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Nuclear Safeguards Research and Development

Program Status Report

September—December 1977

University of California

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Compiled by

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NUCLEAR SAFEGUARDS RESEARCH AND DEVELOPMENT

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ABSTRACT

This report presents the status of the Nuclear Safeguards Research and Development program pursued by LASL Safeguards Groups Q-1, Q-2, Q-3, and Q-4. Topics covered include nondestructive assay technology development and applications, international safeguards, perimeter safeguards and surveillance, concepts and subsystems development (e.g., DYMAC program), integrated safeguards systems, training courses, and technology transfer.

Salient features of the program and technical progress during the current reporting period are outlined in the Executive Summary.

CONTENTS

NUCLEAR SAFEGUARDS RESEARCH AND DEVELOPMENT

EXECUTIVE SUMMARY

G. Robert Keepin
Associate Division Leader for Nuclear Safeguards
and
Director of Nuclear Safeguards Programs

William H. Chambers
Assistant Division Leader for Nuclear Safeguards

The Nuclear Safeguards Program at LASL encompasses four technical groups: Q-1, Safeguards Technology, International Safeguards and Training; Q-2, Detection, Surveillance, Verification, and Recovery; Q-3, Safeguards Subsystems Development and Evaluation; and Q-4, Integrated Safeguards Systems and Technology Transfer. This program status report is divided into four parts, one for each of the Safeguards technical groups; each part begins with a brief overview of the Group's function and major programmatic efforts. Although the work of the four Safeguards Groups is closely interrelated, these program status reports are structured so that each Group contribution can, if necessary, be read independently of the others.

Inspectors at the International Atomic Energy Agency (IAEA) require the capability of nondestructive assay (NDA) techniques for determining the ^{235}U content of highly enriched uranium. Because of the very low spontaneous fission yields in the uranium isotopes, ^{235}U assay usually requires an active interrogation technique. The two widely used active systems, the random driver (RD) and ^{252}Cf Shuffler, are too bulky and complex to meet IAEA transportability requirements.

To meet the IAEA needs, LASL is taking two approaches in the development of relatively simple, rugged, and transportable instruments for the assay of uranium isotopes. One is the high-level neutron coincidence counter (HLNCC), a passive well counter, which is one of the most useful NDA instruments for plutonium assay in field applications. We investigated the feasibility of combining the HLNCC with an AmLi neutron interrogation source to measure uranium as well as plutonium and concluded that the HLNCC can be used for the assay of uranium samples but that the assay precision will be poor. The relative errors for a 1000-s measurement of 200-g and 400-g samples of ^{235}U are 15% and 10%, respectively.

In the second approach, an active well coincidence counter (AWCC) is being developed which uses the same principles as the modified HLNCC, namely, interrogation with an AmLi neutron source and counting of coincidence neutrons in ^{3}He detectors, in a more optimum configuration. We fabricated a prototype AWCC system and performed initial sensitivity tests, with encouraging results. The relative error for a 1000-s measurement of 200 to 400 g of ^{235}U is 2 to 3%.

Absorption-edge densitometry is being developed for simultaneous measurement of multiple concentrations of special nuclear materials (SNM) and source materials in a variety of chemical compositions, including solutions, ash, and fuel bundles. The method is based on the measurement of the transmission of gamma rays with energies just above and just below the K or L_{III} absorption edges. Because the discontinuity across the absorption edge depends upon discrete electron binding energies in the electron shell structure of the atom, it is element specific, not isotopic specific as in the case of passive gamma rays emitted from the nucleus. During this reporting period, an L_{III} absorption-edge densitometer was studied for future installation and in-line evaluation at the experimental coprocessing test location at the Savannah River Laboratory (SRL). The concentrations in the solutions to be assayed will range from 20 to 40 g U/ℓ with uranium-to-plutonium concentration ratios of 4 to 8. The densitometer uses an x-ray generator source and incorporates an advanced data

analysis technique which enables a precise determination of the minor plutonium component at the relatively high concentration ratios. Preliminary results indicate that for a 1000-s counting time, the uranium content can be measured to a precision of 0.5% and the plutonium content to 2%. Studies are also being carried out on the use of an energy-dispersive x-ray fluorescence (XRF) technique that would achieve high precisions in the NDA of lower density solutions, eliminate the need for an x-ray generator, and reduce the cost and complexity of the associated equipment.

The investigation of neutron time correlation techniques has been extended to a wider range of samples including metals, oxides, and a carbide. An expression has been identified involving the reduced variance parameters which correlate well with the real coincidence counts for a given type of material, e.g., groups of metallic samples or groups of nonmetallic samples. Attempts to prove a single correlation for all sample types have been unsuccessful.

A reactor power monitor is being developed to meet the IAEA needs for a continuous independent record of the operating power of nuclear reactors under inspection. A monitor incorporating thermal-neutron sensors and a microprocessor-based data acquisition and analysis system has been designed. The hardware for the prototype unit is nearly complete, and the software is being written. Because installation of the DYMAC system at TA-55 has been slowed by slippage in the building construction schedule, plutonium did not enter the facility during this reporting period. Installation did proceed far enough in the advanced carbide fuels laboratory to allow extensive testing of the accountability system and to provide a basis for training the process operators.

An exercise, "Project Sandbag," was performed to test the transaction system for the advanced carbide fuels process and to acquaint DYMAC personnel with the system. Cans of sand were used to simulate SNM, and all components of the DYMAC Software System (DYSS) were tested. Several "bugs" in the program were discovered and corrected, and the exercise confirmed the satisfactory operation of the DYSS.

The safeguards concept-definition report, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility," was completed in draft form and circulated for review. The report (LA-7011) is part of a continuing effort to establish design and performance requirements for cost-effective, integrated safeguards systems for typical nuclear fuel cycle facilities and to assess the impact of these requirements on future plant design criteria. The conversion facility is designed to couple the previously studied fuel reprocessing plant (see LA-6881) and mixed-oxide fuel fabrication plant (see LA-6536). The materials management systems are compatible in all three facilities; they reinforce each other and, when combined, could share the safeguards responsibilities in a reprocessing complex where the components of more than one facility are collocated.

The conversion facility is unique in the civilian power economy because it invariably handles large amounts of concentrated highly purified plutonium in a form that could be an attractive target for diversion or national misappropriation. Our study shows that strategic quantities of SNM can be safeguarded in a high-throughput, nitrate-to-oxide conversion facility at reasonable cost with minimal process disruption.

During the reporting period, safeguards technical support was provided to the Nuclear Production Division's (NPD) alternative fuel cycle technology programs, the Alternative Fuel Cycle Technology (AFCT) program, and the Thorium Fuel Cycle Technology (TFCT) program. The intent of these programs is to evaluate the relative nonproliferation advantages (or disadvantages) of several alternative modes of operations for existing and proposed fuel cycle facilities and to assess their impact on existing fuel cycle facility designs.

Large, fast-critical assembly research facilities, used to simulate plutonium-fueled breeder reactors, are operating in many of the developed countries of the world. These facilities present a unique safeguards problem in that their inventories can include thousands of fuel pieces constituting tons of fissile material in relatively pure form. During this reporting period a draft report, LA-7028-MS, was issued outlining preliminary concepts for materials measurement and accounting in critical facilities and identifying several promising inventory techniques. This report is an initial milestone in a continuing investigation of these facilities.

LASL safeguards staff members have trained, briefed, and transmitted technical data to representatives from various US industrial nuclear facilities as well as domestic, foreign government, and international agencies. Two courses conducted at LASL as part of the DOE Safeguards Technology Training School had an enrollment of 33 domestic and 28 foreign attendees. Safeguards bilateral meetings included presentations and discussions with representatives from Norway, Denmark, Japan, Greece, Belgium, and South Korea.

PART 1

SAFEGUARDS TECHNOLOGY, INTERNATIONAL SAFEGUARDS AND TRAINING

GROUP Q-1

Roddy B. Walton, Group Leader

H. O. Menlove, Alternate Group Leader

Group Q-1 is responsible for the development and application of a broad range of measurement techniques for the nondestructive assay (NDA) of fissionable materials in the many physical and chemical forms found in the nuclear fuel cycle. Specific applications of NDA technology currently under development by Group Q-1 include: plutonium process materials at DOE facilities, highly enriched uranium, enrichment process materials, and spent fuel reprocessing. Prototype instruments for these applications are developed, calibrated, and tested in operating plant environments. The Group Q-1 NDA technology program provides technical support to material control demonstration systems, such as dynamic materials control (DYMAC), and a base of measurement technology for integrated safeguards system studies.

To implement the transfer of NDA technology to various types of plants and facilities in the nuclear community, Group Q-1 is a major contributor to the DOE Safeguards and Technology Training Program, which is offered to safeguards inspectors (both domestic and international) and qualified government and industry personnel. Additional Q-1 efforts to facilitate technology transfer include briefings, consultations, and issuance of design documentation, operations manuals, interlaboratory assay comparisons, and consensus standards.

A major Q-1 effort is technical assistance to IAEA international safeguards. This effort includes development of portable NDA instrumentation to address specific inspection situations, as well as associated calibration and evaluation of this instrumentation, and personnel training.

I. SPENT FUEL ASSAY

Nondestructive Gamma-Ray Analysis of Highly Enriched Spent MTR Fuel (J. R. Phillips, S. T. Hsue, K. Kaieda, E. G. Medina, and L. R. Cowder)

We are investigating passive gamma-ray techniques for the accurate determination of burnup in highly enriched irradiated fuels. The fuels that we studied had been irradiated in a materials testing reactor (MTR) to ~30% burnup and cooled for 1 to 5 yr. Previous studies of MTR fuel elements show 5 to 9% average differences between operator-declared burnup values and passive gamma-ray-measured burnup values.[1,2] One method of applying the passive gamma-ray technique involves the measurement of the isotopic ratio of two fission products. If one of the fission products is proportional to the square of the thermal neutron flux and the second is proportional to the flux to the first power, the ratio is proportional to the thermal flux and, in principle, can be used as a burnup monitor. Two nuclides, ^{134}Cs and ^{154}Eu, are formed from the neutron activation of fission products ^{133}Cs and ^{153}Eu, respectively, and are therefore approximately proportional to the flux squared. The ratios of the activity of each of these two isotopes to the ^{137}Cs activity (a direct fission product proportional to the neutron flux) may be used to obtain a correlation with fuel burnup in

MTR elements. We measured isotopic ratios both as a function of axial position along the fuel element and for the complete fuel element.

Figure 1 shows the experimental apparatus, which consists of a Ge(Li) detector assembly and a watertight collimator positioned over a fuel rod. By

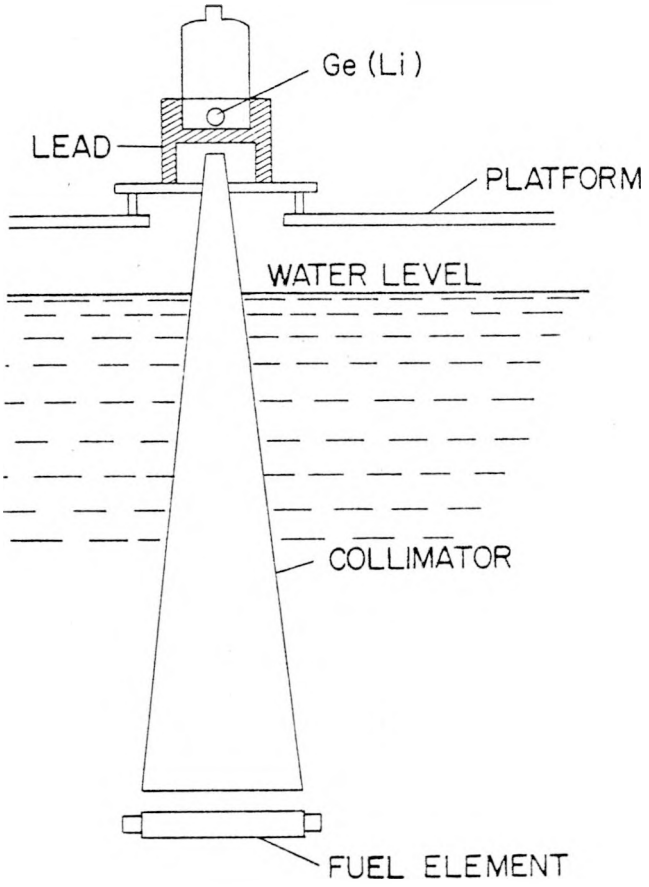


Fig. 1.
Experimental arrangement for measuring MTR spent fuel elements.

a rotation of 90°, the same collimator can be used both for axial scanning and for observing the complete fuel element. Figure 2 shows relative activity profiles for ^{137}Cs, $^{134}Cs/^{137}Cs$, and $^{134}Cs/(^{137}Cs)^2$. The $^{134}Cs/^{137}Cs$ activity ratio is higher than the corresponding ^{137}Cs activity at all axial positions (except the normalization points). This deviation can be explained by the fact that the ^{134}Cs production is sensitive to the epithermal flux because of epithermal resonances in the $^{133}Cs(n,\gamma)^{134}Cs$ reaction. The $^{134}Cs/(^{137}Cs)^2$ plot should be constant at each axial position if both the $^{134}Cs/^{137}Cs$ ratio and the ^{137}Cs activity are proportional to the thermal flux.

In previous studies by other investigators, the fuel elements were measured only at the midpoint. Our axial scans indicate that the position sampled is important. By collecting gamma-ray spectra for the entire fuel element, one avoids the problem of selecting a representative section. We measured 10 complete MTR fuel elements whose declared burnup values based on calculations ranged from 61.36 to 73.87 g ^{235}U. The least-squares regression lines with 95% confidence bounds for the $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ isotopic ratios related to grams of ^{235}U burnup are presented in Figs. 3 and 4, respectively. The bounds may be interpreted as defining a region within which one is 95% confident that an individual burnup value will fall for a specifically measured ratio. The results indicate that the burnup of MTR fuel elements can be predicted with a 2.1 and 3.3% relative standard deviation (1σ) using the $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ ratios, respectively.

The information obtained from these exercises will be applied to the nondestructive measurement of burnup in Light Water Reactor (LWR) fuel elements and assemblies.

