014T}.$$

These corrections were on the order of 2%. Errors due to statistics, deadtime corrections, detector height response, and uncertainty in the mass of the standards were estimated to be $\leq 1\%$. The corrected data, illustrated in Fig. 5 and given in Table IV, show a significant upward curvature.

Self-multiplication in the PuO_2 cans was calculated with the two-dimensional transport code TWOTRAN⁸ with S_4 angular segmentation and Hansen-Roach 16-group neutron cross sections.⁹ The calculations assumed that the cans were bare (unreflected). Future calculations will account for the effects of the cadmium-lined polyethylene detector. The calculated leakage multiplication (number of leakage neutrons per source neutron) is given in Table IV. Several of the transport calculations were checked with the neutron Monte Carlo code MCN.¹⁰ Agreement between the two methods was very good.

To account accurately for the effects of multiplication on coincidence counts, the discrete leakage multiplication (probability of 1, 2, 3, etc., neutrons leaking per source neutron) should be used rather than the average multiplication calculated here. The MCN code will be revised to yield this information. For the present we will estimate coincident multiplication as follows:

Fig. 5.

Self-multiplication in PuO_2 . Measurement errors are about equal to the size of the circles. Deviation of the data from the straight line is believed due to self-multiplication.

(1) The coincident count rate is

$$C \propto \frac{\epsilon^2 \overline{\nu(\nu-1)}}{2!} - \frac{\epsilon^3 \overline{\nu(\nu-1)(\nu-2)}}{3!}$$

where ϵ is the detection efficiency and ν the multiplicity.

(2) The distribution of actual neutron numbers emitted during spontaneous fission can be included

TABLE IV
SELF-MULTIPLICATION IN PuO₂

Total Plutonium (g)	²⁴⁰ Pu (g)	Assumed Plutonium Density (g/cm ³)	Fill Height (cm)	Coincident Counts g ²⁴⁰ Pu-s	Leakage Multiplication, M	Coincident Multiplication, C	Coincident Counts $\times \frac{1}{C}$ g ²⁴⁰ Pu-s
20	1.22	---	---	2.35 ± 0.02	---	---	2.34 ^a
60	3.84	0.15	7.4	2.42 ± 0.02	1.0048	1.012	2.39
120	7.67	0.30	7.4	2.53 ± 0.02	1.0101	1.026	2.46
480	37.40	1.29	6.8	2.99 ± 0.03	1.0459	1.119	2.67
459	43.38	1.29	6.5	2.98 ± 0.03	1.0449	1.116	2.67
556	54.92	1.29	7.9	3.03 ± 0.03	1.0482	1.125	2.69
615	64.96	1.29	8.7	3.08 ± 0.03	1.0499	1.130	2.73
779	81.34	1.29	11.0	3.26 ± 0.03	1.0542	1.141	2.86

^aEstimated.

by summing over the discrete probabilities P(n), as determined experimentally, or by using a gaussian distribution

$$P(\nu) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{-(\nu-\bar{\nu})^2/2\sigma^2}$$

where $\bar{\nu} = 2.17$ for ²⁴⁰Pu, and $\sigma = 1.08$ as recommended by Terrell.¹¹ Both methods yield similar results here. If we fold in the gaussian distribution,

$$C \propto \frac{\epsilon^2}{2} [\sigma^2 + \bar{\nu}(\bar{\nu} - 1) - \epsilon\sigma^2(\bar{\nu} - 1) - \frac{\epsilon}{3} \bar{\nu}(\bar{\nu} - 1)(\bar{\nu} - 2)]$$

through second order.

(3) Self-multiplication inside the can takes place on a time scale small compared to the die-away time of the detector. Thus a leakage multiplication M implies a new average multiplicity of M $\bar{\nu}$. (This cannot hold for induced fissions from (α ,n) reactions, because there is no time correlation between the induced and spontaneous fissions.) Then

$$C \propto \frac{\epsilon^2}{2} [M^2\sigma^2 + M\bar{\nu}(M\bar{\nu} - 1) - \epsilon M^2\sigma^2(M\bar{\nu} - 1) - \frac{\epsilon}{3} M\bar{\nu}(M\bar{\nu} - 1)(M\bar{\nu} - 2)]$$

This coincidence multiplication is much greater than the leakage multiplication, as indicated in Table IV.

The correction for self-multiplication is appreciable despite the low density of the PuO₂ (about 1.3 g/cm³). However, it is clear both from Fig. 5 and the last column of corrected coincident counts/g-s in Table IV that we have removed only about half of the curvature in the data. To increase our understanding of this problem, we will investigate (α ,n) reactions, reflection by the well-counter, and an improved handling of coincidence multiplication.

C. Dual Range Thermal-Neutron Coincidence Counter (M. L. Evans, H. O. Menlove, and L. G. Speir)

For many applications of neutron well coincidence counters, it is desirable to assay samples that cover a

mass range from <1 g to >2 kg of PuO_2 . To achieve this wide-range capability, the dual range coincidence counter has been designed and fabricated. The dual range capability is achieved by having two removable cadmium sleeves near the ^3He detectors. These sleeves can be inserted for low-efficiency operation with short die-away times and removed for high-efficiency counting with a long die-away time.

The geometry of the counter is shown in Fig. 6. The cadmium sleeves on both sides of the middle polyethylene cylinder (ring 2) are removable. The detector consists of 20 ^3He tubes of 2.54-cm diam filled to a pressure of 18 atm. The inner and outer CH_2 cylinders (rings 1 and 3, respectively) are each 3.0 cm thick. The cadmium sleeve (1.0 mm thick) on the inside of the well stops low-energy neutrons from returning to the sample position, thereby minimizing criticality problems and reducing multiplication for high-mass loadings. The outer cadmium sleeve improves the effectiveness of the exterior CH_2 shield.

Thus, there are two modes of operation: (1) the low-mass range with both removable cadmium sleeves removed for maximum efficiency and (2) the high-mass range with all the cadmium sleeves in place to give a short die-away time and correspondingly short electronic gate width in the coincidence circuitry. For operational mode (1), the singles efficiency is 20-25% and the neutron die-away time $50\ \mu\text{s}$. For operational mode (2) the efficiency decreases to 6 to 10% and the die-away time goes to 15 to 25 μs .

Monte Carlo studies were performed to determine the physical dimensions of the counter that would yield the desired efficiency, die-away time, matrix material insensitivity, and optimum values for other parameters. The efficiency is especially dependent on the thicknesses of the polyethylene moderator and reflector. In the Monte Carlo program the thicknesses of rings 1 and 3 were taken to be equal and were varied simultaneously to determine the optimum thickness. The results of this study are shown in Fig. 7 for both configurations (cadmium sleeves in and out). The results indicate that a moderator (and reflector) thickness of about 3.5 cm combines a high efficiency with relative insensitivity to matrix effects for both configurations. At this design thickness, the equivalent hydrogen of about 1-cm thickness of CH_2 can be added to the sample matrix with little change to the counting efficiency as can be seen in Fig. 7. The calculated efficiencies

Fig. 6.
Dual range neutron coincidence counter for the assay of plutonium samples in the mass range 1-2000 g.

Fig. 7.

Design data for the dual range counter showing efficiency and die-away time as a function of polyethylene moderator (and reflector) thickness as determined from Monte Carlo studies.

of the counter shown in Fig. 6 were about 25% in the high-efficiency configuration and ~7% in the low-efficiency configuration. The corresponding moderator die-away times were approximately 45 and 11 μ s. These calculated design parameters agree well with the preliminary measured values.

D. Portable Neutron Assay System for Light Water Reactor (LWR) Fuel Assemblies (J. D. Brandenberger, H. O. Menlove, and E. G. Medina)

During the current reporting period, work continued on development of the active interrogation portable assay assembly for pressurized water reac-

tor (PWR) fuel assemblies. These assemblies are large arrays of as many as 16 by 16 fuel elements. Hence, lack of penetration by the low-energy interrogating neutrons makes it difficult to obtain useful information in the interior of the fuel assemblies.

The geometry of the assay system described in Ref. 2 was used with a 15- by 9-rod fuel assembly and provided useful assay information in all regions (1-12) shown in Fig. 8. Additional fuel rods were used to construct a 15- by 15-rod fuel assembly having only a few missing rods at its back. Under these conditions, the system showed saturation effects in the sense that if, for example, fuel rods in region 10 were removed, the previously detected fission neutrons from this region were essentially compensated for by additional fission neutrons from deeper

Fig. 8.

PWR fuel assembly assay system. Small circles represent individual fuel rods in part of a 15 x 15 rod assembly. Circles with an X are possible locations for control rods.

rows. The regions farther from the Am-Li source had not previously been filled.

To gain sensitivity for regions in the interior and far from the source, the ^4He fission-neutron detectors required some directional sensitivity. Of several geometries tried, we adopted the one consisting of four 2.54-cm-diam ^4He detectors that viewed the assembly through 1.3- by 28-cm slits in the polyethylene arms (Fig. 8). The results of the initial sensitivity tests are shown in Table V. The central regions have a sensitivity of 4 to 6% for replacement of fuel rods by iron rods.

Because of the partial shielding of the detectors, the effective efficiency is decreased and it takes longer to obtain good counting statistics. The sensitivity of the assay system might be improved by using improved geometry or more efficient neutron detectors.

The design of portable electronics that can be stabilized over long periods or quickly calibrated remains to be done.

E. Upgrading of the Segmented Gamma Scanner Software to Measure ^{241}Pu and ^{239}Pu Simultaneously (E. R. Martin)

The assembly language codes that operate the SGS were modified to permit the simultaneous measurement of ^{241}Pu and ^{239}Pu . For routine plutonium fissile assays, ^{76}Se , which has a gamma peak at 400 keV, is usually used as the transmission source to measure the transmission at the ^{239}Pu 414-keV energy. The selenium also has a reasonably intense gamma line at 265 keV, which is used to measure the transmission at the ^{241}Pu 208-keV gamma energy. A μ -ratio correction must be applied because these two energies occur at a point where the mass attenuation coefficients are changing rapidly. However, if the attenuation coefficients for the matrix material are known approximately, a μ -ratio correction can be entered to give reasonably good values for the extrapolated transmission. In most common isotopic concentrations for plutonium, the ^{239}Pu is more prevalent by several orders of magnitude than ^{241}Pu , but the intensity of the 208-keV gamma from ^{241}Pu is greater than the ^{239}Pu 414-keV intensity by several orders of magnitude. Hence the simultaneous measurement of these two isotopes is quite practicable. The attenuation problems for the 208-keV energy are much more severe than at the higher ^{239}Pu energy; therefore, there will be many cases in which a quite reasonable measurement of ^{239}Pu can be made yet no measurement of ^{241}Pu is possible. For most actual applications, both isotopes can be measured.

In changing the computer codes, provision is now made for entering three, four, or five assay peaks. The computer assumes the three-peak assay (livetime peak, fissile peak, and transmission peak) to be either ^{239}Pu or ^{238}Pu , and the livetime peak energy to be the 356-keV peak from ^{139}Ba . The four-peak assay is assumed to be ^{235}U with the livetime peak energy at 122 keV from ^{57}Co . The five-peak assay is for ^{239}Pu and ^{241}Pu as described. In each case, since the livetime peak energy is known, the computer automatically makes a detector resolution

TABLE V

**SENSITIVITY TEST RESULTS OF PORTABLE PWR ASSAY SYSTEM
FOR A 15- BY 15-ROD FUEL ASSEMBLY**

Region ^a	Decrease in Counts per Rod per 2000 s ^b	Sensitivity at the 2 σ Level	
		Rods Exchanged for Detection	Rods Exchanged (%)
1	405	1	0.5
2	151	2	0.5
3	205	1	0.5
4	72	4	1.8
5	87	3	1.3
6	245	1	0.5
7	20	13	5.8
8	50	5	2.2
9	91	3	1.3
10	28	10	4.4
11	23	12	5.3
12	78	3	1.3

^aRegion in which steel rods have been substituted for fuel rods.

^bThe standard deviation of the measurements is 200 to 260 counts per rod per 2000 s.

check at the livetime peak energy, thus providing a powerful diagnostic tool for system operation.

The software now further provides a multiple pass option by which calibration runs can be made automatically without operator intervention until a required preset accuracy is achieved, or sample unknowns can be run to required accuracy.

The present software will run ²³⁵U, ²³⁹Pu, ²⁴¹Pu, ²⁴¹Pu on either the small sample table or the barrel handler with automatic resolution checking of the detector and an automatic two-pass assay based on background statistics.

F. Evaluation of the IAEA GAMMA NP361 Five-Channel Analyzer (J. L. Parker)

At the request of IAEA personnel, we have evaluated the performance and reliability of the IAEA GAMMA NP361 portable gamma-ray assay instrument. This small, truly portable instrument employs five preprogrammed single-channel analyzer windows to extract information from the spectrum generated by an intrinsic germanium

detector. The necessary study and work were done during October and November 1976 and the results have been reported to the IAEA authorities.