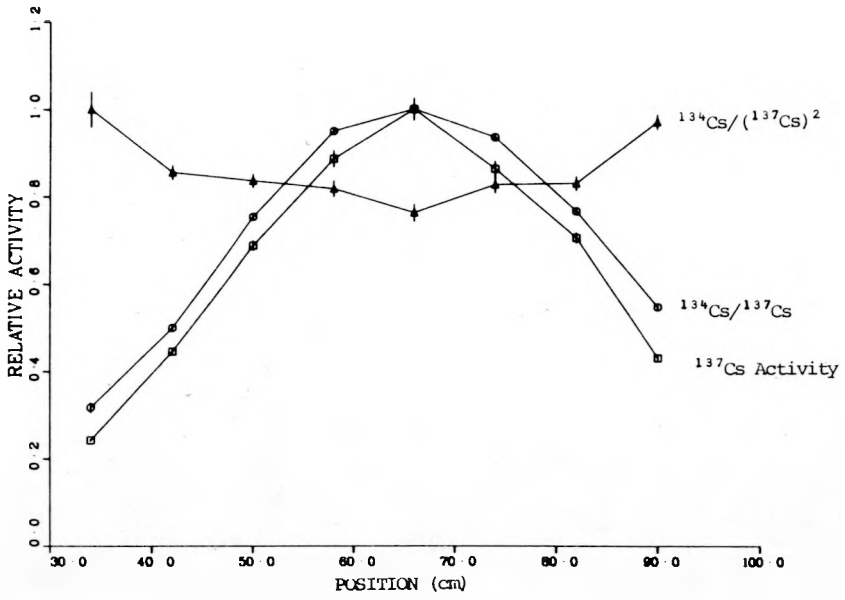


Fig. 2.
Axial distributions of the relative ^{137}Cs activity and the $^{134}Cs/^{137}Cs$ and $^{134}Cs/(^{137}Cs)^2$ activity ratios for an irradiated MTR fuel element.

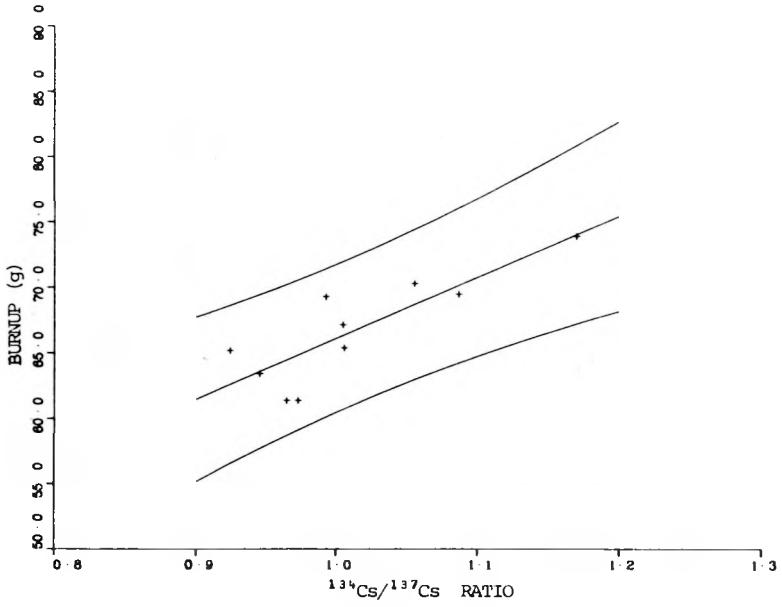


Fig. 3.
The 95% confidence bounds for the $^{134}Cs/^{137}Cs$ activity ratios vs grams of ^{235}U burnup over a range from 61.36 to 73.87 g.

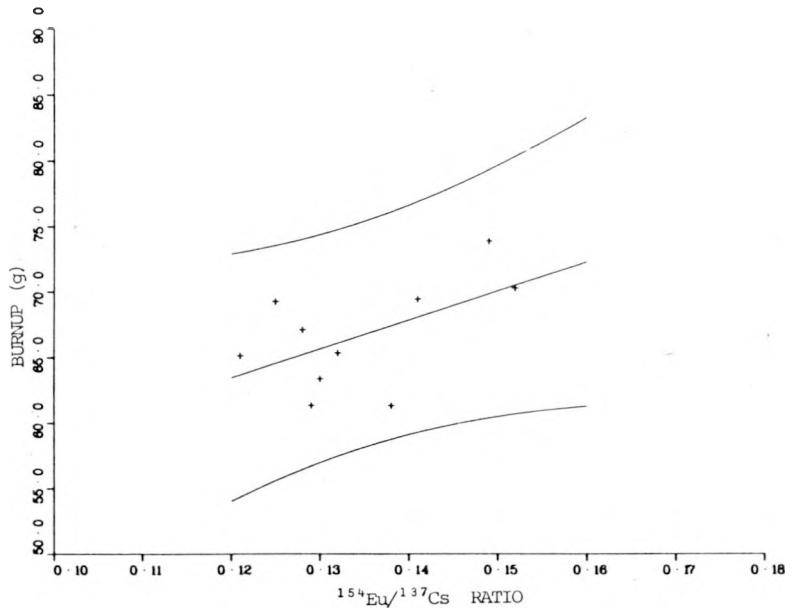


Fig. 4.
The 95% confidence bounds for the $^{154}Eu/^{137}Cs$ activity ratios vs grams of ^{235}U burnup over a range from 61.36 to 73.87 g.

II. NEUTRON ASSAY TECHNIQUE DEVELOPMENT

A. Active Well Coincidence Counter (H. O. Menlove, N. Ensslin, C. R. Hatcher, E. Medina, and J. Foley*)

1. Prototype Active Well Coincidence Counter Design

An active well coincidence counter (AWCC) (Ref. 3, p. 21) is being developed for assay applications to uranium samples, including high gamma-ray background materials such as ^{233}U-Th fuels of special interest for alternative fuel cycle concepts. The counter uses an AmLi source of low intensity ($\sim 10^4$ n/s) to interrogate the assay sample in a ^3He well counter that detects coincidence neutrons from the induced-fission reactions.

Figure 5 is a schematic of an AWCC recently fabricated from a design optimized for counting the induced-fission reactions and discriminating against the lower energy AmLi interrogation background neutrons. The sample cavity is lined with a 2.54-cm-thick nickel reflector to give a more penetrating neutron interrogation. The CH_2 moderator and cadmium sleeves are designed to give maximum efficiency for counting the induced-fission spectrum neutrons and at the same time have a low efficiency for counting the (α,n) neutrons from the AmLi interrogation source. The optimum CH_2 moderator thickness between the sample and the ^3He tube is ~5 cm for fission neutrons and ~2 cm for AmLi neutrons. Thus, the thick CH_2 in the end plugs and wall of the well detector forms a neutron shield to reduce the AmLi counting efficiency, ϵ_α. The CH_2 in the central region of the detector is removed to form an open channel from the sample position to the ^3He tubes, resulting in a high efficiency for fission-neutron counting, ϵ_f.

The total (singles) counting rate is given by

$$T \simeq \epsilon_\alpha S + \text{sample contribution} \quad ,$$

where S is the AmLi (α,n) source intensity. For most uranium assay applications, the sample contributes a negligible fraction of the total neutrons and the

*International Atomic Energy Agency.

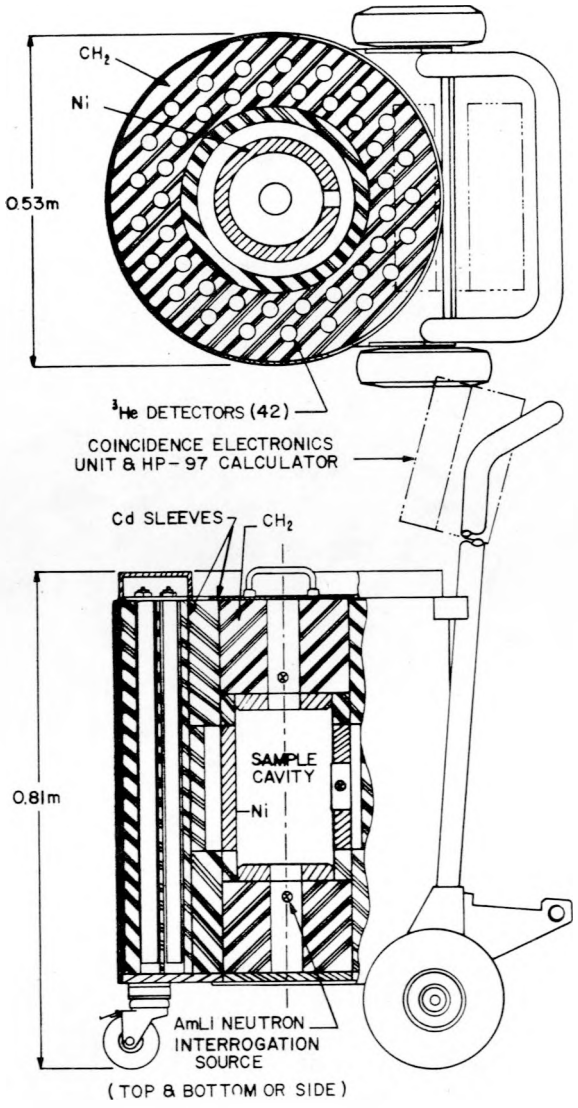


Fig. 5.
Schematic of laboratory prototype of the AWCC. The interior components of nickel, CH_2, and cadmium are removable for laboratory tests and evaluation.

sample term can be neglected. The accidental coincidence rate A is given by

$$A = GT^2 \simeq G(S\epsilon_a)^2 \quad ,$$

where G is the coincidence gate length. For most applications, the minimum error is achieved when the gate length G is set equal to 1.2 times the neutron lifetime τ of the detector. To first-order approximation, the real coincidence rate R is given by

$$R \simeq KUS\epsilon_f^2 \quad ,$$

where U is the mass of ^{235}U and K is a constant that includes the fraction of real events in the coincidence gate length G or 1.2 τ, the fission cross section, the number of neutrons emitted per fission, and the geometric coupling between the interrogation source and the sample. The net signal-to-background ratio is given by

$$\frac{R}{A} = \frac{KU}{1.2\tau} \cdot \frac{1}{S} \cdot \left(\frac{\epsilon_f}{\epsilon_a}\right)^2 \quad .$$

The smaller interrogation sources, S, will give higher signal-to-background ratios. Also any improvement in the $\epsilon_f/\epsilon_\alpha$ ratio will be squared in its effect on the signal-to-background ratio.

The relative statistical error in the measurement is given approximately by

$$E = \frac{\sqrt{R + 2A}}{R} = \frac{\sqrt{KUS\epsilon_f^2 + 2.4\tau S^2\epsilon_a^2}}{KUS\epsilon_f^2} \quad . \qquad (1)$$

For large interrogation source intensities ($S \geq 10^5$), the R term under the radical is negligible and the relative error simplifies to

$$E = \frac{\sqrt{2.4\tau}}{KU} \cdot \frac{\epsilon_a}{\epsilon_f^2} \quad ,$$

and it becomes apparent that significant error reductions can be made by increasing the efficiency for counting the induced fissions, ϵ_f. For instance, if by selective CH_2 shielding and neutron moderation, ϵ_α is reduced by a factor of 2 and ϵ_f increased by a factor of 2, the relative error would be reduced by a factor of 8. Stated differently, the assay time to obtain a given precision would be reduced by a factor of 64.

To evaluate the influence of the source strength S on the relative error, we calculated Eq. (1) for different values of S. The constants K, ϵ_f, and ϵ_α were determined from measurements with the high-level

neutron coincidence counter (HLNCC) (Ref. 3, p. 18) as described in the following section. Figure 6 shows the relative error vs S for the HLNCC and the estimated values for the AWCC shown in Fig. 5. Source strengths as low as ~10^4 n/s can be used with little loss in assay precision and the error is independent of S for values of S greater than 10^5 n/s. A 1000-s measurement for a 200-g ^{235}U sample results in a relative error of ~15% for the HLNCC and 2 to 3% for the AWCC.

2. Adaptation of the HLNCC for Neutron Interrogation

Three HLNCC units have been delivered to the International Atomic Energy Agency (IAEA) for the passive counting of plutonium samples. To adapt these units to the measurement of uranium as well as plutonium, we made a series of experiments using an AmLi neutron interrogation source combined with the HLNCC.

The measurements were carried out with two new AmLi sources that emit 2.4 x 10^4 n/s and 1.2 x 10^5 n/s. Because these sources are smaller than the 5 x 10^5-n/s sources usually used for active assays, it is possible to confirm the dependence of relative error on source strength given by Eq. (1). The optimum source strength and optimum geometry for the HLNCC were then determined by assaying 50-, 100-, and 200-g samples of U_3O_8 (93% ^{235}U) mixed with graphite.

Measurements were made first with the small interrogation source mounted in the side of the well

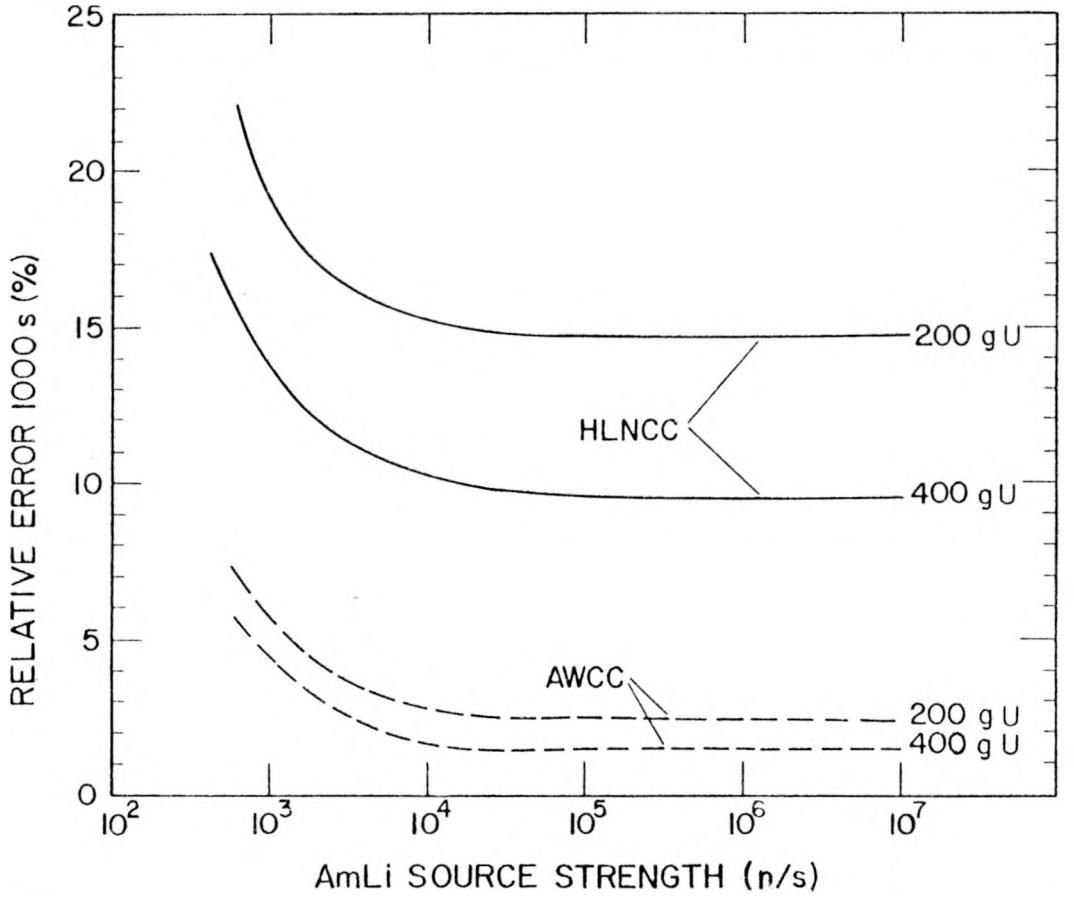


Fig. 6.
Relative precision as a function of neutron interrogation source strength for the HLNCC and the AWCC. The HLNCC results are normalized to measurements at a source strength of 2 x 10^4 n/s, and the AWCC curves are from calculations.

counter. One of the six sides of the detector was replaced by a specially made slab having a nickel-backed source chamber and no ^{3}He tubes. In this geometry the efficiencies for counting source neutrons and fission neutrons were nearly equal and the relative error was high—about 40% for 200 g of uranium assayed for 1000 s. When 2.54 cm of CH_2 were placed in front of the neutron source to obtain a thermal-neutron interrogation, the relative error decreased to 5% but the response was very nonlinear with mass of uranium. The side-mounted source geometry seems unfavorable except for samples larger than ~1 kg.

Measurements then were made with the interrogation source mounted beneath the uranium sample and surrounded by polyethylene to approximate the geometry of Fig. 5. In this configuration, ϵ_α was 4% and ϵ_f was 11%. The coupling constant K was about 3 x 10^{-5}/g. This constant is very sensitive to the distance between the source and the sample. The relative error for 200 g was ~15%, as illustrated in Fig. 6.

We conclude that the HLNCC can be used for the assay of uranium samples, but the assay precision will be poor. The absolute error of a 1000-s measurement will be 20 to 40 g, implying an inaccurate assay of small samples. This error can be decreased substantially by thermalizing the interrogating source, but that would make the response nonlinear and subject to self-shielding for high-enrichment uranium. To minimize geometrical effects, the HLNCC will be equipped with two end caps, each containing a 10^4-n/s AmLi source.

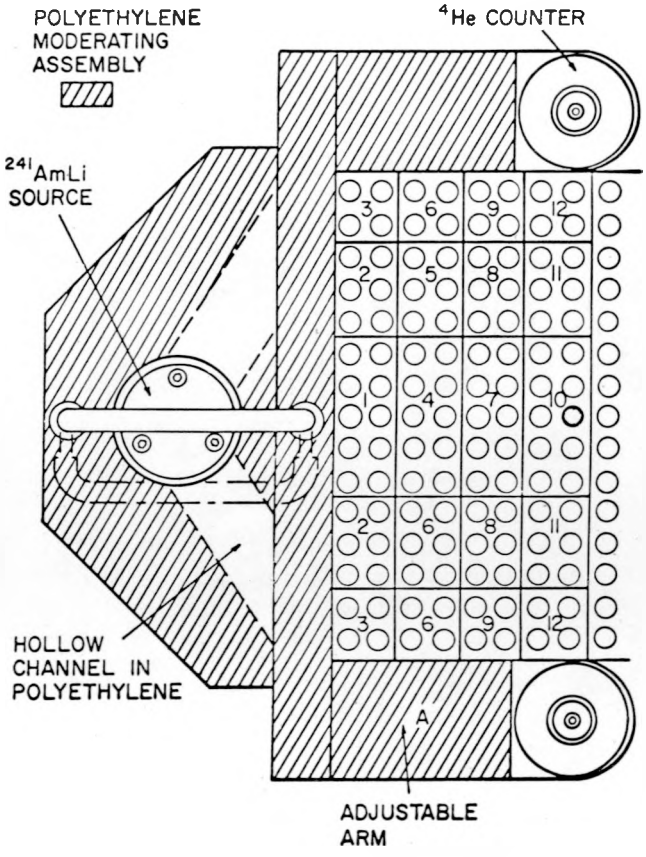


Fig. 7.
PWR fuel assembly assay system. Small circles represent individual fuel rods in part of a 15-by-15 rod assembly. Numbers represent areas used in testing the sensitivity of various regions for fissile material (Table I).

B. IAEA Portable Neutron Assay System for Unirradiated Fuel Assemblies (J. D. Brandenberger, H. O. Menlove, C. R. Hatcher, and E. G. Medina)

During this reporting period we continued to test the assay system on the pressurized water reactor (PWR) mockup fuel element (Ref. 3, p. 27), developed methods for its calibration, and wrote a first draft of the PWR instruction manual. The geometry of the assay system is shown in Fig. 7. We made measurements to determine which detector energy discrimination level gives the best assay precision. We also studied the problem of reproducing this discrimination level—the basic problem of calibration. Calibration methods were developed for a system with discrete electronic modules consisting of an amplifier, discriminator, and scaler and for a system using a Salina BS 27/N multichannel analyzer. Figure 8 shows the number of assay counts of a PWR fuel assembly as a function of small changes in calibration discrimination level for a 2000-s assay. For either a 1% instability or 1% error in determining the calibration discrimination level, the change in counts in a PWR assay is approximately equal to the statistical uncertainty, σ, of the 2000-s assay.

Table I gives the sensitivity of the assay system for detecting missing fuel rods from the various regions of the PWR fuel assembly indicated in Fig. 7.

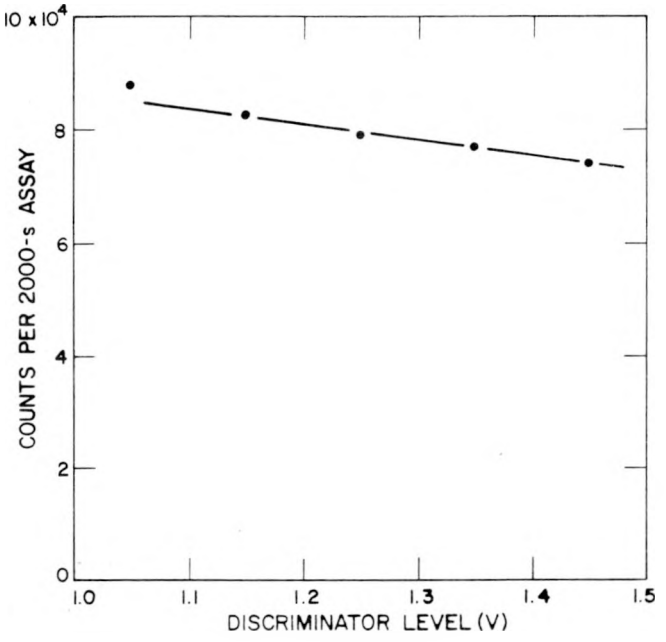


Fig. 8.
Sensitivity of a PWR fuel assembly assay to small changes in the calibration discriminator level of the ^{4}He detectors. Small changes from a desired calibration discrimination level of 1.25 V could occur because of imperfect calibration or electronic instability.

TABLE I

SENSITIVITY OF ASSAY SYSTEM FOR DETECTING MISSING FUEL RODS

Fuel Assembly Region (from Fig. 7)	Minimum No. Rods Missing for Detectability at 95% Confidence Level	Percentage of Rods Missing for Detectability at 95% Confidence Level
1	0.6	0.3
2	0.9	0.4
3	0.8	0.4
4	1.9	0.8
5	1.6	0.7
6	1.0	0.4
7	3.1	1.4
8	2.4	1.1
9	1.7	0.8
10	5.5	2.4
11	3.4	1.5
12	1.9	0.8

Three portable neutron assay systems were fabricated, and the AmLi neutron sources were received from Mound Laboratories. The units were modified for greater adjustment in the supporting arms to fit heavy water assemblies as well as the PWR and boiling water reactor (BWR) assemblies. The IAEA has requested that two of the units be delivered to IAEA, Vienna, and that we demonstrate one unit in collaboration with IAEA staff at a US fuel fabrication facility.

C. Cycle Time Selection for Delayed-Neutron Production from Cyclical Irradiation/Counting Sequences (G. W. Eccleston and H. O. Menlove)

Assay determination of unknown samples that is based on delayed-neutron counting requires an irradiation period for precursor formation, to be followed by a counting period during which the irradiation source is absent. Generally, a cyclical sequence of irradiation and counting is performed so that sufficient data can be collected to reduce the variance of an assay to acceptable levels.

Proper selection of irradiation/counting periods can increase the delayed-neutron yield during the counting cycle, producing improved statistics while reducing the overall measurement time. Selection of cycle times for previous ^{252}Cf Shuffler designs was based on a set of measured ^{235}U delayed-neutron yields for various duty cycles (irradiation time/irradiation + counting time) over a range of cycle periods (Ref. 4, p. 23). Those results are applicable only to measurements of ^{235}U irradiated by a fast-neutron source (^{252}Cf source neutrons were degraded below the fission threshold of ^{238}U). In general, the similarity between the relative half-lives and yields of corresponding delayed-neutron groups of the fissile isotopes permits these data to be used also for selection of measurement times for ^{233}U and ^{239}Pu. The data, however, are not applicable to the fertile isotopes of ^{238}U and ^{232}Th. Also, if the energy of the irradiation source is reduced to near-thermal energies or if the transfer time of the source to and from the sample is significantly altered, the measured data of Ref. 4 will not adequately describe the relative delayed-neutron yield as a function of cycle time.

The following general expression was derived to permit direct calculation of delayed-neutron group yields for any specified cycle times:

$$Y_i = \bar{\nu}\Sigma_f \phi(t)\beta_i \frac{\left(1 - e^{-\lambda_i t}\right)\left(1 - e^{-\lambda_i (t+T_I)}\right)}{\lambda_i^2 t} \cdot \left[\frac{1 - e^{-\lambda_i T_C}}{1 - e^{-\lambda_i T_P}}\right] e^{-\lambda_i T_D} ,$$

where

Y_i = yield of delayed neutrons from group i per cycle during counting window T_C,

$\bar{\nu}\Sigma_f\phi(t)\beta_i$ = production rate of delayed neutrons from group i during the irradiation,

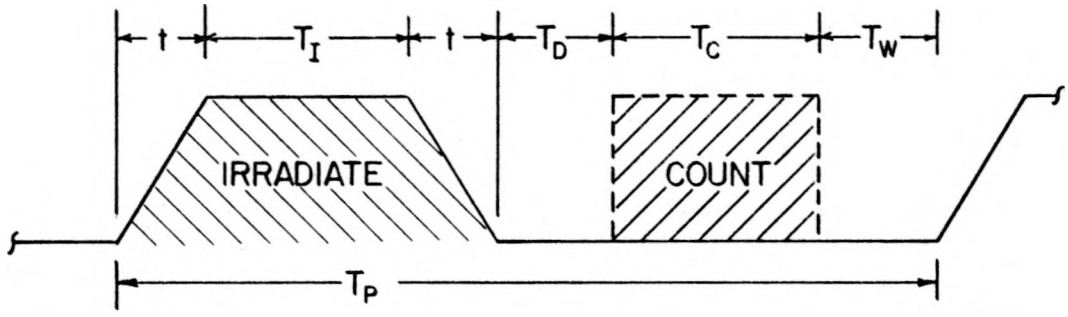
λ_i = decay constant of group i,

$t, T_I, T_D, T_C, T_W, T_P$ = cycle parameters of Fig. 9.

With the above equation one can calculate the number of delayed neutrons appearing during the counting time (Fig. 9) for a sample that is being irradiated cyclically by a trapezoidal flux pulse $\phi(t)$.

A comparison of calculated relative delayed-neutron yields with the measurements of Ref. 4 is shown in Fig. 10. The calculations were based on the six-group delayed-neutron yield and decay constants measured by Keepin et al. (Ref. 5, pp. 82-92). The measured data had a total source transfer time of 0.45 s. The source was moved through a shielded region and was assumed for calculational purposes to contribute negligibly to sample irradiation during the outer 2/3 (0.3 s) of its transfer time. The inner 1/3 (0.15 s) of source transfer was assumed to contribute in a linear manner to sample irradiation, reaching a maximum at the irradiation position. The flux pulse therefore was assumed to be trapezoidal with t = 0.15 s and $T_D = T_W = 0.3$ s. A 67% duty cycle with a 12.9-s period would have an irradiation time (T_I) of 8 s and a counting time (T_C) of 4 s with the delay, wait, and transfer times as specified above.

The data of Fig. 10 show close agreement between the measured and calculated relative delayed-neutron yields for periods of 4 s and longer. At



t = Linear ramp time during which the irradiation flux is changing because of flux transfer.
T_I = Irradiation time.
T_D = Delay time from end of irradiation to counting.
T_C = Counting time.
T_W = Wait time from end of counting to irradiation
T_P = Total cycle time ($t + T_I + t + T_D + T_C + T_W$).