G. Prototype for Neutron Interrogation of 55-gal Barrels (T. W. Crane)

The components have been fabricated for the lead-lined 55-gal barrel assay chamber addition to the Shuffler system (see Ref. 2, p. 2). The prototype unit is now being assembled on the second level of the safeguards building (TSL27) behind the hot cell test area.

A schematic of the assay chamber is shown in Fig. 9. Monte Carlo neutron transport codes were used to investigate the expected behavior of the system. The first problem considered was to estimate the approximate number of delayed neutrons to be counted as a function of the strength of the ²⁵²Cf interrogation source. This problem can be separated into three independent steps: (1) estimate the number of fissions in the barrel per source neutron, (2) calculate the number of delayed neutrons emitted during the

Fig. 9.
Schematic of the 55-gal assay chamber for the Shuffler system. Numbers shown by tubes give contribution to response $\pm 25\%$.

count cycle per fission, and (3) estimate the detection probability per delayed neutron. The delayed-neutron count per source neutron is roughly the product of the above steps.

To estimate the source coupling to the barrel, the chosen matrix material was carbon ($\rho = 1.0 \text{ g/cm}^3$) with $10 \text{ g } ^{239}\text{Pu}$ uniformly interspersed. For this idealized situation the transport code estimates 2.36×10^{-4} fissions per gram ^{239}Pu per source neutron. An additional output of the code is the spatial variation of the fission rate within the barrel. Table VI gives the fission response at various longitudinal and

radial positions with the angular response averaged out by rotation of the barrel. Without rotation the maximum variation increases by a factor of 4.

The next step is to estimate the number of delayed neutrons emitted per fission during the counting period of the Shuffler data acquisition cycle. To reduce the number of parameters in this calculation we chose the experimentally determined optimum cycle of 4s for both the irradiation and count times (see Ref. 12, p. 23). Also we assume the data are taken with a large number of cycles so that the equilibrium condition dominates. Thus, the fraction of neutrons emitted during the counting time for a given delayed neutron precursor is:

$$F_c = \frac{(1 - e^{-\lambda t_1})(1 - e^{-\lambda t_2}) e^{-\lambda t_3}}{\lambda t_1 (1 - e^{-\lambda T})}$$

where

λ = decay rate, ($\tau_{1/2} = \ln 2/\lambda$)

t_1 = irradiation time

t_2 = count time

t_3 = source transfer time

T = total cycle time ($t_1 + t_2 + t_3$).

The source transfer time is 0.45 s for the Shuffler system, and for simplicity in the calculation it is assumed that the interrogation source is off during the transfer time. Experimental data for ^{239}Pu delayed-neutron emission are reproduced in Table VII (see Ref. 13, p. 203) to facilitate the calculation of the expected rate. Thus, we estimate 2.5×10^{-3} delayed neutrons per ^{239}Pu fission.

Finally, the neutron detection efficiency was estimated by the transport codes. A uniform delayed-neutron emission source was used in the simulation. This assumption yields results only slightly different from those using the calculated distribution presented in Table VI. The total neutron detection efficiency was calculated at $9.3 \pm 0.4\%$. The detection efficiencies for individual tubes (relative error $\approx 25\%$) are indicated in Fig. 9. Note that the tubes near the corners are inefficient.

The approximate delayed-neutron counting rate is $5.55 \times 10^{-6}/\text{g-}^{239}\text{Pu}$ per interrogation neutron. Although this result is for an idealized case, we will use it to estimate an approximate sensitivity for the assay of material in 55-gal barrels. The background

TABLE VI

AVERAGE NUMBER OF ^{239}Pu FISSIONS/ $\text{cm}^3/10^9$
 SOURCE NEUTRONS PER GRAM OF ^{239}Pu UNIFORMLY
 INTERSPERSED WITHIN THE BARREL

	Center $0 \leq r \leq$ <u>10.16 cm</u>	$10.16 < r \leq$ <u>20.32 cm</u>	Outer Surface $20.32 < r \leq$ <u>30.48 cm</u>
Midplane			
$0 \leq z \leq$ 15.24 cm	1.39 ± 0.25	1.30 ± 0.09	1.18 ± 0.07
$15.24 < z \leq$ 30.48 cm	1.30 ± 0.24	1.14 ± 0.09	0.87 ± 0.05
$30.48 < z \leq$ 45.72 cm	0.45 ± 0.11	0.48 ± 0.05	0.38 ± 0.03
Top or bottom			

TABLE VII

DATA FOR CALCULATING THE NUMBER OF
 DELAYED NEUTRONS PER ^{239}Pu FISSION
 DURING THE SHUFFLER COUNTING TIME

Group Index	$\tau_{1/2}$ (s)	Neutrons/ Fission	Fraction Countable (F_c)	Countable Neutrons/ Fission
1	53.7 ± 3.6	0.25×10^{-3}	0.449	1.12×10^{-4}
2	22.9 ± 1.1	1.78×10^{-3}	0.449	7.11×10^{-4}
3	6.11 ± 0.24	1.29×10^{-3}	0.438	5.65×10^{-4}
4	2.14 ± 0.06	2.53×10^{-3}	0.373	9.44×10^{-4}
5	0.40 ± 0.03	0.80×10^{-3}	0.066	0.53×10^{-4}
6	0.15 ± 0.05	0.05×10^{-3}	0.007	3.5×10^{-7}
TOTALS	---	6.7×10^{-3}	---	2.5×10^{-3}

rate from neutron leakage from the ^{252}Cf source in the "off" position and from the neutron-producing cosmic-ray interactions in the lead shielding is estimated at 50 counts/s by extrapolating from existing small systems. For plutonium in the oxide form (PuO_2), and of an isotopic mixture similar to that expected in reprocessed fuels (say, 80% ^{239}Pu and 20% ^{240}Pu), the background from spontaneous fission and (α, n) reactions is expected to be about 36 counts/g-Pu/s. The ^{252}Cf source in the Shuffler system is now about 200 μg , thus the delayed-neutron counting should be about 22 counts/g-Pu/s during the count cycle of the data acquisition. When the prototype system is completed, these rates will be known more accurately. If we use a data collection cycle consisting of an initial background count followed by interrogation and delayed-neutron counting, the sensitivity for the 1000-s combination is 80 mg ^{239}Pu at the 3σ level. This result should be considered only a rough estimate, because neutron poisons in the matrix material as well as the neutron background rates will be crucial in determining the sensitivity of actual measurements.

H. Calculational Support for the Assay of Fast Flux Test Facility (FFTF) Fuel Bundles (D. A. Close and M. S. Krick)

The Hanford Engineering Development Laboratory (HEDL) is interested in the development of an instrument that can assay FFTF fuel bundles. Such an instrument would be used for verification and quality assurance measurements. The instrument should give the total fissile content and possibly the isotopic content. Initial Monte Carlo calculations have been performed on possible assay systems.

The FFTF fuel is a fluorine-free $\text{UO}_2\text{-PuO}_2$ mixture. The uranium is natural uranium. Table VIII shows a typical FFTF fuel composition used in the Monte Carlo calculations. Most FFTF fuel also has some ^{238}Pu and ^{242}Pu but the atom fraction of ^{238}Pu was so small (less than 6×10^{-6}) that it was ignored, and ^{242}Pu was ignored because it is not in the Monte Carlo cross-section library. The total mass density of the homogenized FFTF fuel bundle is 2.4 g/cm^3 .

A top view of the calculational model is shown in Fig. 10. Cylindrical geometry was used; the height of

TABLE VIII

TYPICAL FFTF FUEL COMPOSITION USED IN NEUTRONIC CALCULATIONS

Isotope	Weight (%)	Density (g/cm^3)
^{16}O	11.85	0.28
^{235}U	0.48	0.01
^{238}U	64.78	1.56
^{239}Pu	19.79	0.48
^{240}Pu	2.68	0.06
^{241}Pu	0.42	0.01

- ZONE 1 FUEL, 3.81-cm RADIUS
- ZONE 2 FUEL, 3.81 cm THICK
- ZONE 3 CADMIUM, 0.1 cm THICK
- ZONE 4 MODERATOR/ REFLECTOR, 5.08 cm THICK
- ZONE 5 POLYETHYLENE, 7.62 cm THICK
- ZONE 6 5% BORATED POLYETHYLENE, 15.24 cm THICK
- ZONE 7 ^3He , 2.54-cm diam AT 4-atm PRESSURE

Fig. 10.

Top view of model used in the Monte Carlo neutronic calculations.

the model was 91.44 cm. The fuel region was divided into two regions, the volume ratio being 1:3. The FFTF fuel bundle has a hexagonal cross section approximately 15.24 cm across the flats. The diameter of the fuel region was taken to be 15.24 cm. The fuel was surrounded by a 0.1-cm-thick cadmium sleeve. Next there was a 5.08-cm-thick moderator/reflector annulus. Surrounding this moderator/reflector annulus was a 7.62-cm-thick annulus of polyethylene. Twelve ^3He tubes (2.54-cm diam, 4 atm, 91.44 cm long) were assumed in the center of this polyethylene annulus. Finally, there was a 15.24-cm-thick annulus of 5% borated polyethylene.

The first series of calculations assumed different moderators/reflectors: vacuum, polyethylene, 5% borated polyethylene, 30% borated polyethylene, beryllium, iron, heavy water, and graphite. The multiplication of the system, the total efficiency for all 12 ^3He tubes, the total number of fissions caused by the incident neutrons normalized to the number of source neutrons, and the ratio of the average fissions per unit volume in the inner fuel region to the

average fissions per unit volume in the outer fuel region were calculated for each of the moderator/reflector materials. This set of calculations is summarized in Table IX, columns 2-5. The incident-neutron energy spectrum was a ^{252}Cf fission-neutron energy spectrum uniformly distributed throughout the volume of the fuel and thus represented a passive-neutron measurement.

The beryllium moderator/reflector yields the highest multiplication because of the $^9\text{Be}(n,2n)^8\text{Be}$ reaction. The fission-neutron energy spectrum incident on beryllium has a multiplication of 1.134 when there is no fuel in the system. The systems having borated polyethylene moderators/reflectors have a lower ^3He efficiency because of the capture of slow neutrons by boron. For these eight different moderators/reflectors, the number of fissions is rather uniform throughout the fuel volume, indicating good penetrability by the fission neutrons. On the basis of calculations for these eight cases, beryllium seems to be the best moderator/reflector.

TABLE IX

MODERATOR/REFLECTOR SENSITIVITY CALCULATIONS FOR AN FFTF
FUEL BUNDLE ASSAY SYSTEM ASSUMING PASSIVE NEUTRONS

Moderator/ Reflector	Multiplication	Total ^3He Efficiency	Total Number of Fissions/ Source Neutron	$\frac{(\text{Fissions/Volume})_{\text{inner}}}{(\text{Fissions/Volume})_{\text{outer}}}$
Vacuum	1.126	0.141	0.059	1.059
Polyethylene	1.150	0.130	0.075	1.025
5% borated polyethylene	1.148	0.068	0.071	1.013
30% borated polyethylene	1.138	0.047	0.065	1.102
Beryllium	1.354	0.226	0.108	0.981
Iron	1.158	0.166	0.070	1.086
Heavy water	1.208	0.186	0.093	0.985
Graphite	1.178	0.180	0.085	1.024

The next series of calculations involved the effect of the thickness of the moderator/reflector on the multiplication of the system, the total efficiency of the 12 ^3He tubes, and the ratio of the number of fissions per volume in the inner fuel region to the number of fissions per volume in the outer fuel region. Three moderators/reflectors were considered here: beryllium, heavy water, graphite. The neutron energy spectrum used here was the same as that described previously.

Figure 11 shows the effect of the thickness of the beryllium moderator/reflector on the above-mentioned three quantities. Because of the $^9\text{Be}(n,2n)^8\text{Be}$ reaction, the multiplication of the system when no fuel is present is also shown in Fig. 11(a). Figure 11(b) shows the variation of ^3He efficiency with beryllium thickness. The variation of the inner volume to outer volume fission ratio as a

function of the beryllium thickness is shown in Fig. 11(c).

Figure 12(a) shows the variation of the multiplication of the system as a function of the heavy water moderator/reflector thickness. When no fuel is present, the multiplication is unity for all thicknesses. Figures 12(b) and 12(c) present the ^3He efficiency variation and the inner volume to outer volume fission ratio variation, respectively.

Figure 13(a) shows the variation of the multiplication of the system as a function of the thickness of the graphite moderator/reflector. When no fuel is present, the multiplication is unity for all thicknesses. Figures 13(b) and 13(a) present the ^3He efficiency variation and the inner volume to outer volume fission ratio variation, respectively, for four different thicknesses of graphite.