Fig. 9.
Irradiation and delayed-neutron counting cycle for a ^{252}Cf Shuffler assay system with a trapezoidal flux pulse.

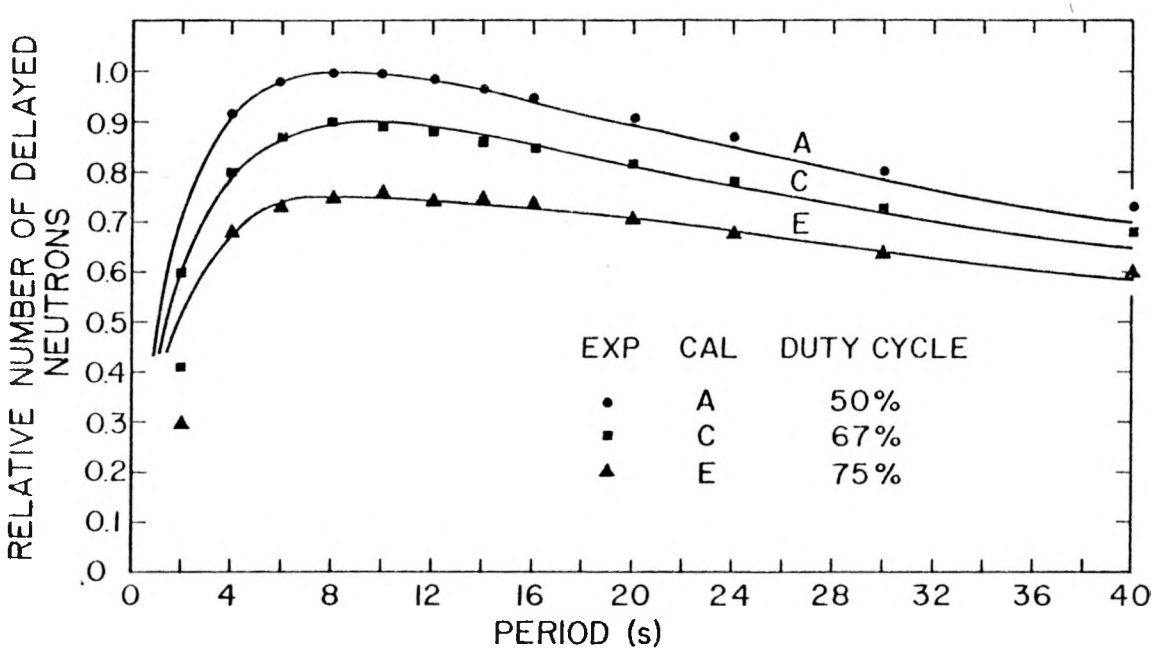


Fig. 10.

Calculated and measured relative delayed-neutron yields for fast fission of ^{235}U for various periods and duty cycles. Solid curves were calculated from data in Ref. 5; measured data are from Ref. 4.

shorter periods the calculations significantly overestimate the relative yields obtained experimentally. The discrepancy is due to the approximation of the flux pulse shape during time t, which overestimates the flux on the sample in comparison to the experiment. The result is an increased calculation of delayed-neutron production at shorter periods, producing a higher relative yield.

The effect of using thermal delayed-neutron parameters to calculate relative delayed-neutron yields from ^{235}U fission is shown in Fig. 11 for a 50% duty cycle. A significant reduction in relative yield for the longer cycle periods is evident for thermal-fission data in comparison to the fast-fission data. The use of the experimental data of Ref. 4 for the selection of cycle times should be limited to fast fission of the fissile isotopes. For mixtures of nuclides at various source energies and cycle times, the above equation will permit calculations of a proper set of curves to be used for cycle time selection.

D. Shuffler Assay Time Correction Factor and Estimation of the ^{252}Cf Source Transfer Time Error (T. W. Crane)

In the normal operation of the Shuffler assay system, the cycle time is comparable to or shorter than the lifetime of about 40% of the delayed-neutron precursors (Ref. 6, p. 13, and Ref. 7). In this mode of operation a buildup of delayed neutrons occurs with each cycle until eventually an equilibrium condition is reached. If the standards and the samples are assayed identically, the buildup does not affect the assay. However, it would be convenient to assay the standards longer than the samples to improve the accuracy on the calibration constants. In addition, a high throughput is necessary at the fuel fabrication facility at the Savannah River Plant (SRP). Thus, if each sample were assayed just long enough to obtain the required precision, the Shuffler could be used more efficiently.

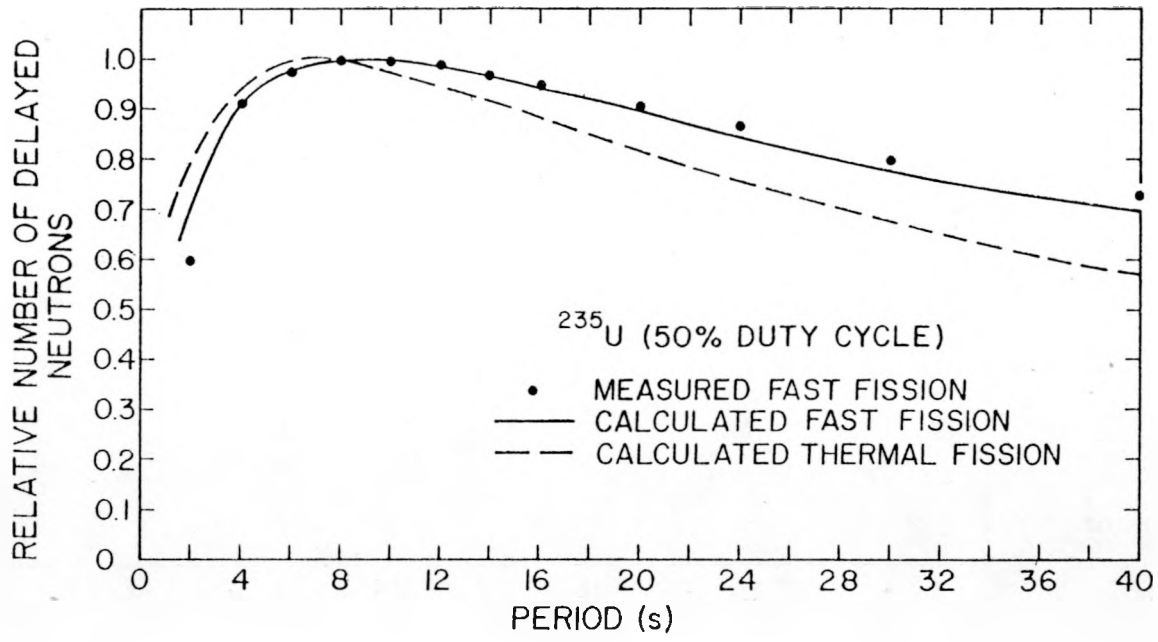


Fig. 11.
Comparison of relative delayed-neutron yields between fast and thermal fission of ^{235}U for various periods and duty cycles.

To accurately measure samples for assay times different from those used for the standards, a correction factor based on fast neutron kinetics must be applied (Ref. 8, p. 13; Ref. 9; and Part 1.II.C). The observed count rate for assay times relative to equilibrium is plotted in Fig. 12. If the response from both the standard and the sample is corrected to the equilibrium value, the overall correction for assay times longer than 200 s is no more than 4%.

During the transfer of the source to and from the interrogation well, a variation in the transfer time could affect the delayed-neutron count rate and thus the assayed mass. The effect of a variation in the transfer time from a typical operating condition (9.55 s irradiate, 9.55 s count, 0.45 s transfer) is plotted in Fig. 13. For small variations in the transfer time (ΔT_t), the relative change in the delayed-neutron response is given by

$$\frac{\Delta R}{R} \cong 0.14\,\Delta T_t \quad ,$$

where ΔT_t is in units of seconds. The transfer of the ^{252}Cf is driven by a stepping motor with stepping-time phase locked to a 16.384-MHz crystal oscillator.[10] The final step rate before stopping is 500 steps/s. Thus, the arrival time should be controllable to within ±2 steps, and ΔT_t would be <0.004 s. This maximum uncertainty in the transfer time would result in a systematic error of only 0.06%. The source transfer therefore is not expected to make a significant contribution to the error in the Shuffler's nondestructive assay (NDA) measurements. In addition, a neutron flux monitor at the sample assay position will pick up any flux variations or system malfunctions.

E. Random-Driver Studies (D. M. Lee, N. Ensslin, C. Shonrock, and T. Van Lyssel)

To understand more fully the physics of the random driver (RD), we made a series of measurements to determine the neutron threshold, the gamma-ray sensitivity, and the physical process that produced the sensitivity to the AmLi source.

With the AmLi source placed in the RD, the linear signals from both phototubes on one of the scintillators were ORed together and pulse-height analyzed. The results are shown in Fig. 14. The

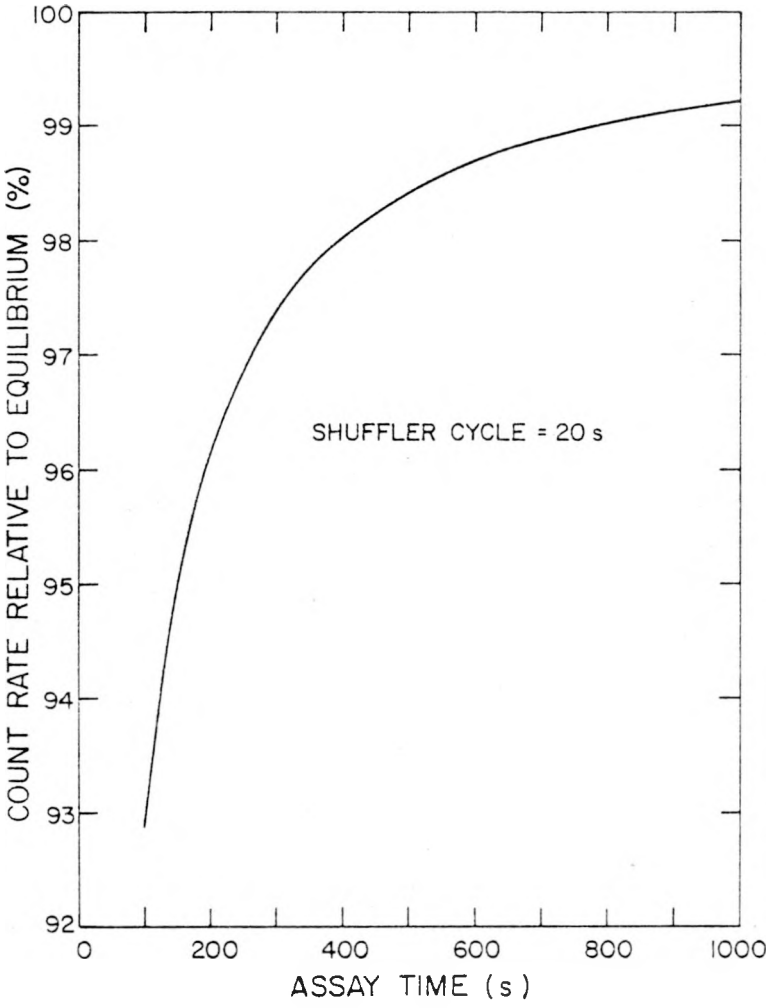


Fig. 12.
Relative count rate for ^{235}U delayed-fission neutrons in a ^{252}Cf Shuffler assay system. A continuous curve is drawn although the function is evaluated only for integer numbers of cycles.

calibration shows that the dominant source peak is at 1.97 MeV (electron equivalent), which corresponds to either a 2.2-MeV gamma ray or a 4.9-MeV neutron. Because the distribution resembles a typical Compton spectrum and no monoenergetic 4.9-MeV neutron is expected, we conclude that a significant fraction of events seen by the RD is due to source neutrons undergoing a $n + p \rightarrow d + \gamma$(2.2 MeV) reaction in the scintillator. Therefore, even though the source neutrons (0.5 MeV) are below the threshold of the scintillator (see below), the RD is still sensitive to slow neutrons via a neutron-capture reaction.

By use of the calibration from Fig. 14, the threshold was determined to be 0.095 MeV (electron equivalent). The relation[11]

$$T_p = \left(\frac{T_e}{0.18}\right)^{2/3} ,$$

where T_p = proton energy and T_e = electron energy, gives a calculated neutron threshold of 0.655 MeV. The efficiency for neutron detection between 2 and 5 MeV is then ~38% (Ref. 12). The electron threshold corresponds to the Compton-scattered electron from

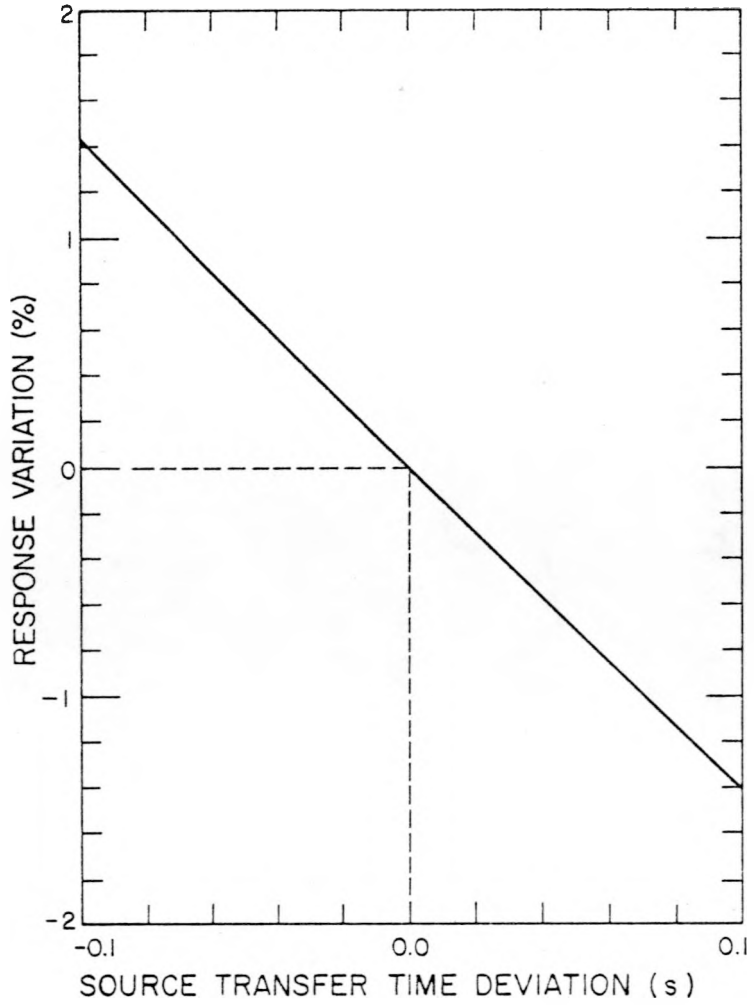


Fig. 13.
Relative variation in response as a function of the change in source arrival time. A negative time corresponds to the source reaching the interrogation well sooner than expected and a positive time corresponds to late arrival of the source.

a 220-keV gamma ray. Therefore, the gamma threshold is 220 keV.

A 5-cm scintillator has a relatively high efficiency (~15%) for Compton-scattering 1-MeV gammas; consequently, all types of coincidences should be possible: n-n, n-γ, and γ-γ coincidences. To determine the percentage of n-n coincidences, we use the setup shown in Fig. 15. A spectrum for all coincidences was first recorded in the multichannel analyzer (MCA). Then the pulse shape discrimination (PSD) circuit was set for gamma rays only and a second spectrum was recorded. In the first ungated spectrum, n-n, n-γ, and γ-γ coincidences are accepted. In the gated spectrum, n-γ and γ-γ coincidences are recorded. The difference between the two spectra is due to n-n coincidences.

The results are shown in Fig. 16. In the 5- to 45-ns gate, comparable to the RD gate, n-n coincidences account for 72 ± 4% of the data and n-γ coincidences account for the rest. In the 0- to 5-ns gate, n-n coincidences account for 54% of the data and n-γ and γ-γ coincidences account for the rest. The coincidence distributions in Fig. 16 are made more understandable in the schematic of Fig. 17, where the various contributions are shown approximately to scale. For the n-n coincidences, the peak at zero

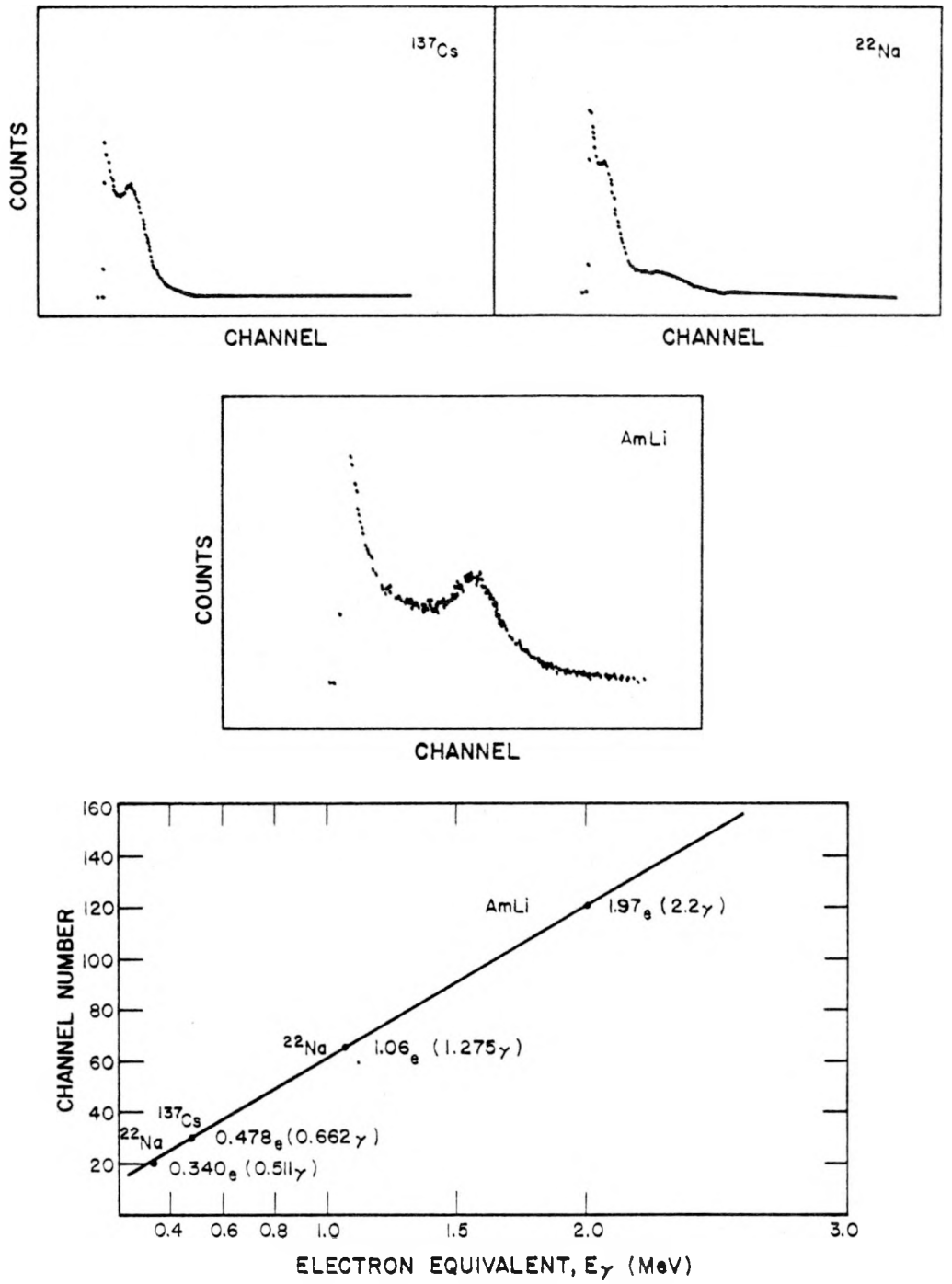


Fig. 14.

Gamma-ray and AmLi spectra and resulting calibration curve for an RD scintillator. Data points correspond to the Compton-scattered electron. The corresponding gamma-ray energies are in parentheses.

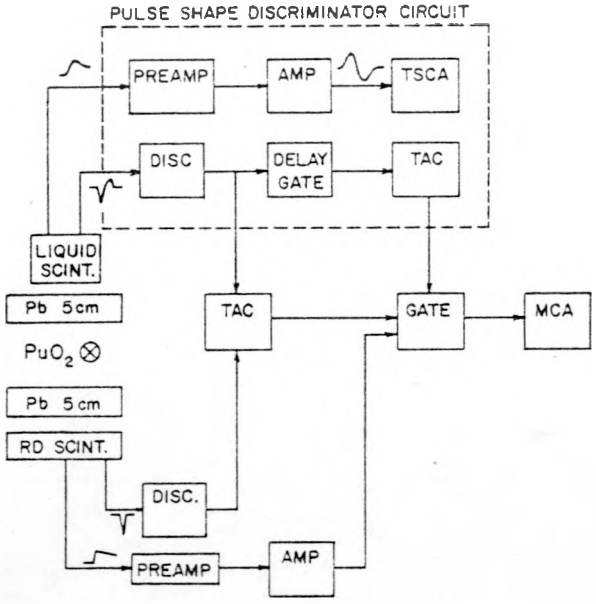


Fig. 15.
Experimental setup for neutron-gamma sensitivity measurement.

time difference indicates that in the most probable case both neutrons are emitted with the same energy, although one cannot determine what that energy is. For the n-γ coincidences the situation is different. Because one gamma is emitted and has a constant flight time, the timing distribution reflects the distribution in the time of flight for the neutron and gives the neutron energy distribution. Because the neutron energy E_n is related to the time of flight via

$$E_n = \frac{1}{2} m \frac{L^2}{t} ,$$

where L is the flight path and t is the time of flight, events closer to t = 0 correspond to higher energy neutrons, and as t increases E_n decreases. The consequence of the gating currently used on the RD can then be stated as follows. By removing the γ-γ peak, one also removes those γ-n coincidences for which the neutron energy is high. By accepting only events between 5 and 45 ns, one preferentially accepts lower energy neutrons from the n-γ coincidences.

One can also legitimately ask whether it is even necessary to remove the γ-γ coincidences. The 5 cm of lead shielding will attenuate low-energy gammas (<1-MeV) more than high-energy gammas. This implies that the gamma rays penetrating the lead are mostly of high energy and therefore comparable to neutrons in penetrability. However, attenuation of a specific energy gamma often does not remove the gamma completely but merely downscatters it to a lower energy. With the 200-keV gamma threshold, a fraction of gammas in the 500- to 1000-keV range might be detected as downscattered gammas. Therefore it was important to determine what range of gamma energies incident on the lead shield produces a detectable signal in the scintillators.

A small test plastic scintillator was set up and the linear signal was processed so that pulse-height analysis could be performed. The threshold was made the same as that for the RD scintillators. Three gamma sources were used: ^{133}Ba ($\gamma_{av} \approx 350$ keV), ^{137}Cs ($\gamma = 662$ keV), ^{60}Co ($\gamma_{av} \approx 1.25$ MeV). For each source the integrated response was measured with and without a 5-cm lead absorber between the source and detector. The ratio of these two sets of data is the transmission for the gamma of that particular energy. The measurements indicate that ~38% of the gamma rays detected in the scintillators correspond to initial-fission gammas of <1 MeV, with the peak in the distribution at ~600 keV. The results of this measurement are shown in Fig. 18.

Contributions from the various coincidences for each gate are tabulated in Table II. The existing RD gate is 5 to 45 ns and ~10% of the coincidences have an associated low-energy (<1-MeV) gamma. If the RD gate is increased to include the 0- to 5-ns interval, then ~11% of the coincidences have an associated low-energy gamma ray. This larger coincidence gate (0 to 45 ns) would increase the coincidence rate but would not seriously change the possible matrix dependence of the assay. With this larger gate, the threshold of the scintillators could be raised to minimize the effects of the low-energy gammas.

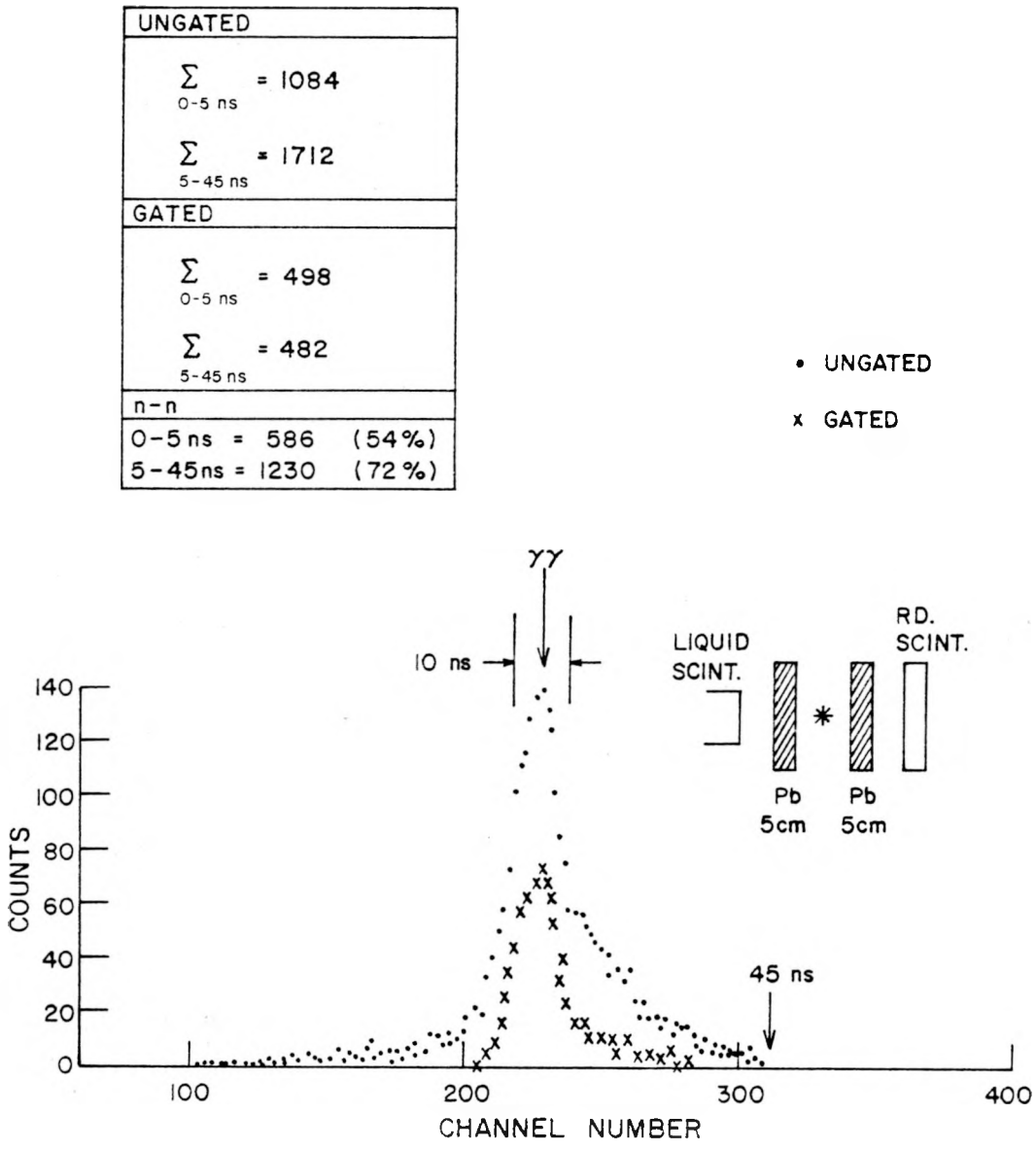


Fig. 16.
RD timing distribution distribution for a 41-g plutonium sample. • includes n-n, n-γ, and γ-γ coincidences. x includes n-γ and γ-γ coincidences.