Fig. 11.

Effect of thickness of the beryllium moderator/reflector on (a) the multiplication of the FFTF fuel bundle assay system, (b) the total efficiency of the 12 ^3He tubes, (c) the inner volume to outer volume fission ratio. The smooth curves serve merely as a guide for the eye.

Fig. 12.

Effect of thickness of the heavy water moderator/reflector on (a) the multiplication of the FFTF fuel bundle assay system, (b) the total efficiency of the 12 ^3He tubes, (c) the inner volume to outer volume fission ratio. The smooth curves serve merely as a guide for the eye.

Fig. 13.

Effect of thickness of the graphite moderator/reflector on (a) the multiplication of the FFTF fuel bundle assay system, (b) the total efficiency of the ^{12}He tubes, (c) the inner volume to outer volume fission ratio. The smooth curves serve merely as a guide for the eye.

Finally, a series of fuel sensitivity calculations was performed. For these calculations, the moderator/reflector thickness was held constant at 5.08 cm. The fuel sensitivity was determined by replacing each uranium and plutonium isotope atom-for-atom with lead. These calculations are summarized in Tables X-XIII.

Tables X and XI assumed the passive neutron system described above. Table X presents the results for a beryllium moderator/reflector; Table XI lists the results for a heavy water moderator/reflector. Both of these passive neutron assay systems show good penetrability of the fuel. The systems appear to be sensitive to the absence of ^{239}Pu , and there is some sensitivity to the absence of ^{238}U . The other isotopes, ^{235}U , ^{240}Pu , and ^{241}Pu , have little effect on

the response of the system because they are present in such small quantities.

In addition to the above passive neutron assay systems, two active neutron systems were mocked up. The results of a ^{124}Sb -Be photoneutron assay system are presented in Table XII. A cylindrical ring neutron source having the neutron energy spectrum of ^{124}Sb -Be was positioned in the middle of the 5.08-cm-thick beryllium moderator/reflector.

The second active neutron system was a ^{241}Am -Li (α, n) neutron source (see Table XIII). There was no moderator/reflector material in this case, but the 5.08-cm thickness was preserved. The neutron energy spectrum of ^{241}Am -Li was assumed to be a cylindrical ring source located in the middle of the moderator/reflector region.

These active neutron systems are also very sensitive to the absence of ^{239}Pu . The ^{124}Sb -Be system is sensitive to the absence of ^{238}U . The total ^3He efficiency of the ^{241}Am -Li system is sensitive to the absence of ^{238}U ; the total number of fissions in the ^{241}Am -Li system is not sensitive to the absence of ^{238}U . The responses of these two active systems are not sensitive to the other uranium and plutonium isotopes. The neutrons from the ^{124}Sb -Be and ^{241}Am -Li sources, being less energetic than fission neutrons, do not penetrate the fuel as well. It remains to be seen whether this is critical.

Further calculations will investigate other hypothetical neutron assay systems, namely ^{124}Sb -Be and ^{241}Am -Li neutron sources and ^3He tubes.

I. Photoneutron Assay of Bulk PuO_2 Samples (M. S. Krick and H. O. Menlove)

Photoneutron techniques are being considered for the assay of full FFTF fuel assemblies, which contain approximately 8 kg of fissile plutonium and uranium per assembly. An experimental photoneutron assay system was assembled previously² to study assay techniques for intermediate-size enriched uranium samples (1 to 60 g ^{235}U). For the study of larger mass plutonium samples, four PuO_2 standards were prepared from commercial-grade PuO_2 by LASL Group CMB-11.

The PuO_2 standards with masses ranging from 531 to 900 g are oxide powders in 8.6-cm-diam by 11.4-cm-high metal cans. The secondary container is a

TABLE X
**FUEL SENSITIVITY CALCULATIONS FOR AN FFTF FUEL
BUNDLE ASSAY SYSTEM ASSUMING PASSIVE NEUTRONS
AND A BERYLLIUM MODERATOR/REFLECTOR**

<u>Fuel Composition</u>	<u>Multi- plication</u>	<u>Total ³He Efficiency</u>	<u>Total Number of Fissions/ Source Neutron</u>	<u>(Fissions/Volume)_{inner} (Fissions/Volume)_{outer}</u>
Normal fuel	1.355	0.226	0.108	0.981
²³⁵ U replaced with Pb	1.347	0.223	0.106	0.964
²³⁸ U replaced with Pb	1.315	0.223	0.098	0.931
²³⁹ Pu replaced with Pb	1.175	0.191	0.025	1.128
²⁴⁰ Pu replaced with Pb	1.349	0.221	0.107	0.971
²⁴¹ Pu replaced with Pb	1.346	0.224	0.106	0.982

TABLE XI
**FUEL SENSITIVITY CALCULATIONS FOR AN FFTF FUEL
BUNDLE ASSAY SYSTEM ASSUMING PASSIVE NEUTRONS AND A
HEAVY WATER MODERATOR/REFLECTOR**

<u>Fuel Composition</u>	<u>Multi- plication</u>	<u>Total ³He Efficiency</u>	<u>Total Number of Fissions/ Source Neutron</u>	<u>(Fissions/Volume)_{inner} (Fissions/Volume)_{outer}</u>
Normal fuel	1.208	0.186	0.093	0.985
²³⁵ U replaced with Pb	1.204	0.189	0.092	0.995
²³⁸ U replaced with Pb	1.152	0.185	0.079	0.914
²³⁹ Pu replaced with Pb	1.061	0.167	0.022	1.141
²⁴⁰ Pu replaced with Pb	1.212	0.195	0.094	0.958
²⁴¹ Pu replaced with Pb	1.202	0.190	0.091	0.958

TABLE XII
FUEL SENSITIVITY CALCULATIONS FOR AN FFTF FUEL BUNDLE
ASSAY SYSTEM ASSUMING A ^{124}Sb -Be NEUTRON SOURCE

<u>Fuel Composition</u>	<u>Multi- plication</u>	<u>Total ^3He Efficiency</u>	<u>Total Number of Fissions/ Source Neutron</u>	<u>(Fissions/Volume)_{inner} (Fissions/Volume)_{outer}</u>
No fuel	1.000	0.224		
Normal fuel	1.182	0.241	0.085	0.793
^{235}U replaced with Pb	1.178	0.240	0.082	0.794
^{238}U replaced with Pb	1.207	0.259	0.103	0.840
^{239}Pu replaced with Pb	1.018	0.213	0.010	0.923
^{240}Pu replaced with Pb	1.191	0.248	0.090	0.812
^{241}Pu replaced with Pb	1.174	0.240	0.081	0.795

TABLE XIII
FUEL SENSITIVITY CALCULATIONS FOR AN FFTF FUEL BUNDLE
ASSAY SYSTEM ASSUMING A ^{241}Am -Li NEUTRON SOURCE

<u>Fuel Composition</u>	<u>Multi- plication</u>	<u>Total ^3He Efficiency</u>	<u>Total Number of Fissions/ Source Neutron</u>	<u>(Fissions/Volume)_{inner} (Fissions/Volume)_{outer}</u>
No fuel	1.000	0.167		
Normal fuel	1.072	0.174	0.038	0.787
^{235}U replaced with Pb	1.072	0.174	0.037	0.784
^{238}U replaced with Pb	1.078	0.184	0.039	0.840
^{239}Pu replaced with Pb	1.007	0.171	0.004	0.932
^{240}Pu replaced with Pb	1.074	0.176	0.040	0.798
^{241}Pu replaced with Pb	1.068	0.173	0.036	0.773

10.8-cm-diam by 12.4-cm-high metal can. The primary container is centered radially and rests on the bottom of the secondary container; the volume between the containers is filled with MgO powder. Package data are shown in Table XIV; mass spectrometric data are shown in Table XV.

The four PuO₂ samples were measured with the experimental photoneutron assay system described previously.² Unlike the previous measurements on the uranium samples, measurements on the PuO₂ samples were made with a lead cover over most of the assembly to improve the spatial distribution of the source neutrons. The PuO₂ samples were also measured without the cover for comparison. The

response of the system per second per gram of fissile plutonium is plotted against fissile plutonium mass in Fig. 14 for measurements made with and without the lead cover. No flux monitor was used to correct for flux depression. Not surprisingly, the responses of the covered system averaged 44% higher than those of the uncovered system. Surprisingly, the covered system was not appreciably more uniform in response than the uncovered one. The response per gram of the 900-g sample was 13% lower than that of the 531-g sample for the covered assembly; the corresponding decrease for the uncovered assembly was 17%.

TABLE XIV
PACKAGE DATA FOR FOUR PuO₂ STANDARDS

<u>Standard Identification</u>	<u>Oxide Mass (g)</u>	<u>Pu Mass (g)</u>	<u>PuO₂ Height (cm)</u>	<u>Total Package Mass (g)</u>
PEO-385	531	459	6.0	1628
PEO-382	655	556	7.9	1779
PEO-381	720	615	9.2	1833
PEO-447	900	779	11.0	2011

TABLE XV
MASS SPECTROMETRIC DATA FOR FOUR PuO₂ STANDARDS

<u>Standard Identification</u>	<u>Isotopic Abundance (%)</u>				
	<u>²³⁸Pu</u>	<u>²³⁹Pu</u>	<u>²⁴⁰Pu</u>	<u>²⁴¹Pu</u>	<u>²⁴²Pu</u>
PEO-385	0.030	89.96	9.22	0.697	0.093
PEO-382	0.030	89.42	9.62	0.825	0.109
PEO-381	0.038	88.59	10.27	0.981	0.118
PEO-447	0.045	88.75	10.07	0.981	0.155

Fig. 14.

Response of photoneutron assay system (counts/s-g) vs the fissile plutonium mass with covered and uncovered assemblies.

From these measurements we estimate that an FFTF fuel assembly in a cylindrically symmetric photoneutron assay system using 20 biased ^4He proportional counters (5-cm diam, 61 cm long, 18 atm) for fast-neutron detection would produce a count rate in excess of 10^4 counts/s for a 1-Ci ^{124}Sb -Be source or its equivalent. Approximately one-fourth of the counts would be due to spontaneous emission from the fuel assembly and would be measured separately from ^{124}Sb -Be neutron interrogation.

J. Bremsstrahlung Photoneutron Sources (M. S. Krick)

Nondestructive assay systems based on photoneutron interrogation have been investigated by LASL's Nuclear Safeguards Research Laboratory for many years. One system was designed to assay spent Rover fuel elements (Ref. 14, p. 8) and another was designed to assay cold or spent LWR fuel rods (Ref. 15, p. 16). Other photoneutron systems are under consideration for full FFTF fuel assembly assay and for bulk sample uranium and plutonium assay.

Generally, the $^9\text{Be}(\gamma, n)$ and $^2\text{H}(\gamma, n)$ reactions are used for photoneutron sources because the small, negative Q-values (-1.67 and -2.23 MeV, respectively) permit the use of isotopic gamma-ray sources such as ^{124}Sb and ^{228}Th . Photoneutrons should have

energies high enough to allow good sample penetration (the photoneutrons are usually not thermalized), yet low enough so that (1) fertile isotopes are not fissioned and (2) the photoneutrons are easily separated from the prompt fission neutrons by energy discrimination.

The photon source ideally would produce monoenergetic photons with an appropriate energy and would have a conveniently long half-life. Unfortunately, no isotopic photon source is close to the ideal. All have at least one of the following deficiencies: (1) the half-life is too short, (2) the neutron energies are too widely spread, (3) the neutron yield per source photon is too low, (4) the cost per neutron is too high.

Antimony-124, which is used with the LWR fuel rod assay system (Ref. 15, p. 16), is an excellent source except for its short half-life of 60.2 days. Radium-226, which was used with the Rover fuel rod assay system (Ref. 24, p. 8) has a long half-life (1620 yr), but is not as good as ^{124}Sb in all other respects. Some promising photoneutron sources such as accelerator-produced ^{88}Y (107-day half-life) and fission-product ^{144}Ce (285-day half-life) are not yet available in sufficient quantities for practical applications. Table XVI summarizes the qualities of some photoneutron sources.

Bremsstrahlung sources are also being considered for photoneutron production. An isotope that beta-decays with a Q-value above a photoneutron threshold can produce photoneutrons via $(h\nu, n)$ reactions from bremsstrahlung photons ($h\nu$). The neutron yield per beta decay is very low because (1) only a small fraction of the beta particles have energies above the photoneutron threshold and (2) only a small fraction of these energetic beta particles will produce a photon above the photoneutron threshold. The thick-target bremsstrahlung energy distribution for monoenergetic electrons is¹⁶

$$\frac{dE}{d\nu} \cong c [Z(\nu_m - \nu) + 0.0025 Z^2],$$

where

$dE/d\nu$ is the bremsstrahlung energy per unit frequency,
 Z is the atomic number of the target,
 ν_m is the maximum photon frequency, and
 c is independent of Z and ν .