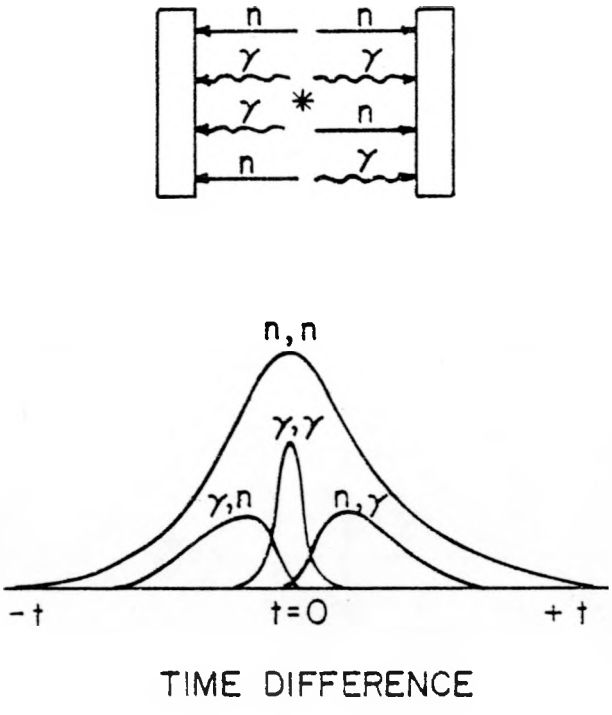


Fig. 17.
Idealized timing distribution for random driver. Coincidence contributions are approximately to scale.

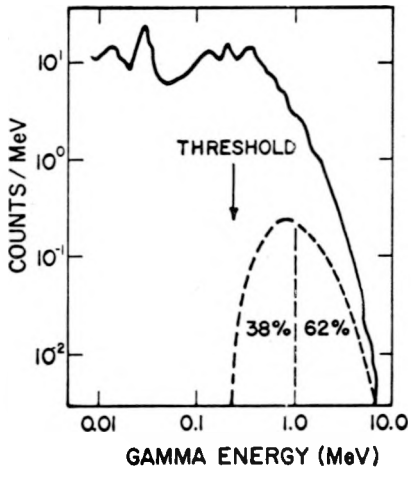


Fig. 18.
Spectrum of gamma rays produced by the fission process. The solid line is obtained from data presented in Ref. 5, p. 66. The dashed line is the spectrum detected by RD.

TABLE II

CONTRIBUTIONS OF VARIOUS COINCIDENCES FOR DIFFERENT COINCIDENCE GATES

Coincidence Gate (ns)	Coincidence Type (%)		
	n-n	n-γ	γ-γ
0 to 5	54	21	25
5 to 45	72	28	0
0 to 45	65	25	10

III. GAMMA-RAY ASSAY TECHNIQUE DEVELOPMENT

A. Application of L-Edge Densitometry to Solutions Containing Multiple SNM Components (T. R. Canada, J. W. Tape, S. T. Hsue, and D. G. Langner)

K absorption-edge densitometry has been used successfully for the simultaneous determination of the thorium and uranium contents of high-temperature gas-cooled reactor fuel having a thorium-to-uranium density ratio of 4 (Ref. 4, pp. 10-13). Preliminary studies have also shown that L_{III} absorption-edge densitometry is applicable to plutonium and uranium assay in solutions containing a similar uranium/plutonium concentration (Ref. 13, p. 10). The purpose of the present study is to develop an in-line L_{III} absorption-edge densitometer for rapid simultaneous measurement of multiple concentrations of special nuclear materials (SNM). Such an instrument has been proposed for installation in the Savannah River Laboratory's (SRL) experimental coprocessing setup, where solutions can be drawn from processing lines that provide concentrations in the range of 22 to 37 g U/ℓ, 2 to 5 g Pu/ℓ, and uranium-to-plutonium concentration ratios of 4 to 8.

The assay of multiple SNM can be performed conveniently when the two components are present in roughly equal amounts (ratios between 1 and 4). If the concentration ratios are >4, most of the pulse-processing time will be spent on the major component (uranium in the SRL case); consequently, the minor component (plutonium in the SRL case) will be poorly determined. When the solution contains uranium and plutonium, problems arise because their energies at the L_{III} edge are so close (17.168 and 18.066 keV, respectively). However, the relevant transmissions can be measured in the limited energy range between the two L_{III} edges.

The statistics around the absorption edge of the minor SNM component can be improved in several ways. The x-ray beam intensity near the uranium absorption edge can be attenuated relative to the beam near the plutonium edge. One can also use higher counting rates, but that worsens the detector resolution and narrows the range between the absorption edges in which transmission measurements can be made.

Another approach to improved statistics is to use as many data as possible. A typical x-ray spectrum transmitted through a uranium- and zirconium-bearing solution is shown in Fig. 19. The zirconium can be used to simulate the plutonium because the zirconium K edge at 17.998 keV is representative of the plutonium L_{III} edge at 18.066 keV. Selected regions of data have been extrapolated (straight-line) to the absorption edges to determine the transmission ratios, as shown in Fig. 19.

The transmission through a solution of thickness x containing a single SNM is given by

$$\ln T = -\mu_s \rho_s x - \mu_m \rho_m x \qquad (2)$$

where μ and ρ are the mass-absorption coefficient and density, and subscripts s and m refer to the SNM and matrix material, respectively. The difference in ln(T) at a definite energy step is given by

$$\ln \frac{T_i}{T_{i-1}} = -(\mu_{s,i} - \mu_{s,i-1})\rho_s x - (\mu_{m,i} - \mu_{m,i-1})\rho_m x \qquad (3)$$

A plot of the differences is shown in Fig. 20. For a narrow energy range, the following assumption can be made:

$$\mu_{m,i} - \mu_{m,i-1} = \text{constant} = C_m$$

$$\mu_{s,i} - \mu_{s,i-1} = \text{constant} = \begin{cases} C_L & \text{below the absorption edge} \\ C_U & \text{above the absorption edge} \\ \Delta\mu_s & \text{across the absorption edge.} \end{cases}$$

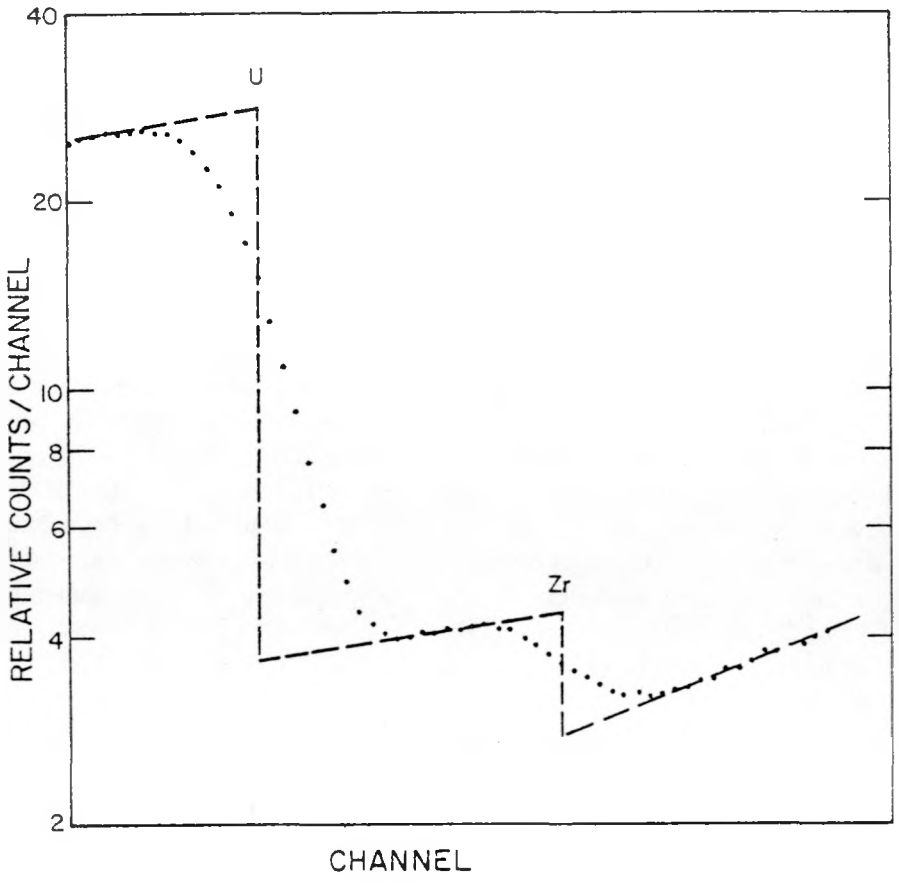


Fig. 19.
Continuous x-ray spectrum transmitted through a solution containing uranium and zirconium. Data are straight-line extrapolated to the absorption edges to determine the transmission ratios at the edges.

Summing over N channels of the "peak" shown in Fig. 20,

$$\sum_{i=1}^{N} \ln \frac{T_i}{T_{i-1}} = -C_L \rho_s x \frac{N}{2} - C_U \rho_s x \frac{N}{2} - C_m \rho_m x N - \Delta\mu_s \rho_s x \qquad (4)$$

The first three terms on the right-hand side of Eq. (4) represent the "background" under the peak and can be subtracted. So the net area of the peak is proportional to ρ_s, the density of the SNM. The advantage of this approach as a measure of the SNM content is that the increased number of data points permits a more precise determination.

We used this approach in measuring a solution containing 6.2 g Zr/ℓ and 37 g U/ℓ. From repeated runs of 1000-s counting time, we found that uranium content can be measured to 0.5% and zirconium content to 2%.

B. Uranium-Plutonium Solution Assay by Transmission-Corrected X-Ray Fluorescence (P. Russo, M. P. Baker, and T. R. Canada)

Techniques of absorption-edge densitometry applied to the NDA of SNM-bearing solutions have been successful over a wide range of solution concentrations (Ref. 14, Vol. II, App. A). The magnitude of the measured transmission discontinuity across the

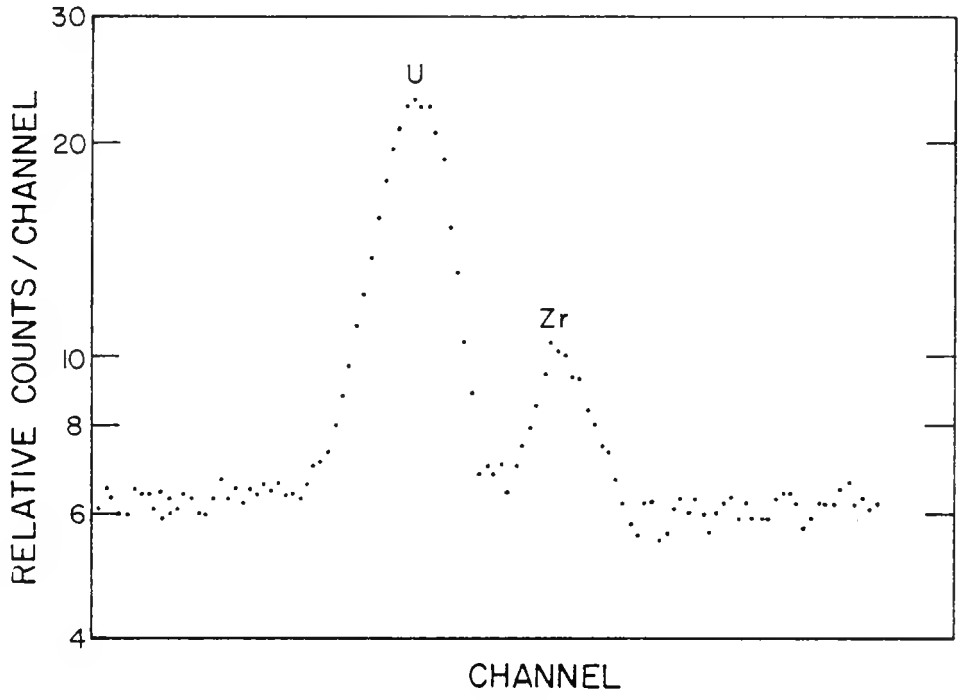


Fig. 20.
Difference in the log of transmission for a fixed-energy increment as a function of energy. Transmissions are measured by passing a continuous x-ray spectrum through a solution containing uranium and zirconium.

absorption edge determines the density of a single element rather than its isotopic content, thus distinguishing these measurements from assays obtained by direct counting of radiations emitted by the sample. In the higher concentration ranges (50 to 400 g/ℓ), discrete gamma rays that bracket the absorption edge are sufficient to perform measurements; at lower concentrations, an x-ray machine that generates a continuum across the energy region of the absorption edge is required to obtain comparable accuracy. Furthermore, for reasonable counting times the uncertainties increase to more than a few per cent for sample concentrations <2 g/ℓ.

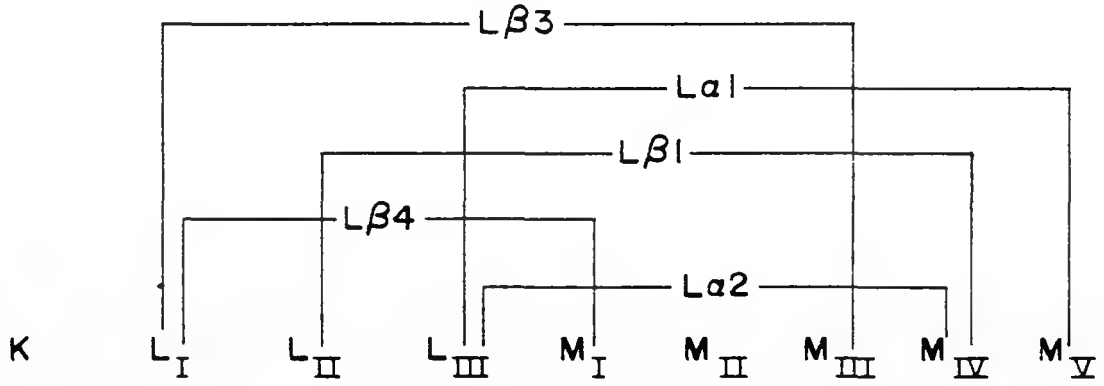
To achieve higher precision in the NDA of lower density solutions, we began the present study, in which we use techniques of energy-dispersive x-ray fluorescence (XRF). Like absorption-edge densitometry, XRF also measures the total elemental concentration. However, for lower concentrations the x-ray machine can be eliminated, thereby reducing the cost and simplifying the NDA equipment.