TABLE XVI

ESTIMATED QUALITIES OF SOME PHOTONEUTRON
SOURCES USING BERYLLIUM

Photoneutron Source	Half- Life	Neutron Energies	Neutron/ Photon Ratio	Cost Per Neutron
^{88}Y	P	E	E	U
^{90}Sr (bremsstrahlung)	E	G	P	F(U)
^{124}Sb	P	G	E	E
^{144}Ce	F	G	F	U
^{228}Th	G	G	F	P(U)
^{226}Ra	E	F	F	F(U)

E excellent

G good

F fair

P poor

U unknown or uncertain in the quantities and configurations required for practical assay systems.

Except for a small contribution near ν_m , $dE/d\nu$ is proportional to Z . Therefore, the photoneutron yield per curie from a bremsstrahlung source can be increased by mixing the radioactive isotope with a high- Z material.

The most promising isotope for a bremsstrahlung source is ^{90}Sr because (1) the maximum beta-decay energy from the decay chain is 2.27 MeV (0.61 MeV above the $^9\text{Be}(\nu, n)$ threshold), (2) the half-life is 28.1 yr, and (3) ^{90}Sr is available in large quantities as a byproduct of nuclear fuel reprocessing operations. The useful beta particle is emitted by ^{90}Y in the decay chain.

A 92-Ci ^{90}Sr source was obtained from ORNL in the form of 2 g of SrCO_3 mixed with 9.75 g of lead and sealed in a stainless steel capsule. The SrCO_3 and lead were intimately mixed and compressed into three cylindrical pellets to enhance the brems-

strahlung production per curie of ^{90}Sr . The cylindrical stainless steel capsule is 6.5 cm long and 8 mm in diameter with a 1.3-cm-long handling post at one end; the wall thickness is 1.3 mm. Pulse-height distributions measured with a Ge-Li spectrometer for the ^{90}Sr -Pb source and for a ^{124}Sb source are shown in Figs. 15 and 16, respectively; these distributions have not been unfolded to produce photon energy distributions.

The photoneutron yield was measured for the ^{90}Sr -Pb-Be source with an energy-independent long counter¹⁷ and compared to the yield from a ^{124}Sb -Be source. A 7.5-cm-diam by 10-cm-high beryllium sleeve was the converter for both sources. The ^{90}Sr -Pb-Be source yielded ~ 370 neutrons/s-Ci and the ^{124}Sb -Be source yielded $\sim 3 \times 10^4$ neutrons/s-Ci. Thus, to produce a given neutron source strength about 8000 Ci of ^{90}Sr is required for each curie of ^{124}Sb . At \$0.50/Ci for ^{90}Sr a bremsstrahlung source yielding 10^4 neutrons/s would cost $\sim \$1400$ for the ^{90}Sr . However, for small production runs the packaging costs would dominate the total price.

Fig. 15.

Pulse-height distribution obtained with a Ge(Li) spectrometer from a ^{90}Sr -Pb bremsstrahlung source. The vertical line at 1.665 MeV illustrates the $^9\text{Be}(\text{h}\nu, \text{n})$ threshold.

Fig. 16.

Pulse-height distribution obtained with a Ge(Li) spectrometer from a ^{124}Sb bremsstrahlung source. The vertical line at 1.666 MeV illustrates the $^9\text{Be}(\text{h}\nu, \text{n})$ threshold.

For photoneutron assay systems using ^4He proportional counters for fast-neutron detectors, shielding is required to protect the ^4He detectors from source photons, otherwise pulse pileup makes the assay difficult or impossible. Also, for measurements performed outside of a hot-cell environment additional shielding is required for radiation safety. The shielding requirements for ^{124}Sb and $^{90}\text{Sr-Pb}$ sources were compared simply by placing lead bricks around the sources and measuring the dose rates with gamma-ray survey meters. The results are shown in Table XVII. The dose rates measured with 10 cm of lead may be too high because of scattering effects in the imperfectly arranged shielding mockup. Nevertheless, for a given neutron source strength the dose rates from a $^{90}\text{Sr-Pb}$ source are higher by about an order of magnitude than those from a ^{124}Sb source under practical shielding conditions.

The photoneutron energy distribution from the $^{90}\text{Sr-Pb-Be}$ source was not measured; however, the end-point beta-decay energy for ^{90}Y is 2.27 MeV and so the maximum neutron energy from a beryllium converter is ~ 540 keV. The average neutron energy will be about 100 keV; few neutrons will be produced above 300 keV.

A PuO_2 sample containing 416 g of fissile plutonium was counted with a biased ^4He detector to determine the response of a simple assay system using $^{124}\text{Sb-Be}$ and $^{90}\text{Sr-Pb-Be}$ sources. The response for the $^{124}\text{Sb-Be}$ source was 50.3 counts/s-kg-Ci and for the $^{90}\text{Sr-Pb-Be}$ source was 0.00661 counts/s-kg-

Ci. For an 8000-Ci $^{90}\text{Sr-Pb-Be}$ source the response would be 52.9 counts/s-kg, very close to the response for 1 Ci of ^{124}Sb .

These experiments demonstrate that $^{90}\text{Sr-Pb-Be}$ sources can be used in photoneutron assay systems for the measurement of bulk plutonium and uranium samples or for the measurement of fuel assemblies. The virtues of ^{90}Sr are its suitable beta energies, its long half-life, and its unlimited availability. If the short half-life of ^{124}Sb does not present a serious problem for a potential assay application, then $^{124}\text{Sb-Be}$ is clearly superior to $^{90}\text{Sr-Pb-Be}$ as a photoneutron source. If a long half-life photoneutron system is essential, the choice between ^{226}Ra and $^{90}\text{Sr-Pb}$ would have to be determined by direct comparison for the particular application.

K. Upgrading of the MEGAS Box Counter (T. W. Crane and E. R. Martin)

We plan to upgrade the MEGAS box counter by incorporating intrinsic germanium detectors. The present NaI-based system has insufficient resolution for reliable assay of combustible waste for transuranic (TRU) contamination if fission products are present. The resolution of a germanium detector will in many cases permit an unambiguous determination of the TRU waste at the 10-nCi/g fiducial even when fission products are present. Table XVIII

TABLE XVII
PHOTON DOSE RATES FROM ^{124}Sb AND $^{90}\text{Sr-Pb}$ SOURCES

Configuration		Dose Rate (rem/h)	
Distance from Source (cm)	Lead Shielding Thickness (cm)	~ 1 Ci ^{124}Sb	8000 Ci ^{90}Sr
50	0	4	500
50	5	0.2	4
50	10	0.02	0.3
5	5	6	100
10	10	0.2	2

TABLE XVIII

**PLUTONIUM AND ^{241}Am DETECTABILITY
LIMITS FOR A 1000-s COUNT**

Photo Energy Region (keV)	Isotope	Detectability ^a (nCi/g)	
		No Fission Products	With Fission ^b Products
13.6, 17.3, 20.5 (L x-rays)	Pu ^c	1.2	1.3
59.5	^{241}Am	0.004	0.006
94.7, 98.4 (K x-rays)	Pu ^c	35	40
38.7, 51.6, 129.3	^{239}Pu	9	10

^a3 σ level, 57-l box with 2.4 kg waste, detector placed 40 cm from face of box.

^b5300 nCi, ^{137}Cs .

^cPlutonium with 94% ^{239}Pu and 6% ^{240}Pu .

shows that the presence of the fission product ^{137}Cs (5300-nCi contamination) only slightly affects the assay sensitivity of the germanium detector. We estimate that an assay based on the L x-rays alone in the presence of 300 μCi of ^{137}Cs would be sensitive to 10 nCi/g (3 σ or better) of transuranics with a 1000-s counting time.

The resolution of the germanium detectors will also facilitate the use of more sophisticated analysis techniques. When a differential absorption correction is applied to the three L x-ray groups, the discrepancy between groups is reduced from a factor of 2 to only a $\pm 7\%$ variation for the two TRU-contaminated boxes referenced to a standard (Ref. 2, p. 6).

A novel technique based upon Compton scattering within the waste box can also be used to estimate attenuation corrections and to flag boxes that do not meet the criteria for a reliable assay. The principle can be explained with the aid of an idealized gamma-ray peak ($E_\gamma \geq 50$ keV) depicted in Fig. 17. For low-density material such as combustible waste, the ratio of the Compton-scattered gamma rays to the number of counts in the photopeak is proportional to the attenuation factor μx , where μ is the

linear attenuation coefficient at the photopeak energy and x is the average transmission thickness. From the spectrum parameters shown in Fig. 17, the Compton-to-peak ratio C can be computed:

Fig. 17.

Pulse-height spectrum of a single photopeak illustrating the general procedure for determining photopeak area and Compton scattered events.

$$C = (B_1 - n_1 B_2 / n_2) / A$$

and the error in C is given by

$$\sigma(C) = \frac{1}{A} [(1 + nC/2n_1)^2 B_1 + C^2 P + (n_1/n_2)^2 (1 - nC/2n_1)^2 B_2]^{1/2}$$

where

B_1 = number of counts in the n_1 background channels,

P = number of counts in the n photopeak channels,

A = photopeak area. (see Ref. 18, p. 29) as calculated by

$$A = P - [(n/2)(B_1/n_1 + B_2/n_2)]$$

The attenuation coefficient can be obtained from

$$\mu x = \ln(A_0/A)$$

where A_0 is the unattenuated photopeak area.

The linear relationship between C and μx ($\mu x < 1$) is shown in Fig. 18. Agreement between the Monte Carlo computer calculations and experimental measurements is reasonable. The proportionality constant between C and μx can be determined either by measurement or by calculation. The value depends upon the gamma-ray energy, the energy width of the Compton window, and the geometry used in the measurement.

L. Adding the DYMAC Data Link to the CMB-11 Segmented Gamma Scanner (SGS) (E. R. Martin)

The SGS software and hardware were changed to permit automatic data transfer to the DYMAC central computer upon operator command. An interface added to the SGS system serializes the assay data, fissile value, and error estimate in a fixed format dictated by DYMAC computer constraints, and transmits it upon operator command. The SGS

Fig. 18.

Data for Compton scattering attenuation correction.

codes were changed to recognize the new operator interrupt command and make the hardware transfer as required.

M. Determination of the Total Uranium Content in Materials Test Reactor (MTR) Fuel by K-Edge Densitometry (D. G. Langner, S. T. Hsue, and T. R. Canada)

Fuel bundles for MTRs nominally contain 220 g of 93.15%-enriched ^{235}U . To verify that a bundle contains the nominal 18 plates, the uranium in the bundle must be measured to a 1σ accuracy of plus or minus one-quarter of a plate, or 1.4%. A simple technique to achieve the required accuracy is k-absorption-edge densitometry.^{2,10} The ratio of transmissions on both sides of the uranium's 116-keV k-edge is measured to determine the density of

uranium. If the energies are close enough to the k-edge, the measurement becomes virtually independent of other materials present in the bundle.

To demonstrate this technique, transmission measurements were made at 109.8, 130.5, 177.2, and 198 keV using a ^{109}Yb source for 1, 4, 18, and 19 MTR plates. To remove the effects of the aluminum in the bundles, the measured transmission values $T(E)$ were extrapolated to the k-edge of the uranium at 116 keV. The extrapolation was performed by fitting $\ln [-\ln T(E)]$ vs $\ln E$, where E is the gamma-ray energy, to a straight line. A value of the transmission $T(116^+)$ on the high-energy side of the k-edge was then computed from the fit. The 109.8-keV transmission data were extrapolated to obtain a transmission $T(116^-)$ on the low-energy side of the k-edge using

$$\ln[-\ln T(116^-)] = \ln[-\ln T(109.8)] + \alpha \ln(116/109.8).$$

For uranium, $\alpha \cong -2.44$.

Figure 19 shows a plot of the log of the ratio of transmissions R obtained by extrapolation versus the number N of MTR fuel plates. A fit of these data gave

$$\ln R = (-0.122 \pm 0.001)N - (0.001 \pm 0.011)$$

which to within the uncertainties is equal to the theoretical matrix-independent relation for uranium

$$\ln R = -0.123 N.$$

The relative error in the measured number of plates is related to the relative error in the ratio of transmissions by

$$\frac{\Delta N}{N} = \frac{-1}{(\Delta\mu/\bar{\rho}x)N} \frac{\Delta R}{R}$$

where $\Delta\mu$ is the change in the mass absorption coefficient for uranium at the k-edge and $\bar{\rho}x$ is the average density of the uranium per plate. For extrapolated transmissions and an 18-plate bundle, this becomes

$$\frac{\Delta N}{N} \cong 0.45 \frac{\Delta R}{R}.$$

Fig. 19.

Log of the ratio of transmissions R obtained by extrapolation vs the number N of MTR fuel plates.

Thus to measure $\Delta N/N$ to 1.4%, $\Delta R/R$ must be determined to 3%. The errors propagate through the extrapolation according to

$$\begin{aligned} \frac{\Delta R}{R} \cong & \left[\left(\frac{\ln T(116^+)}{\ln T(130.5)} \right)^2 \left(\frac{\Delta T(130.5)}{T(130.5)} \right)^2 \right. \\ & + \left(\frac{\ln T(116^+)}{\ln T(177.2)} \right)^2 \left(\frac{\Delta T(177.2)}{T(177.2)} \right)^2 \\ & + \left(\frac{\ln T(116^+)}{\ln T(198.0)} \right)^2 \left(\frac{\Delta T(198)}{T(198)} \right)^2 \\ & \left. + \left(\frac{\ln T(116^-)}{\ln T(109.8)} \right)^2 \left(\frac{\Delta T(109.8)}{T(109.8)} \right)^2 \right]^{1/2}. \end{aligned}$$

For a 30-min count on an 18-plate bundle, we obtained $(\Delta T/T)$ at 130.5 to be approximately equal to 1.5% and $(\Delta T/T)$ at 109.8, 177.2, and 198 to be approximately equal to 0.5%. This gave $\Delta R/R \sim 2.4\%$, which is within the required limit.

Does the extrapolation technique reduce matrix effects enough to warrant this additional analysis? Suppose one wished to verify that a nominal 18-plate fuel bundle indeed contained 18 uranium-filled plates. But suppose one plate in the bundle had (1) the uranium replaced by lead (high-Z), or (2) the uranium replaced by aluminum (low-Z). The measurement is performed such that there is a 1.4% (1σ) accuracy for the nominal 18-plate bundle. The ratio of transmissions is computed by two methods;

Method I: R is determined using the transmissions at 109.8 and 130.5 keV corrected for the effects of the known aluminum in the bundle.

Method II: R is determined using the extrapolation technique. No aluminum correction is performed.

The results of the experiment are classified according to the following simple criterion: if the predicted number of plates falls between $N - 0.5$ and $N + 0.5$ for N an integer, the conclusion will be that there are N plates in the bundle. How often would cases (1) and (2) be correctly classified as 17 plates containing uranium?

Table XIX gives the percentage of correct classification predicted for this "thought experiment." Since Method I gives a relatively low confidence level for correctly classifying high-Z material substitution, the extrapolation technique is necessary to ensure that both high- and low-Z substitutions can be detected at a 95% confidence level. The results obtained from Method I, however, demonstrate an interesting benefit of the k-edge technique. Although the classification of the bundle with a high-Z substitution would be wrong 36% of the time when Method I was used, the bundle would be misclassified as containing 16, not 18, uranium-filled plates. The error flag would still be raised to indicate that the bundle contained too little uranium. In fact, for any substitution, the classification will always be 17, or fewer, uranium-containing plates. With at least a 95% confidence level, the bundle will not be misclassified as containing the nominal 18 plates.

N. The ERDA Safeguards Technology Training Program (T. R. Canada, J. L. Parker, and J. W. Tape)

Two courses of the ERDA Safeguards Technology Training Program four-course sequence were given in Fall 1976. The "Fundamentals of Nondestructive Assay of Fissionable Materials Using Portable

TABLE XIX
CORRECT CLASSIFICATION FOR HIGH- AND LOW-Z
SUBSTITUTION IN MTR FUEL

Type of Analysis	Substitution	Mean No. of Plates Predicted	Correct Classification as 17 Plates (%)
Method I (aluminum correction)	Pb	16.59	64.1
	Al	16.97	95.3
Method II (extrapolation)	Pb	16.997	96.3
	Al	17.023	95.4

Instrumentation" training session was conducted from November 1-5 with 15 IAEA representatives and 15 domestic U.S. participants in attendance (see Table XX). Seventeen Group R-1 staff

members were involved in lectures, demonstrations, or laboratory instruction. Because the groups had only three or four students, the instructors could

TABLE XX

**FUNDAMENTALS OF NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIAL
USING PORTABLE INSTRUMENTATION: COURSE ATTENDEES**

<u>Organization</u>	<u>Representative</u>
Exxon Nuclear Company	Michael W. Andrews
IAEA	Uldis Barda, Latvia Calvin H. Delegard, U.S.A. Elmir Dermendjiev, Bulgaria Ichiro Hiramatsu, Japan John B. Hornsby, United Kingdom Jacek T. Kaniewski, Poland Hayao Kawamoto, Japan Alberto Lumetti, Italy Arrigo Massera, Italy Albano de Moncada, Portugal Ved Narang, India Pierre Noir, France Eric Payne, Canada Ken Sanders, U.S.A. Ijaz Umar, Pakistan
US NRC	James Blaylock James P. Patterson Thomas U. Tuten
Sandia Laboratories	R. J. Gonzales Tony R. Lopez
Battelle Pacific Northwest Laboratory	Clark O. Harvey
Rockwell International	Marvin E. Hughes Jill Irvin
Mason and Hanger	Elmer Laughter
Atlantic Richfield Hanford Co.	George Westsik
LASL	Richard Dooley Richard Imprescia Evita Medina Joseph Sapir

spend a considerable amount of time with each student (Fig. 20). Although the "Fundamentals" course stressed the use of portable instrumentation, other techniques were touched upon. In addition to the usual topics on the use of portable instrumentation for passive neutron and gamma-ray assay of uranium and plutonium in small cans, the Random Driver and neutron well coincidence counter were demonstrated. A special session on the verification of MTR fuel plates and elements was also offered for the benefit of the IAEA inspectors. Lectures covered techniques involved in holdup measurements, practical assay problems and solutions, and hypothetical design criteria for the next generation of portable NDA equipment.

A new course, "In-Plant Nondestructive Assay Instrumentation," was given during the week of

December 6. Twenty-seven trainees (Table XXI), including eight for the IAEA, spent a day with each of four different nondestructive assay instruments (two neutron and two gamma-ray assay systems) suitable for use in nuclear fuel cycle plants (see Fig. 21). A Random Driver for the active assay of large-mass uranium samples constituted one-half of the neutron-based assay instrumentation. This Random Driver was the latest model of a series and employed modern electronics and minicomputer technology for data analysis. The other half of the neutron assay instrumentation involved a specially constructed, dual range well coincidence counter for use in this school. Plutonium samples were used to calibrate this dual range detector and to study its characteristics.

Fig. 20.

November 1-5 Fundamentals of NDA course in progress. Instructors Jack Parker (left foreground) and Diana Langner (right background) explain basics of gamma-ray spectroscopy as it applies to nondestructive assay of SNM. Parker is pointing out a high-resolution, high-efficiency, Ge(Li) gamma-ray detector while Langner manipulates the controls of a sophisticated multichannel pulse-height analyzer.

TABLE XXI**IN-PLANT NONDESTRUCTIVE ASSAY INSTRUMENTATION:
COURSE ATTENDEES**

<u>Organization</u>	<u>Representative</u>
IAEA	Winston Alston, South Africa Marco Ferraris, Italy Ahmed Keddar, Algeria Ilia Mishev, Bulgaria D. R. Terrey, United Kingdom Leslie Thorne, United Kingdom Joan Todorescu, Romania Stanley Turel, U.S.A.
Westinghouse Hanford Co.	Dennis Fazzari
US NRC	Anthony T. Gody Donaid R. Joy E. Thomas Shaub Philip Ting
Goodyear Atomic	James R. Griggs
Nuclear Fuel Services, Inc.	Ron Hawkins
Rockwell Internation	Jill Irvin James Lawless
Atlantic Richfield Hanford Co.	Cecil H. Kindle
US ERDA	Ray Lux Cathy S. Morimoto J. F. Olivier Craig S. Smith Barbara M. Wilt
Babcock and Wilcox	R. Dee Sherrill
LASL	Richard Dooley Evita Medina Joseph Sapir

Gamma-ray-based assay techniques were represented by a Uranium Solution Assay Device (USAD) and the segmented gamma scanner (SGS). Both systems use high-resolution detectors, nuclear spectroscopy electronics, and a computer-based multichannel analyzer for scanning control (the SGS) and data accumulation and analysis. Two ver-

sions of the SGS were available: one setup for uranium assay and the other for plutonium. The USAD was used for assay of uranium solution varying in concentration from a few parts per million to hundreds of grams per liter with accuracies as good as 0.5%. Participants were encouraged to become familiar with the equipment in a hands-on fashion.

Fig. 21.

December 6-10 "In-Plant Nondestructive Assay Instrumentation" training session in progress. Willy Hsue, LASL instructor, teaches Philip Ting (NRC) how to type in data as Joe Sapir (LASL, R-1) and Kathy Morimoto (ERDA, ALOO) look on. The typewriter at which Ting sits is connected to one of the two computer-based segmented gamma counters used during the course.

Lectures on dynamic materials control (DYMAC) systems for the LASL plutonium facilities were given in conjunction with a tour of the new LASL plutonium facility. An unusual amount of time was spent in preparing the instrumentation course manual and the lectures for this course, but participants felt it was time well spent.

Another new course, "Integrated Nondestructive Assay Systems—Concepts and Implementation,"

will be offered in March 1977. The "Gamma-Ray Spectroscopy for Nuclear Material Accountability" course will be given for the third time in May 1977. Current plans are to repeat the "Fundamentals" course in October 1977 and the "In-Plant" course in December 1977.

PART 2

DEVELOPMENT AND DEMONSTRATION OF DYNAMIC MATERIALS CONTROL—DYMAC PROGRAM (R-Division, CMB-Division, and E-Division Staffs)

I. CONCEPTS AND SUBSYSTEM DEVELOPMENT

A. NDA Instrumentation

1. Self-Regulating Thermal-Neutron Coincidence Counting Efficiencies and Effective Masses (N. Baron)

A phenomenological approach to passive non-destructive assay by thermal-neutron coincidence counting is described. The principal objective is to account accurately for any modification of the apparent spontaneous-fission coincidence count rate caused by self- and/or induced-multiplication as well as matrix effects. This technique requires the determination of an apparent coincidence counting efficiency defined to be the quotient of the measured true coincidence count rate divided by the average creation rate of prompt-neutron pairs resulting from spontaneous fission processes. If this efficiency is not an invariant, then the presence of multiplication and/or matrix effects is implied. This efficiency can be written as

$$\text{eff} \equiv \frac{\dot{N}^B}{\sum_{i=1}^n \frac{1}{2} a_i \bar{\nu}_i m_i} \quad (1)$$

where

$\dot{N}^B$ is the coincidence count rate associated with a given gate width and is measured by using the outputs of all the neutron counting tubes in a well-counter having two concentric rings of tubes,

a_i is the number of spontaneous fissions per second per gram of isotope i ,

$\bar{\nu}_i$ is the average number of prompt neutrons emitted per spontaneous fission event of isotope i , and m_i is the mass of isotope i .

Equation (1) can be rewritten as

$$\text{eff} \equiv \frac{2}{a_{240} \bar{\nu}_{240}} \left(\frac{\dot{N}^B}{M_E} \right) \quad (2)$$

where M_E is the effective mass of ^{240}Pu and is defined to be

$$M_E \equiv \sum_{i=1}^n \frac{a_i}{a_{240}} \frac{\bar{\nu}_i}{\bar{\nu}_{240}} m_i \quad (3)$$

By rewriting Eq. (2) as

$$M_E = \frac{\dot{N}^B / \text{eff}}{\text{calibration factor}} \quad (4)$$

we stress that the effective mass of fertile material is determinable from the measured thermal-neutron coincidence count rate if one knows the coincidence counting efficiency, eff , as defined by Eq. (1). The calibration factor in Eq. (4) is the reciprocal of the unbracketed terms of Eq. (2).

The measuring instrument was a thermal-neutron well-counter similar to that described in Ref. 12, p. 13. The inner and outer rings had 12 and 16 ^3He counters, respectively. Using known source strengths, the coincidence count rates were measured (1) with both rings of ^3He counters in the electrical circuit, i.e., $\dot{N}^B$, (2) with only the inner ring of ^3He counters in the electrical circuit, i.e., $\dot{N}^I$, and (3) with only the outer ring of ^3He counters in the electrical circuit, i.e., $\dot{N}^O$. The coincidence count

rates were measured using the previously reported LASL shift register circuit⁷ and a gate width of 32 μ s.

By the following analysis, we can predict the efficiency, eff, defined by Eq. (1). The reason for the success of this analysis is still unclear, but we hope that further work will provide a suitable explanation.