Table III gives the electron binding energies of the K, L. and M edges for lead, thorium, uranium, and plutonium and also indicates the transitions corresponding to the most prominent L x rays. In some cases, proper selection of the energy of the fluorescing source can preclude the creation of some of the L vacancies and simplify the x-ray spectra. An example is the choice of ^{109}Cd as a source for the assay of plutonium. The ^{109}Cd source has a relatively long (435-d) half-life and emits only one gamma ray at 88 keV. This 88-keV gamma ray is accompanied by silver K x rays (daughter of ^{109}Cd) sufficiently energetic ($E_{K\alpha 1}$ = 22.16 keV, $E_{K\alpha 2}$ = 21.99 keV) to create only L_{III} vacancies in plutonium, yet excite vacancies in the three L orbitals of thorium and uranium.

Figures 21a, b, and c show the expected distribution of L x-ray intensities for plutonium, thorium, and uranium excited by ^{109}Cd. These are empirical results[15] for L_I, L_{II}, and L_{III} x rays normalized to the most intense member of each group. Equal probability is assumed, in the cases of thorium and uranium, for the excitation of L_I, L_{II}, and L_{III}

TABLE III

ELECTRON BINDING ENERGIES (keV)



Pb	88.005	15.861	15.201	13.035	3.851	3.554	3.067	2.586	2.484
Th	109.651	20.472	19.693	16.300	5.182	4.831	4.046	3.491	3.332
U	115.606	21.758	20.948	17.168	5.548	5.181	4.304	3.728	3.552
Pu	121.818	23.102	22.266	18.057	5.933	5.546	4.562	3.973	3.778

vacancies. The triangular line shapes are drawn with a full width of 350 keV at half maximum. (Such resolution is achieved routinely with Si(Li) detectors at these energies.) The relatively simple plutonium x-ray spectrum induced by ^{109}Cd activation makes the plutonium assay possible even in the presence of thorium or uranium. This feature has been exploited to innovate transmission-correction techniques in the present XRF measurements. The new approach to transmission-corrected x-ray fluorescence (TC XRF) is discussed below.

In the absence of sample attenuation of the incident and outgoing x rays, the sample concentration ρ_s is proportional to N_s, the number of x-ray events detected. For a sample solution of finite thickness x and total density ρ_t, the exact expression is

$$\rho_s = \frac{N_s}{K} \frac{\ln\alpha}{1-\alpha}$$

where

$$\alpha = e^{-\mu_i \rho_t x} e^{-\mu_o \rho_t x}$$

K = constant.

The quantities μ_i and μ_o are the sample attenuation coefficients for the incoming and outgoing x rays, respectively. Therefore, the first exponential factor in the quantity α is the transmission of the source x rays through a sample of thickness x, and the second factor is the transmission of fluoresced x rays through the same sample thickness. The challenge to the assay technique is the evaluation of the transmission-correction factor, $\ln\alpha/(1-\alpha)$. One approach to the evaluation of α is shown schematically in Fig. 22. By selection of the elemental composition of the transmission foil such that its characteristic fluoresced x rays bracket the sample x-ray assay energy E, α may be determined at E. The ratio of the foil x-ray intensity, with and without the sample in place, determines α (or the transmission) as a function of x-ray energy. Assuming $\ln\alpha$ is a linear function of energy over a limited energy range, $\alpha(E)$ can be determined by linear interpolation. This TC XCF technique often will allow a simple, accurate assay to be made on SNM-bearing solutions in a single measurement.

A study of such factors as dependence on sample density, count rates, energy resolution, and precision of assay measurements was carried out using

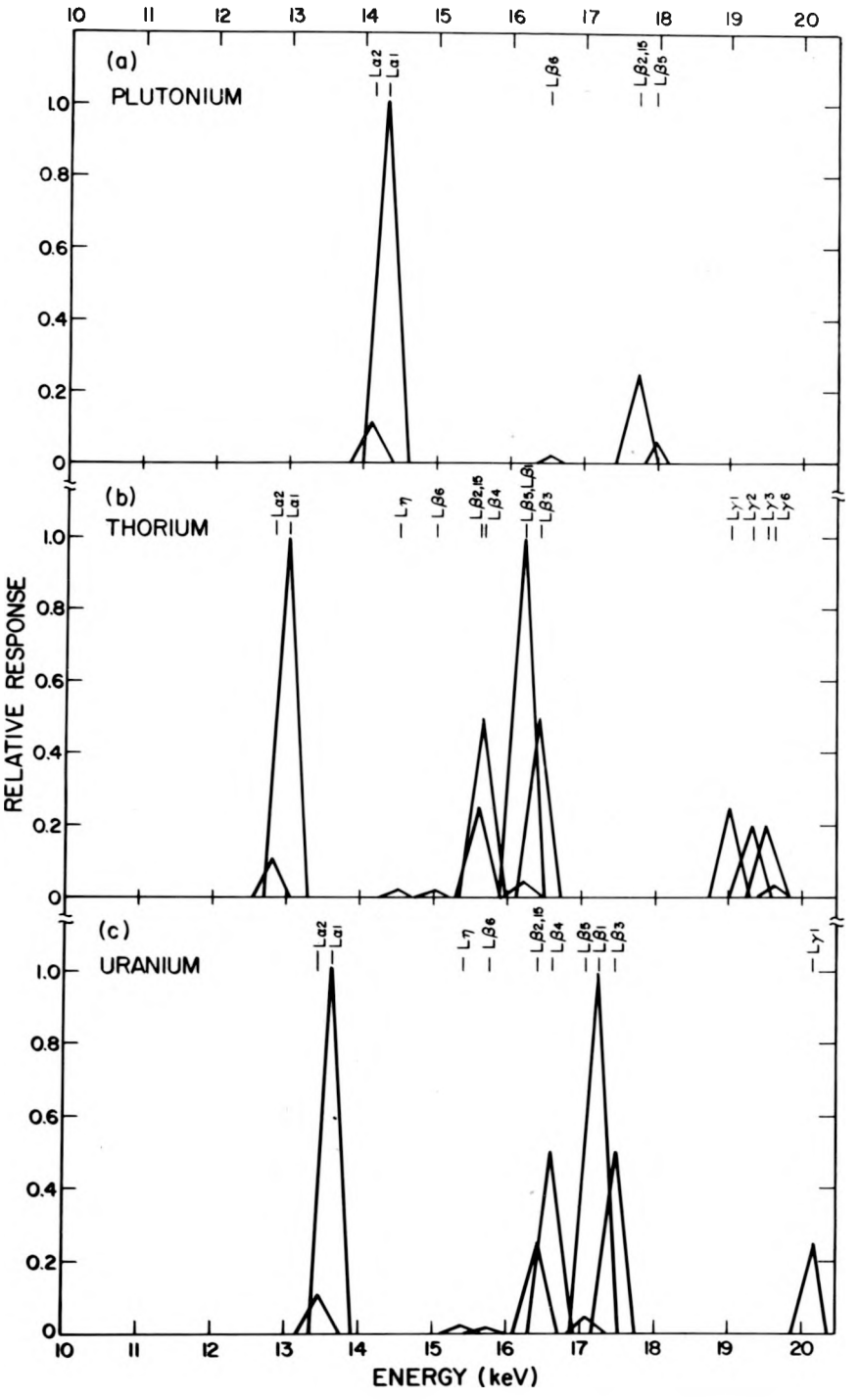


Fig. 21.

L x-ray spectral lines for ^{109}Cd excitation of (a) plutonium, (b) thorium, and (c) uranium displayed with triangular line shapes corresponding to a resolution of 350-eV full width at one-half maximum. Equal probability is assumed for creation of L_I, L_{II}, and L_{III} vacancies in thorium and uranium. The relative intensities of L_I, L_{II}, or L_{III} x rays were obtained from a compilation of empirical results.

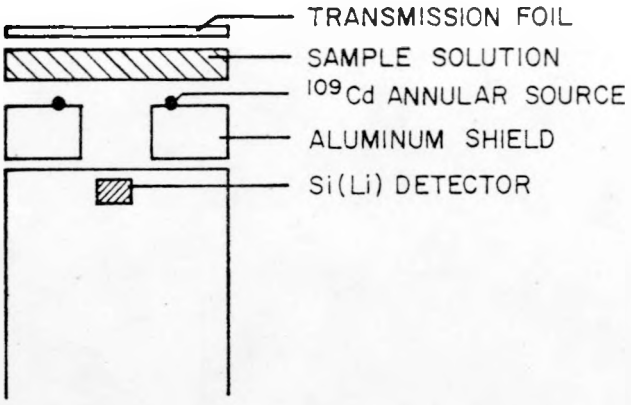


Fig. 22.
Arrangement of Si(Li) detector, ^{109}Cd source, uranium solution sample, and transmission foil for TC XRF assay.

solutions of dissolved $UO_2(NO_3)_2 \cdot 6H_2O$, a ^{109}Cd source, and the experimental arrangement shown in Fig. 22.

The detector was shielded from the annular ^{109}Cd gamma source by 1 cm of aluminum. The sample solutions were contained in plastic disk-shaped holders that provided for a uniform 4-mm sample thickness. The ~100-μCi activity of the annular ^{109}Cd source and the small (~12-mm^2) Si(Li) x-ray detector gave rise to total count rates of 10 to 30 s^{-1}. The counting times were increased accordingly to compensate for the temporary lack of a stronger source and larger detector. The energy resolution of the x-ray lines was ~300 eV. The measurements were made on solutions containing uranium in the concentration range of 1 to 20 g/ℓ. A sample spectrum is shown in Fig. 23 for a 1-g/ℓ uranium solution and a thorium transmission foil. This can be compared with Figs. 19b and c, which show the relative, unattenuated L x-ray intensities for thorium and uranium, respectively.

The assay of a single element by TC XRF using L x rays should involve a unique combination of sample, foil, and source to minimize the overlap of x-ray lines. The L x-ray spectrum of thorium is not well suited to the TC XRF assay of uranium in a single measurement because the groups of Lβ lines for the two elements overlap near 16.5 keV (near channel 570 in Fig. 23). The same is true to a lesser extent for the Lα lines at lower energies. However, a two-pass assay (with and without the thorium foil) permits one to subtract the uranium contributions from the thorium peaks, thus providing a demonstration and proof of principle. The assay peak for uranium corresponds to the Lα line at 13.6 keV. The higher uranium peak at 17.3 keV, while sufficiently intense and well-resolved from that of thorium, cannot be used for the transmission-corrected assay. This peak lies ~100 eV above the L_{III} edge of uranium, invalidating a transmission correction by extrapolation.

The solid points in Fig. 24 are plots of concentration vs data corresponding to the count rate per unit concentration for the uranium-assay peak. Significant self-attenuation is evident at concentrations >2 g/ℓ.

Figure 25 shows the transmission T, measured at the three thorium peak energies, plotted against energy, for four of the five solutions. (Transmission data for the 1-g/ℓ uranium solution were not obtained.) The negative deviation of the high-energy point from the line drawn through the two low-energy points is due to the energy dependence of the gamma-ray attenuation and perhaps also to the possible presence of contaminant thorium in the uranium solutions. Because the thorium L_{III} edge lies at 16.3 keV, the thorium Lβ3 line at 16.4 keV (which contributes approximately one-third of the events falling into the high-energy thorium peak) is attenuated in proportion to the concentration of the thorium contaminant. Therefore, a linear interpolation of the measured transmission values to 13.63 keV was performed for each solution using only the two lower energy thorium data points. The correction factors, derived as described previously, were applied to the data plotted in Fig. 24. The corrected values of the count rate per unit concentration, normalized to the uncorrected value for the 1-g/ℓ solution, are plotted as open circles in Fig. 24. The weighted average of these values, indicated by the horizontal line, has a 0.5% statistical uncertainty.

The success of the thorium-transmission corrections with a measurement precision of 1% or better for the low-concentration uranium solutions is a guarantee that the TC XRF plutonium assay with a thorium foil will also succeed since the extraction of peak areas is more straightforward for the simpler spectra obtained (see Fig. 21). Furthermore, the use of a stronger source and larger detector will reduce counting times by two to three orders of magnitude,

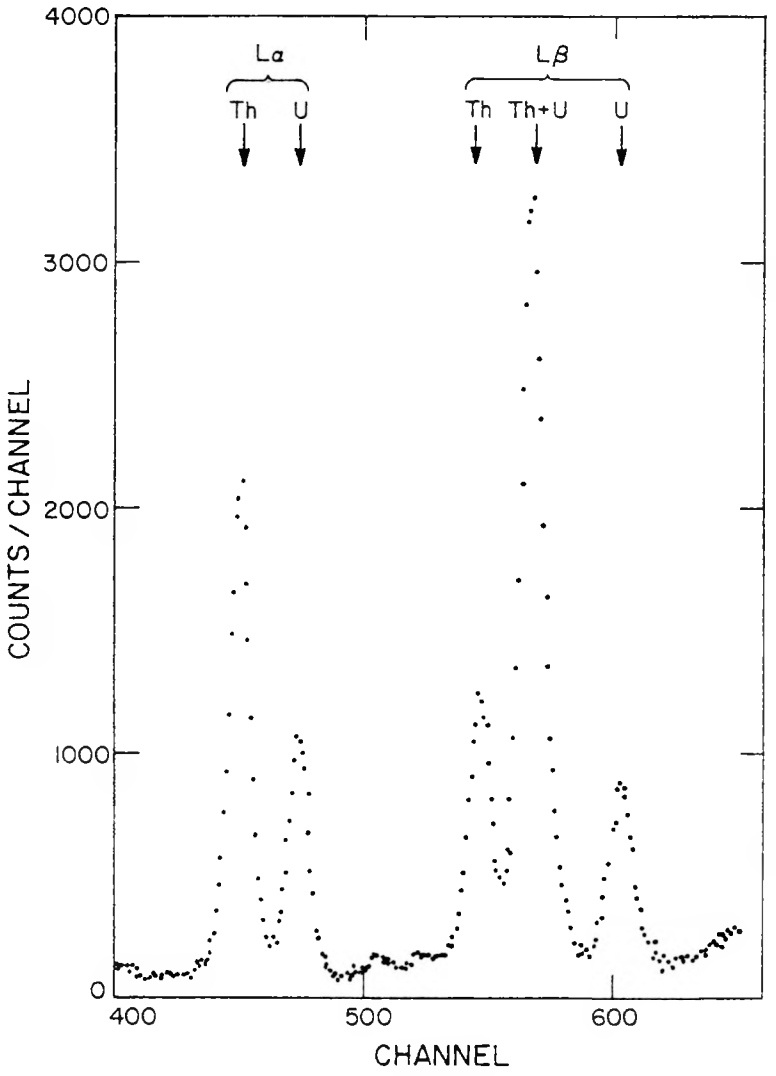


Fig. 23.
Lα x-ray spectrum obtained from 1-g/ℓ uranium solution and a thorium transmission foil using a ^{109}Cd source.

perhaps lowering backgrounds and improving the precision.