If the calculable coincidence count rate, given by

$$\dot{N}^B(\text{calc}) = \dot{N}^I + \dot{N}^O + 2(\dot{N}^I \dot{N}^O)^{1/2}, \quad (5)$$

satisfies the relation

$$\frac{\dot{N}^B(\text{calc}) - \dot{N}^B}{\dot{N}^B} < 2\sigma, \quad (6)$$

where σ is the statistical counting uncertainty in $\dot{N}^B$, then the efficiency, eff, defined by Eq. (1) can be predicted reasonably well by the relation

$$\text{eff} = \kappa \frac{(\dot{N}^I / \dot{N}^O)}{(\dot{T}^I / \dot{T}^O)}, \quad (7)$$

where $\dot{T}^I$ and $\dot{T}^O$ are the "singles" count rates measured by the inner and outer tube rings, respectively.

Otherwise, the coincidence counting efficiency, eff, is correctly given by

$$\text{eff} = \kappa \frac{(\dot{n}^O / \dot{N}^O)}{(\dot{T}^I / \dot{T}^O)} \quad (8)$$

where $\dot{n}^O$ is obtained from the measured values of $\dot{N}^B$ and $\dot{N}^I$ by the relation

$$(\dot{n}^O)^{1/2} = (\dot{N}^B)^{1/2} - (\dot{N}^I)^{1/2}. \quad (9)$$

The proportionality constant κ in Eqs. (7) and (8) is taken to be the average value obtained by measuring p standards and is given by

$$\kappa = \frac{1}{p} \sum_{i=1}^p \left[\text{eff} \frac{\dot{T}^I / \dot{T}^O}{C / \dot{N}^O} \right]_i \quad (10)$$

where

$$C = \dot{N}^I \quad (11)$$

if relation (6) is satisfied; otherwise,

$$C = \dot{n}^O. \quad (12)$$

Once the coincidence detection efficiency is determined by the criteria set forth in Eqs. (5) through (12), the unknown effective mass of spontaneously fissioning material M_E is given by Eq. (4).

Table XXII shows the results of the analyses of the measurements on six standards with effective masses of ²⁴⁰Pu ranging from 3.84 to 37.4 g. The fact that those standards having a matrix of MgO have a greater count rate than similarly loaded standards having a matrix of diatomaceous earth is probably due to induced fissions caused by neutrons produced by the large Mg(α ,n) reaction. Columns 4 and 7 in Table XXII show that the efficiencies calculated with Eqs. (7) and (8) agree well with the efficiencies defined by Eq. (1). A comparison of columns 4 and 6 demonstrates the self-regulating nature of the count rate ratios relative to variations in coincidence counting efficiencies, and the linear nature of this relationship is shown in Fig. 22.

The results of conventional calculations of effective masses are shown in column 11 and the errors are listed in column 12 of Table XXII. The calibration factor for this calculation was taken to be the average of the six measured coincident count rates per gram, $\dot{N}^B/M_E$, listed in column 10 of the table. The results of similar effective-mass calculations in which the coincidence count rates were corrected for efficiency as defined by Eqs. (7) and (8) are shown in column 8 of Table XXII, and the errors are listed in column 9. The effective masses are generally more accurately predicted by this phenomenological approach than by the conventional technique. In fact, the sum of the effective masses predicted by this efficiency correction technique, obtained by summing the values in column 8 of Table XXII, is in error by only 0.3%. This is to be compared to the total resulting from the conventional technique, obtained by summing the values in column 11 of Table XXII and which is found to be in error by 4.1%.

The linear relationship of the coincidence count rate corrected for efficiency and the effective mass are emphasized in Fig. 23 where, for comparison, is also shown the coincidence count rate uncorrected by the coincidence efficiency factor.

TABLE XXII

COINCIDENCE EFFICIENCIES AND RESULTANT EFFECTIVE MASSES

Column											
1	2	3	4	5	6	7	8	9	10	11	12
M_E (g)	$\dot{N}^B$ (counts/s)	Material Form and Matrix	eff (%) Eq. 1	κ Eq. 10	Count Rate Ratios ^a	eff (%) Eq. 7 or 8	M_E (g) Eq. 4	Percent Error	$\dot{N}^B/M_E$	M_E (g) ^b	Percent Error
3.84	89.8	PuO ₂ + Diatomaceous Earth	4.58	0.0479	0.9558	4.69	3.75	-2.3	23.42	3.41	-11.2
3.84	100.7	PuO ₂ + MgO	5.13	0.0478	1.074	5.27	3.74	-2.6	26.23	3.82	-0.5
7.67	184.1	PuO ₂ + Diatomaceous Earth	4.70	0.0510	0.9210	4.52	7.96	+3.8	24.00	7.86	+2.5
7.67	219.7	PuO ₂ + MgO	5.60	0.0495	1.132	5.56	7.73	+0.8	28.63	8.33	+8.6
15.3	439.0	PuO ₂ + MgO	5.60	0.0495	1.132	5.56	15.4	+0.6	28.60	16.6	+8.5
37.4	1025.0	PuO ₂	5.37	0.0486	1.107	5.44	36.9	-1.3	27.41	38.8	+3.7
(75.72) _{total}			(0.0491) _{av}			(75.48) _{total}			(26.30) _{av}	(78.82) _{total}	

^aCalculated using $\frac{\dot{N}^I/\dot{N}^O}{\dot{I}^I/\dot{I}^O}$ or $\frac{\dot{N}^O/\dot{N}^I}{\dot{I}^O/\dot{I}^I}$

^bConventional calculation; $M_E = \dot{N}^B$ / response per gram, where the response per gram is the average of those values listed in column 10.

Fig. 22.

Coincidence counting efficiency vs measured count rate ratio.

Fig. 23.

Coincidence count vs effective mass.

2. Neutron Monitoring of Direct Oxide Reduction (R. S. Marshall, W. R. Severe, and R. Siebelist)

Group CMB-11 is conducting research on the direct reduction of $^{238}\text{PuO}_2$ to ^{238}Pu metal. Group R-1 monitored the progress of the reduction reaction by measuring the emitted neutron flux. We expected that the initial neutron flux, a combination of spontaneous fission neutrons from ^{238}Pu plus (α, n) neutrons, would decrease as the $^{238}\text{PuO}_2$ was converted to ^{238}Pu metal.

Figure 24 shows the temperature and neutron flux history during a reduction experiment conducted in a molten CaO matrix. Neutron counting was started during the last 8 min of a heating cycle during which the mixture was heated to 820°C . The heating furnace was then cut back to stabilize the molten CaO at 820°C and a stirrer was started. The stirrer brought the PuO_2 (sitting below the less dense molten CaO) into contact with the calcium metal (floating on top of the molten CaO). An exothermic reaction occurred, as can be seen by the temperature spike in Fig. 24. Notice the rise in neutron flux corresponding to the activation of the stirrer. We attribute this rise to a more favorable counting geometry between the $^{238}\text{PuO}_2$ and the SNAP II neutron detector. The rise is followed by a rapid drop in neutron flux closely paralleling the

Fig. 24.

Neutron flux and temperature history of a $^{238}\text{PuO}_2$ reduction. Arrows indicate time that stirrer was turned on and off.

temperature drop. Both indicators suggest completion of the reduction reaction.

We expected the after-reduction neutron flux to decrease to about 75% of the before-reduction flux and to remain constant after the reaction; Fig. 24 shows that neither occurred. Examination of emission spectrographic analyses of the PuO_2 feed material and the plutonium metal product revealed that boron was present in the metal at much higher concentrations than in the feed PuO_2 (less than 1 ppm vs 400 ppm). Because boron has a very high cross section for the (α, n) reaction, it seems likely that the dissolution of boron in the molten plutonium explains the high and rising neutron flux.

B. Data Acquisition

Automatic Label Readers (T. Gardiner)

Evaluation work has begun on the use of portable label readers for the inventorying of item control areas. Reliability experience at other facilities has been mixed, prompting the need for a thorough review of available options. At present, we have ordered a bar label printer, which uses the CODE-A-BAR format, and three readers from different manufacturers. Later we will evaluate character recognition equipment for application to the inventory problem.

C. Data Base Management (R. F. Ford,* J. Hagen, C. L. Slocomb,* and T. M. Sandoski*)

Transaction Data Integrity

DYMAC is concerned with data integrity in that the information placed in the data base must be sufficiently correct to allow generation of an accurate book inventory. To ensure data integrity, the DYMAC program relies on a number of techniques that can prevent, detect, and subsequently correct the majority of errors occurring during operation. To minimize the domino effect of error propagation, the system emphasizes early detection of invalid data prior to data-base update.

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Interactive Man-Machine Dialogs

In cases where sequences of data or transactions are required, a set of protocols is imposed via the dialogue at the transaction terminal.

Syntactical Checking of Responses

Operator responses are examined in detail. For example, an account number must be three digits, a person's name may contain no numerics. Incorrect syntax of the response generates an explicit error message to the operator and the prompt reappears on his display.

Image Files

The program supplies all fixed information in the transaction to reduce operator error.

Use of Known Data-Base Information

The operator enters only the lot ID (name) of the material. Other pertinent information, such as location and amount of SNM, is supplied from the inventory record.

Inventory Pre- and Post-Transaction Searches

When a transaction is initiated, the program searches the inventory to ensure that the lot ID exists. On completion of the dialogue, the program again searches the inventory to ensure that either (1) the new lot ID already exists, in the case of a mixing transaction, or (2) does not exist, for all other transaction types.

Logical Relationship Check

The program checks to validate the relationship between the transaction types, lot ID, account numbers, locations, amounts, and names of the operators involved.

Preupdate Edit of Transaction

Before completion of a transaction and subsequent file update, the entire transaction appears on

the display and the operator can make any changes needed. He can only change those items that he entered; he cannot edit an image parameter or a quantity supplied by the data base.

Protocol for Consistent Data Collection

All data sent to the computer from any line, terminal, or NDA instrument look alike, that is, they have ASCII characters with even parity. The program checks each character and echoes it back to the source for display or comparison. In this way, data collection and checking are the same regardless of source. Data conversion is thereby avoided and the likelihood of introducing errors is reduced.

Management Reporting in Near Real Time

Every morning, process supervisors will receive a list of the previous day's activity which allows them to monitor the accuracy of their inventory books.

Inquiry and Correction Facilities

Inquiry and correction programs allow management to trace back from inconsistent book totals to discover which transactions were in error and to write the required corrective transactions.

Error Feedback

All errors are classified by type and logged. The results are used to update operator training programs and serve as the basis for possible changes or enhancements to the DYMACE software.

Measurement Control Program

The measurement control program monitors the calibration and accuracy of NDA instruments and generates control charts for each one. The program prevents a suspect instrument from handling transactions until it is back in good working order.

χ^2 -EVALUATION OF NEUTRON WELL
COINCIDENCE COUNTER DATA

Experience with the Measurement Control Program

As described in Ref. 1, a formal program for measurement control has been written. After official approval it will be used as an operational procedure for both the DP-Site counting room and the metal-to-oxide conversion process. The program contains several control procedures. The t-distribution will be the control parameter for accuracy of response. The precision parameters will be the F-distribution for weighing devices and the χ^2 -distribution for counting devices. All parameters will be controlled by warning limits at the 95% confidence level and action limits at the 99% confidence level.

The χ^2 precision control parameter has already demonstrated its value by detecting an electrical problem in the 2- ℓ neutron coincidence well-counter. A series of 15 replicate measurements was made with the 2- ℓ coincidence counter. The machine-calculated standard deviation, based on the Poisson distribution, was compared with the standard deviation of the replicate measurement values. Table XXIII shows the replicate values and their Poisson standard deviations. The average Poisson standard deviation was 349.3, whereas the standard deviation of the replicate measurement values was 1597.8. The χ^2 -parameter is given by

$$\chi^2 = s^2 / \bar{\sigma}^2$$

where σ and s are the Poisson and replicate standard deviations, respectively. For this case the value of the χ^2 -parameter is 20.92. For 15 measurements the χ^2 -distribution gives control limits of 1.69 at 95% confidence and 2.08 at 99% confidence. The instrument was definitely out of control. Installation of a

<u>Reals</u>	<u>Uncertainties</u>
73138	352
73594	353
74193	354
73288	353
73888	353
68678	341
71483	348
70318	345
71518	347
71019	347
70047	344
72553	350
72817	350
73108	351
72998	351
$\bar{x} = 72176$	$\bar{\sigma} = 349.3$
$s = 1597.8$	

more effective instrument ground corrected the problem. Inspection of the Table XXIII data might have resulted in the same conclusion as that reached by the χ^2 -parameter, but use of the parameter provided a sound statistical basis for the decision.

Evaluation of the SGS and the 2- ℓ thermal-neutron coincidence counter is not yet complete. Preliminary results indicate that both instruments will be able to perform well over a wide range of plutonium values. However, matrix may prove to be a problem for both systems. This evaluation will continue.

II. DYMAC IMPLEMENTATION

A. DP-Site Test and Evaluation Phase (R. H. Augustson).

We have reported²⁰ that the ash-leach process had been selected as the prototype line for test and evaluation of DYMAC hardware and software. Instrument installation and software development were essentially complete when ERDA requested Group CMB-11 to convert the ash-leach line to a metal-to-oxide conversion process because of programmatic commitments. We decided to convert the DYMAC evaluation program to monitor the new process. This entailed many software changes within a short time. We decided to use the installed hardware (terminals and NDA instrumentation) as is, although in some cases measurement capability was compromised. The conversion is now complete and the DYMAC system is in operation again.

1. Metal Dissolution Unit Process Description (J. Hagen, T. M. Sandoski,* W. S. Severe, R. Siebelist, R. P. Wagner,** and W. M. Olson**)

A different unit process, metal dissolution, has been incorporated into the DYMAC system at DP Site. Material flow, NDA measurement points, and balance areas are shown in Fig. 25. In this process plutonium ingots weighing approximately 2 kg are received and unpacked. Accumulated oxide is scraped from the ingot. The ingot is then cut into quarters. Each quarter-ingot is dissolved in nitric acid and any residue is filtered out. At this point various nitrate solutions are blended together to assure the proper isotopic composition of the finished oxide product. Peroxide is added and the solution is filtered. Americium is removed in the filtrate. The peroxide cake is redissolved with nitric acid, reduced, and precipitated with oxalic acid. The oxalate cake is then calcined. Cans of this oxide are screened, blended, and canned for shipment.

Four unit process accounting areas have been defined in the metal dissolution unit process.

a. Receiving balance area. Here the input is the shipper's value and the measured outputs are the scraped oxide, the clean ingot, and the shipping materials. Any material remaining on the books is assigned to the shipper-receiver difference account.

b. Cutting balance area. The input is the measured clean ingot. The measured outputs are the four quarters, the metal sawdust, and the worn-out saw blades. Material remaining on the books is assigned to the material in process (MIP) account for the cutting box.

c. Dissolution balance area. The quarter-ingot is the input and the measured outputs are nitrate solution residues, undissolved metal, peroxide and oxalate filtrates, and the calcined oxide. Remaining book material here is assigned to the MIP account for this balance area.

d. Screening balance area. Here the input is the measured calcined oxide and the output is the measured product. Any differences are assigned to the MIP account for this screening area.

These processes will be monitored by the DYMAC system in parallel with the paper system and the monitoring will continue for several weeks, after which the data will be analyzed.

2. Software Conversion from Ash-Leach Recovery Process to Oxide Conversion Process (R. F. Ford,* J. Hagen, C. Slocomb,* and W. C. Barnett*)

All of the accounting software has been modified and tested to allow rapid and easy inclusion of any new transactions or alterations to existing ones. The primary software modification has been to adopt a packet-like scheme for the accountability program. That is, each transaction is characterized by a packet or list of parameters such as the operator

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- S* SS value and % purity at shipper's value
W bulk weight measurement $\times$ % purity = SS value
F bulk weight measurement $\times$ % purity $\times$ oxide factor = SS value
G segmented gamma scan measurement of ^{238}Pu mass
SA solution assay measurement of ^{238}Pu mass
N neutron coincidence count of ^{240}Pu mass
Z ratio of volume to be blended to the initial volume

Fig. 25.

Flow diagram of metal dissolution process showing NDA measurement points and material balance area.

prompts, responses, or edits required, the measurements necessary, the types of file searches needed, and the kind of file updates required. A packet is created for each transaction, and the accounting program is driven by this list. With this scheme the software has the necessary flexibility to cope with process changes, such as the one from the ash-leach to the metal dissolution. Other software modifications include creation of an executive task to manage a pool of worker tasks to obviate the need for a task per terminal, installation of a timer task to allow recovery from slow or nonexistent operator input or a failure of the communications line; improved logging, and error handling.

3. Refinement-of-Accuracy Study for the Plutonium Solution Assay Instrument (J. L. Parker and R. Siebelist)

In the previous progress report² we discussed the results of an initial program to evaluate the solution assay instrument (SAI) for the assay of 0.5- to 5.0-g/l ash-leach solutions. Eighteen samples had been assayed both by the SAI and by isotopic dilution mass spectroscopy. At the time the average of the ratio of the SAI results to the isotopic dilution results was 0.981, with a standard deviation of 0.013. Discovery and correction of a significant bias in the calibration of the SAI have changed the average ratio to 0.995, with the standard deviation still at 0.013. All other comments in the previous report still stand, but all the ratio values in Table XVI of Ref. 2 should be multiplied by 1.015.

The biasing effect was loss of mass, apparently by diffusion, from the solution standards with which the SAI was calibrated. This was approximately 0.1 g per month, which in about 1.5 yr of storage changed the volume of the standards sufficiently to produce a 1.5% change in specific response relative to samples with the correct volume. The effect is due primarily to the operation of the $1/r^2$ law and is a consequence of the necessarily small distance (about 4.3 cm) between the sample bottle and detector.

4. Applications of the Solution Assay Instrument (SAI) (W. R. Severe, J. L. Parker, R. P. Wagner,* and R. Siebelist)

The SAI is currently being used for two programs. The first program involves measuring filtrate solutions generated in the metal-to-oxide conversion line. There are peroxide filtrates and oxylate filtrates. Both types have a plutonium concentration range of 0 and 0.5 g/l. For a nominal filtrate concentration of 0.25 g of plutonium/l, and a 20-min counting time, 1σ precisions of 15% relative standard deviation (RSD) are obtained. The SAI solution sample is then sent to Group CMB-1 for radioassay.

We plan to evaluate the relative precision and accuracy of these two methods for several months. If a significant bias is noted, final judgement of system accuracy will be made by comparison with isotopic dilution mass spectroscopy. The radioassay results are currently used for material accountability.

The second program involves special measurement projects. One of these special projects was the determination of the plutonium metal dissolution rate for the metal-to-oxide conversion process. The concentration of the nitrate solution determined by the SAI is plotted against time in Fig. 26. When correlated with other variables, this and other dissolution-rate studies made with the SAI helped production personnel optimize the dissolution process.

A second special project was to provide rapid measurements of the initial peroxide and oxylate filtrates. The timely measurements made possible by the SAI allowed the final process parameters to be set with minimal delay.

In a third special project, the SAI was used for plutonium metal enrichment measurements. A special collimator and a plutonium metal standard have been fabricated. Plutonium metal entering the metal-to-oxide conversion line will be measured, and an evaluation of the practicability of such a measurement system will be evaluated.

*Group CMB-11.

Fig. 26.

Plutonium metal dissolution rate.

5. In-Line Neutron Coincidence Unit (W. R. Severe, R. Siebelist, and N. Ensslin)

The in-line thermal-neutron coincidence counter was installed during the previous reporting period. It was originally designed to measure low-level waste for the ash-leach process. When the ash-leach process was replaced by the metal-to-oxide conversion process, the primary function of the counter changed to the measurement of kilogram quantities of plutonium. The change in function required extensive instrument modification. Modifications have been completed, and calibration is in progress using standards ranging from 500 to 1000 g of plutonium. The counter will provide DYMAC measurements for the pure-product oxide cans produced by the metal-to-oxide conversion process. The product cans will also be measured by sampling and analytical chemistry, hence the metal-to-oxide conversion process will provide an excellent method for evaluating the counter's accuracy through measuring large quantities of plutonium.

6. Plutonium-Button Verification Measurements (R. S. Marshall and R. Siebelist)

LASL received a shipment of 48 plutonium metal buttons from Atlantic Richfield Hanford Co. (ARHCO). Each button weighed approximately 2 kg and was packaged in a sealed metal can. ARHCO supplied complete plutonium isotopic ratios and

metallic impurity information, but the physical dimensions of each button were unknown.

We made verification measurements using the equipment illustrated in Fig. 27. Our measurements can be considered field measurements in that:

- (a) counting time was limited to 30 s per button;
- (b) can-to-detector distance of 0.5 m was maintained by visually centering each can on the rotator;
- (c) measurements were made in a high background area just outside a plutonium storage vault;
- (d) measurements were made beside an open door: temperature in the measurement area ranged from an estimated -1°C (30°F) to 4°C (40°F); and
- (e) only 2 h were allotted for equipment setup, calibration, and button measurements.

We performed calibration by repeated measurements of a single 2-kg plutonium button which contained 6% ^{240}Pu . Unfortunately the button had an unusual shape: both diameter and height were 5.21 cm. Such a configuration has a neutron multiplication of about 1.74. Generally, 2-kg buttons have a much lower profile, i.e., diameter-to-height ratios of about 5 to 1, and are characterized by neutron multiplication of about 1.5 (see Ref. 18, Table 9.3 and Ref. 14, Sec. E, pp. 20-23).

Table XXIV lists the kilograms of plutonium (by ARHCO lot ID, either DZA or NX) found by LASL for each button and the value assigned by ARHCO. The LASL-1 column lists the kilograms of plutonium found per button based on a comparison with the calibration button. The LASL-1 data are biased 17% lower than the ARHCO data. This size of bias would be expected for buttons of 5-to-1 diameter-to-height ratio. Notice that measurements for the DZA buttons are biased considerably more than those for the NX buttons. It turned out that

Fig. 27.

Verification measurement setup.

TABLE XXIV

LASL VERIFICATION MEASUREMENTS OF PLUTONIUM METAL BUTTONS

DZA Lot ID				NX Lot ID			
Button Number	Plutonium (kg)			Button Number	Plutonium (kg)		
	ARHCO	LASL-1	LASL-2		ARHCO	LASL-1	LASL-2
520	2.00	1.57	1.99	148	2.04	1.90	2.10
392	2.05	1.59	2.02	036	1.94	1.64	1.82
523	1.94	1.58	2.00	037	1.56	1.17	1.48
376	1.98	1.60	2.04	008	2.06	2.03	2.24
371	2.04	1.59	2.02	033	1.89	1.59	1.75
438	1.99	1.56	1.90	146	2.01	1.78	1.97
390	1.98	1.53	1.94	161	2.05	2.00	2.22
385	1.86	1.43	1.81	162	2.05	1.94	2.15
378	2.04	1.59	2.02	147	2.02	1.82	2.01
381	2.05	1.64	2.10	226	2.21	1.81	2.07
391	2.02	1.56	1.98	227	2.02	1.86	2.06
516	2.04	1.63	2.07	224	2.04	1.88	2.15
248	2.16	1.75	2.23	140	2.05	1.92	2.13
099	2.14	1.71	2.17	035	1.93	1.64	1.82
249	1.71	1.26	1.60	015	2.05	2.11	2.33
396	1.99	1.60	2.05	034	1.93	1.67	1.81
394	2.03	1.61	2.05	149	1.87	1.68	1.86
393	2.06	1.61	2.05				
442	2.04	1.63	2.07				
443	2.05	1.65	2.10				
435	2.02	1.62	2.06				
420	2.05	1.61	2.05				
421	2.00	1.57	1.99				
397	1.96	1.52	1.94				
080	2.11	1.63	2.07				

the DZA buttons had a higher diameter-to-height ratio than the NX buttons.

The LASL-2 data were calculated by calibrating the SNAP II detector against the entire group of DZA or NX buttons. Specifically,

$$C_{DZA} = \frac{\sum (\text{neutron counts}/30 \text{ s})_{DZA}}{\sum (\text{plutonium mass})_{DZA}}$$

$$C_{NX} = \frac{\sum (\text{neutron counts}/30 \text{ s})_{NX}}{\sum (\text{plutonium mass})_{NX}}$$

These calibration constants were then used to calculate the plutonium content of each button. Results are listed in the LASL-2 column.

Table XXV contains statistical information about the data in Table XXIV. Although this information

TABLE XXV

STATISTICAL INFORMATION FOR PLUTONIUM BUTTON MEASUREMENTS

Button Type	Number of Buttons	Relative Bias* (%)	Standard Deviation ^b
DZA	25	-20.3	0.025
NX	17	-9.7	0.068

*Relative bias = $\left\{ \frac{\sum [(LASL-1) - ARHCO]}{\sum (LASL-1)} \right\} \times 100$.
^bStandard deviation as calculated from (LASL-2)-ARHCO values, in kilograms of plutonium for each button.

does not reveal a discrepancy in the total shipment, individual buttons that deviate from listed values by more than 3σ can be identified.

If button diameter-to-height ratios are known within 25% and suitable matching standards are available, button verification measurements made with portable equipment may be possible to $2\sigma = 100$ g of plutonium.

B. DYMAC for New LASL Plutonium Facility

1. DYMAC Instrumentation (R. S. Marshall, V. S. Reams, and M. M. Stephens)

Approximately three-fourths of the new glove boxes for TA-55 have been installed. Glove-box and floor space have been allocated for DYMAC NDA instruments and associated electronics. Electronic bins usually are located beside their respective NDA instruments; however, in a few cases signals from the instruments will have to be sent to electronic racks across aisles, and wire-runs of about 15 m will be required.

Nine glove boxes have been modified for adaptation of neutron counter wells. The wells have been fabricated and will be installed during the next quarter. Most of the wells are 17 cm in diameter, sized to accept containers commonly used to transfer concentrated SNM within LASL glove boxes. Four 26-cm-diam wells are being installed to

permit measurement of large weapon parts and for analysis of large trash packages and dismantled equipment.

Most of the thermal-neutron well coincidence counters will have double range capability. Items displaying a very high neutron flux will be counted by approximately one-fifth of the ^3He tubes surrounding the sample well. Items displaying low neutron flux will be counted by all of the ^3He tubes. The smaller, 17-cm-diam wells will require 20 ^3He tubes, whereas the 26-cm-diam wells will require 30. The high- and low-efficiency counting arrays will be used to assay material categories listed in Table XXVI. Details of the ^3He tube array, cadmium, and polyethylene which make up the thermal-neutron coincidence detectors will be included in the January-April 1977 progress report.

Approximately 35 electronic balances will be used in the TA-55 DYMAC system. The balances will be either 0 to 2 kg or 0 to 15 kg with 0.1- or 1-g readout, respectively. All but three of the balances will be split into a sensing module (located within glove boxes) and a digital readout module (located outside glove boxes). The three remaining balances will be used as integrated units located entirely outside of glove boxes.

Two balances from one manufacturer have been under evaluation for several months. Balances from two other manufacturers are on order and their evaluation should begin during February 1977.

2. Computer Procurement for TA-55 (R. F. Ford,* J. Hagen, and C. Slocomb*)

The computer has been ordered and is scheduled for delivery in January 1977. It will be a Data General Eclipse Model C330 with 128k words of core memory, two 5-megaword disks, two 75 IPS 9-track magnetic tape drives, two system consoles, one 600 LPM line printer, and a 128-line communications multiplexor. Preinstallation meetings have been held with Data General and the TA-55 building personnel to ensure a smooth installation in the TA-55 computer room. The vendor has supplied schedules for training courses about INFOS data-base management software and RDOS system courses.

*Group E-5.

TABLE XXVI
MATERIAL CATEGORIES FOR NEUTRON COUNTING

Material Category	Low-Efficiency Array (g)	High-Efficiency Array (g)
Plutonium as plutonium metal, PuO ₂ , or Pu(NO ₃) ₄	>500	<500
Impure plutonium with high (α ,n) interference	>100	<100
²³⁸ Pu as plutonium metal or PuO ₂	>5	<5

Two people who have completed training on the INFOS system and one who has completed the RDOS system course have shared the course information with other members of the software group. At present, the group is designing the data-base structure for TA-55.

3. TA-55/DYMAC Communication System (R. B. Glascock,* R. F. Ford,* and R. S. Marshall)

The communication system for the DYMAC program to be installed at TA-55 will connect process line terminals and instruments to a computer to create a real-time nuclear materials control system. The numbers of terminals and instruments required for this control system are listed in Table XXVII, which also indicates possible expansion provided by the wiring system (spares). In all, 96 locations are to be implemented at this time. The computer multiplexor has 128 ports and an ultimate capacity of 256 ports for terminals or instruments (locations).

Figure 28 shows a block diagram of the system. Present plans call for a trunk cable for each wing. The number of pairs in the trunk is constant for the entire length of a wing, with junction boxes (three or four) along the trunk where all pairs are available for individual runs to each terminal or instrument. The availability of all pairs anywhere in a wing will allow

the maximum number of terminals to be located at any point in the wing. This feature also simplifies the construction of the trunk in a wing as all junction boxes in a wing are identical.

Supervisory stations, which consist of a hard-copy terminal (30 or 180 characters/s) plus a cathode-ray tube (CRT) terminal (480 characters/s), will be serviced by independent computer ports and wire pairs. This will allow each to operate independently and offer the flexibility to change the speed characteristics of either terminal by reprogramming the computer port. Thus the CRT could be used for other transactions while the hard-copy unit is printing a report.

The 20-mA current loop will be the signaling medium between the computer and the individual instruments and terminals. (This represents a change from the approach previously reported in Ref. 2, Sec. B, pp. 35-36.) This loop was chosen for maximum noise immunity and minimal equipment (i.e., no modems will be required). The distances (up to 120 or 150 m) and data rates (up to 4800 or 9600 baud) could be marginal using the RS-232C interface. Individually shielded pairs should be used to reduce crosstalk and errors due to extraneous noise sources.

The junction boxes provide minimal runs of cables to individual locations and reduce the size of trunk cable bundles. The actual number used will be determined during the design process. The

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TABLE XXVII
INSTRUMENT, TERMINAL, AND SPARE LOCATIONS

Wing	Instru- ments	No. CRTs	Hard-Copy Terminals		Spares	Total
			30 char/s	180 char/s		
1	11	8	1	---	47	67
2	3 ^a	2	1	---	48	54
3	15	9 ^b	2 ^b	1	40	67
4	34 ^a	7	2	---	38	81
Totals	63	26	6	1	---	269

^aIncludes an outlet for an instrument or terminal.

^bIncludes a CRT plus hard-copy terminal in Rooms 112 and 131.

Fig. 28.

TA-55 cable interconnections for terminals and instruments to the DYMAC computer.

number of trunks will depend on the route used and other considerations which may reduce the number of trunk lines to two.

Cables consisting of two pairs of wires will connect individual locations to the trunks at the junction boxes. The instruments and terminals will have a short cable (2 to 3 m long) with a four-prong phone plug to connect into the system at a mating phone jack. (Plugs and jacks are of a type used by telephone companies for extension phones in residential and commercial installations.)

The trunks will terminate in the DYMAC computer room on panels with connectors (24 pairs/panel). Enough cable will be provided to serve the totals listed in the last column of Table XXVII. These panels will be connected to the jackfield with cables, and outputs of the jackfield will use 24-pair cables with 24-pair connectors on one end and twelve 2-pair connectors on the other for connection to the computer multiplexor. The use of 24-pair equipment with connectors reduces the manual wiring by over 1500 pairs.

The jackfield (Fig. 29) serves as a maintenance panel and manual patching point. A failed MUX port can be replaced by a spare with a patch cord between the line side to a location and the spare MUX drop side. The monitor jack is always in parallel with the line side for monitoring purposes.

The layout of the jackstrips in the jackfield is shown in Fig. 30. The top two chassis will be test equipment, as indicated. The locations in each wing will be identified by a wing number and a series of numbers starting with 1 for each wing, thus 3-5 identifies location 5 in wing 3. An attempt has been made to arrive at a design in which the locations correspond to the MUX port number and are sequential within the jackfield. This could be implemented, but the sequential numbering would fail when additions are added.

Jackstrips will be installed to serve the number of locations listed on the right half of the parentheses in Fig. 30 (shown at right-hand side under Wing No.), but only the number of terminals/instruments

Fig. 29.
Forty-eight-position modular jackstrip.

NOTE. NUMBERS IN PARENTHESES FOR EACH WING INDICATE THE NUMBER OF TERMINALS AND INSTRUMENTS INITIALLY IMPLEMENTED TO THE NUMBER OF AVAILABLE LOCATIONS.

Fig. 30.

TA-55/DYMAC jackfield mounted in an equipment rack.

III. TECHNOLOGY TRANSFER (R. H. Augustson)

Attendees of the December 1976 in-plant NDA instrumentation course were introduced to DYMAC. Topics covered basic principles underlying a materials control system, considerations involved in the use and location of NDA equipment, fundamen-

tals of measurement control; and a detailed description of the type of computer hardware and software needed to implement the real-time aspect of such a system. We will offer a course in March 1977 which will extend and amplify these topics.

PART 3

PERIMETER SAFEGUARDS AND MATERIAL VERIFICATION

I. VERIFICATION SIGNATURE CALCULATIONS (H. F. Atwater)

A statistical method based on hypothesis testing has been applied to the comparison of multicomponent nuclear signatures. This method was specifically directed to the problem of comparing the two verification signatures obtained when nuclear material is placed in and removed from a storage vault. The basic problem is to determine, within an acceptable confidence limit, whether a sample removed from a particular vault location is the same sample that was previously stored in that location.

The statistical method is applicable to the general problem of comparing two multicomponent variables. As such, this method may also be useful in other Group R-2 problems, such as identification based on comparison of few-region neutron or gamma-ray spectra. Two advantages of the hypothesis testing method are the quantitative specification of a single confidence level and the inclusion of correlation effects between individual signature components.

To apply these signature comparison techniques, a minimum number of sample-in and sample-out signature measurements are required. The sample-in signature must be measured $M > p$ times, where p is the number of signature components. The sample-out signature must be measured $N \geq 2$ times. The sample-in signature measurements can

be made consecutively without moving the sample from its position in the verification station. The same situation exists when the sample-out measurements are made. As the number of signature measurements increases beyond the required minima, the probability of Type I and Type II²¹ errors decreases.

A computer program called TSQTEST has been written to perform signature comparisons using the T^2 statistic hypothesis testing method. This program was used to analyze signature data taken with eight plutonium samples. The TSQTEST calculations were made with the CDC 7600 computer and employed several fairly involved procedures such as double-precision matrix inversion and numerical evaluation of F distribution parameters. Such procedures require substantial computer capability for efficient execution and probably could not be easily performed with hard-wired portable equipment. Signature data acquired by portable detection equipment would probably have to be analyzed later at separate computer facilities.

The TSQTEST program will be used to analyze additional data obtained when the demonstration version of the verification station becomes operational.

II. REAL-TIME INVENTORY (N. Nicholson, T. H. Whittlesey)

The verification chamber has been assembled in Building I at TA-18, and is ready for installment of the neutron and gamma detectors, the 25-kg scale, and the associated electronic components.

Two MOD III prototype shelf monitors were assembled and checked out. They were then tied to a

digital data simulator that provided the input signals to the detectors. Detector outputs were observed on a scope monitor. Both detectors performed as designed. Since the new detector design offers a very workable computer-compatible system, 16 of these MOD III detectors will be built and used

in the 16-can demonstration system. These detector boards should be completed and ready for installation during the next reporting period.

The Data General NOVA 3/12 computer has been ordered and should arrive by the end of January 1977.

III. VERIFICATION STATION FOR THE 280-CAN SANDIA DEMONSTRATION SYSTEM (N. Nicholson, D. R. Millegan, T. H. Whittlesey)

Phase I. The Sandia verification station for Phase I has been completed and delivered to Sandia. Two BF_3 neutron detectors and a GM tube gamma detector were installed in the station, along with the associated preamps. The logic modules, HV power supplies, amplifier-discriminators, and scalers were

installed in a standard NIM bin and transferred to Sandia. This system will be integrated into the Sandia 280-can demonstration system and will remain there until it is replaced with the Phase II verification station.

IV. HAND-HELD PERSONNEL AND VEHICLE MONITORS (W. E. Kune, W. H. Chambers, C. N. Henry, S. W. France, G. M. Worth, R. D. Hastings, D. R. Milligan)

A light, rugged monitor, shown in Fig. 31, has been developed for SNM searches at material-access and protection-area exits. This monitor accepts gamma-ray pulses from a $\text{NaI}(\text{Tl})$ detector, integrates for a preset counting interval (typically 0.3 s), and produces an audible alarm whenever the counts in the interval exceed the trip level that is a preset multiple of the stored background count. Because the monitor is silent except when the alarm is operating, personnel with little special training can conduct more effective searches in a noisy and distracting environment than they can with conventional audible monitoring of individual radiation counts. The monitor is also more sensitive than conventional monitors that provide audible indication of the count rate.

The design of the instrument was sent to 12 commercial vendors for a request for price quotation. National Nuclear Corp. was the successful bidder and delivered the first of 63 units as an evaluation prototype in December 1976. The delivery of the balance of the order should begin in March 1977. These units have been ordered by LASL and several other facilities in the U.S.

Fig. 31.
Hand-held personnel and vehicle monitor.

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GLOSSARY

ALOO	Albuquerque Operations Office
ARHCO	Atlantic Richfield Hanford Co.
DNA	delayed neutron assay
DYMAC	dynamic materials control
FFTF	fast flux test facility
HEDL	Hanford Engineering Development Laboratory
HLNCC	high-level neutron coincidence counter
LASL	Los Alamos Scientific Laboratory
LWR	light water reactor
MIP	material in process
MTR	materials test reactor
NBL	New Brunswick Laboratory
NDA	nondestructive assay
PWR	pressurized water reactor
RR-II	Round Robin II
RSD	relative standard deviation
SAI	solution assay instrument
SGS	segmented gamma scanner
SNM	special nuclear material
SSAS	small sample assay station
TRU	transuranic
USAD	uranium solution assay device