While the TC XRF assay of uranium solutions is difficult to perform with thorium, lead is a suitable alternative in terms of the L x-ray spectrum. The Lα, Lβ, and Lγ lines for lead are conveniently grouped at approximately 11.5, 12.5, and 15 keV, respectively, permitting resolution of three peaks for each of the two elements in a single spectrum. Because of the lower energy of the lead x rays, the uranium assay with lead foil requires thinner samples than those designed to optimize the relative thorium-plutonium peak intensities.

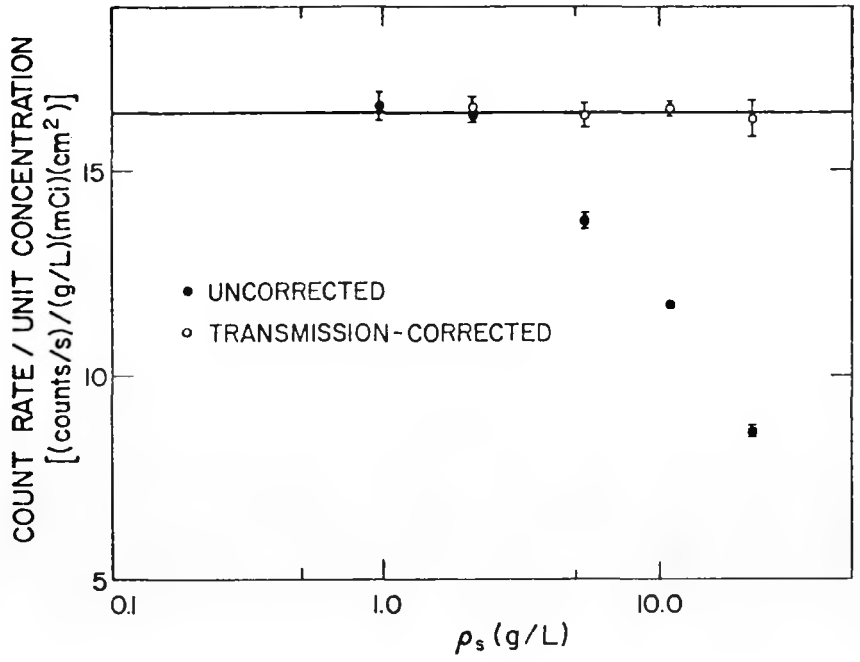


Fig. 24.
Count rate for the uranium 13.6-keV assay peak per unit concentration (per 1 mCi of source activity and 1-cm² detector area) plotted vs uranium concentration. The solid points show the uncorrected data. The open points are corrected for self-attenuation, as described in the text, and normalized to the uncorrected point for the 1-g/ℓ solution.

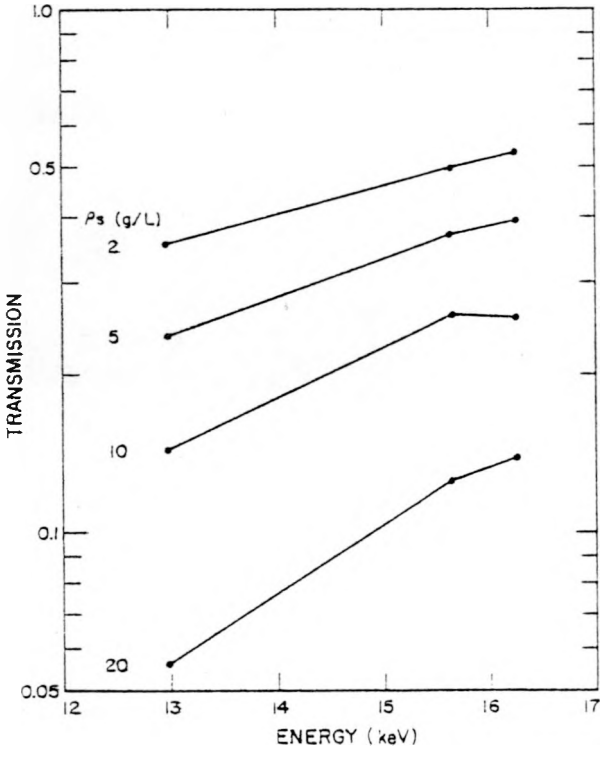


Fig. 25.
Fractional transmissions of the three thorium lines plotted against energy for each of four uranium solution concentrations.

IV. FIELD TESTS AND EVALUATIONS

A. Design of the ^{252}Cf Shuffler for SRP (T. W. Crane, G. W. Eccleston, and L. G. Speir)

A Shuffler unit employing a fast but still primarily subthreshold neutron interrogation with delayed-neutron counting is being designed for assaying the ^{235}U content of various materials at the SRP fuel fabrication facility (Ref. 3, p. 34). The materials to be assayed include uranium-plutonium ingots as long as 25 cm, assorted pieces of rejected fuel elements and scraps, and waste which includes lathe chips, saw chips, floor sweepings, and flux (LiF, AlF_3) ingots produced during leaching operations.[16] Most of the materials will be packaged in cans 17.8 cm in diameter and 30.5 cm in height. The Shuffler will be used to assay ~35 items per day. Thus, with sample handling, data logging, and calibration checks the actual sample assay time will have to be 500 s or less. To facilitate the operation of the Shuffler unit and data reduction, a computer will be monitoring and controlling the assays (see Part 1, Sec. V-B).

A schematic of the Shuffler designed for SRP is shown in Fig. 26. The assay begins with a weight measurement of the sample, which is checked against the sample weight entered by the operator. The assay proceeds with a background count of the sample with the ^{252}Cf source still in the storage position. The active portion of assay consists of a cyclic neutron activation of the sample with the source, followed by delayed-neutron counting with the source in the storage tank. Upon completion of the assay, a hardcopy output gives the NDA measurement of the ^{235}U content and its measurement uncertainty as well as the total mass of the sample.

The mechanical hardware for the Shuffler shown in Fig. 26 includes a sample interrogation/counting well and a source storage tank that isolates the ^{252}Cf source during the delayed-neutron counting period. In the interrogation well the ^{252}Cf source is moderated, so the contribution from ^{234}U, ^{236}U, and ^{238}U is <2% of the ^{235}U signal. The inside surface of the interrogation well is lined with boral and cadmium to maintain a penetrating neutron-energy spectrum. The interrogation-counting well contains load cells for weighing the sample and a turntable for rotating the sample. Surrounding the interrogation-counting well on the sides and bottom are the ^{3}He-filled neutron detectors used for counting delayed neutrons.

The design of the SRP Shuffler was based upon a series of Monte Carlo neutron-transport calculations, and serious consideration was given to ease of construction and system reliability. The calculation of the system's response consists of two independent simulations using the LASL-developed MCN code.[17] First, a calculation of the neutron interrogation is made; the principal result is the number of fissions induced in the sample as a function of isotope and position. From these data the uniformity and penetrability of the neutron interrogation are obtained. The second step is a simulation of the emission of delayed neutrons from the sample, yielding the neutron-detection effficiency of the ^{3}He tubes.

An elevator for positioning the sample is not necessary to obtain a uniform interrogation over the sample volume.[3] During this reporting period more detailed calculations were made to obtain a response that was linear with the sample's ^{235}U mass. For cast ingots the mass is proportional to the height, with the result that the low-mass samples are somewhat pancake-shaped and the high-mass samples are tall cylinders. This change in the sample geometry with mass affects the response of the system. For the low-mass samples, the response per gram rises sharply once the length is less than the radius because the large surface-to-volume ratio results in less neutron self-shielding in the sample. On the other hand, the response per gram for large samples tends to increase because of multiplication.

To compensate for both of these effects, the neutron-tailoring assembly was designed so that the interrogating flux would be slightly lower at the top and bottom of the assay chamber. The flux depression was selected to cancel the increased response of short samples at the bottom of the assay chamber and to offset the multiplication present in the heavy samples. Figure 27 shows the calculated fission rate as a function of position in the sample for a 27.94-cm-high, cyclindrical uranium-aluminum ingot. The density of the ingot was assumed to be 3 g/cm^3 and uniform, with a relative ^{235}U density of 15%. The decreased fission rate near the bottom and top

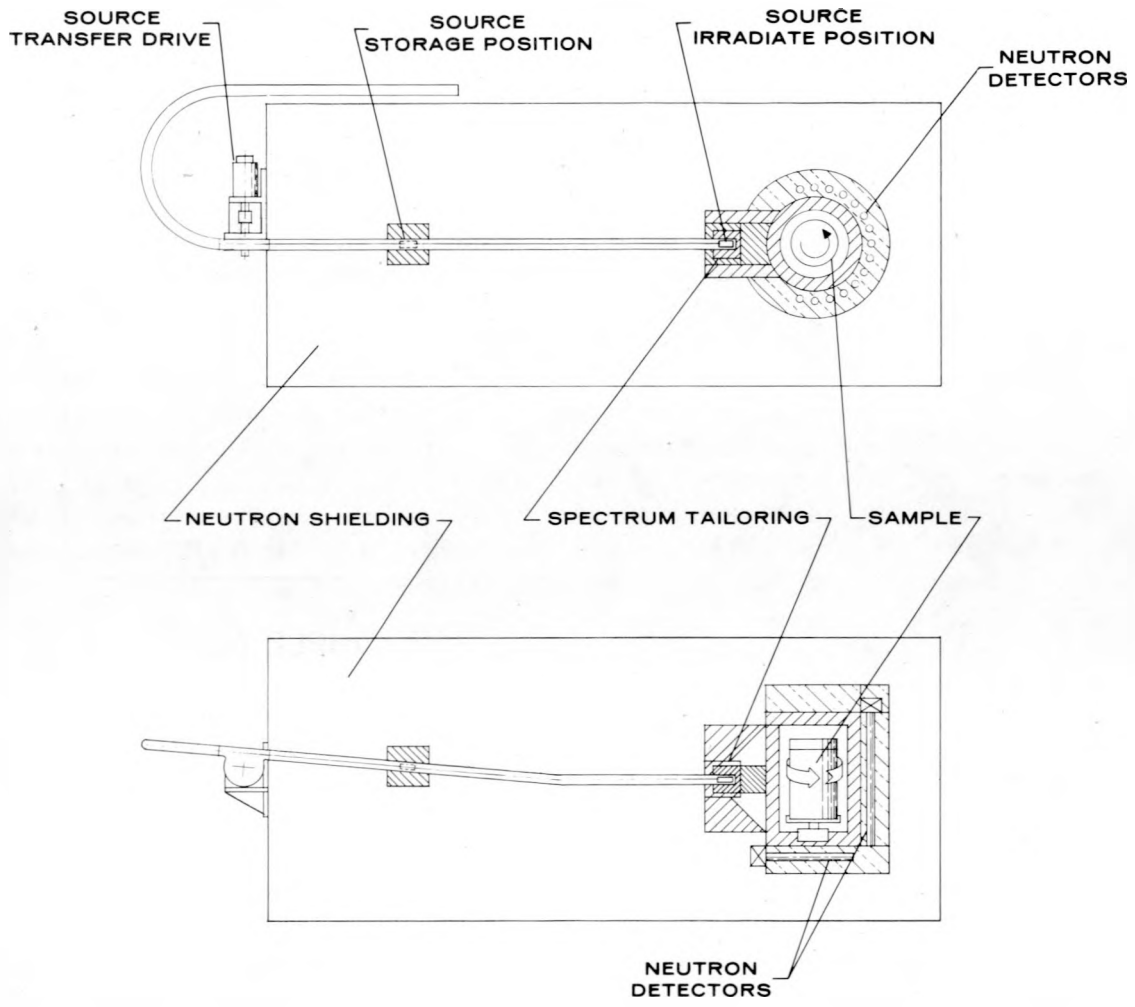


Fig. 26.
Schematic of the Shuffler assay system for the SRP. The sample is shown in the interrogation-counting well.

of the cylinder can be observed. Figure 28 shows the relative response of the system, including the calculation of interrogation and delayed-neutron counting efficiency. The results indicate a linear response by the Shuffler over a sample-height range of 2.54 to 27.94 cm to within the accuracy of the calculation (±3%). Heights for the SRP materials are expected to be 5 to 25 cm, with the tallest samples having a ^{235}U mass of ~2.5 kg.

Potential small deviations can be included in the calibration function or in a correction factor. If the correction-factor approach is used, a measure of the sample height can be obtained from the ratio of delayed neutrons detected in the side counters to the total number of delayed neutrons counted (side plus bottom detectors). A plot of this ratio (based on the Monte Carlo calculations) is shown in Fig. 29. The ratio is linear except for the tallest cylinders.

B. Preliminary Design of a ^{252}Cf Shuffler for the Idaho Chemical Processing Plant (G. Eccleston and H. O. Menlove)

Preliminary design specifications are complete for the addition of a ^{252}Cf Shuffler system to the Idaho Chemical Processing Plant (ICPP). Title I (preliminary) design for the addition is complete,

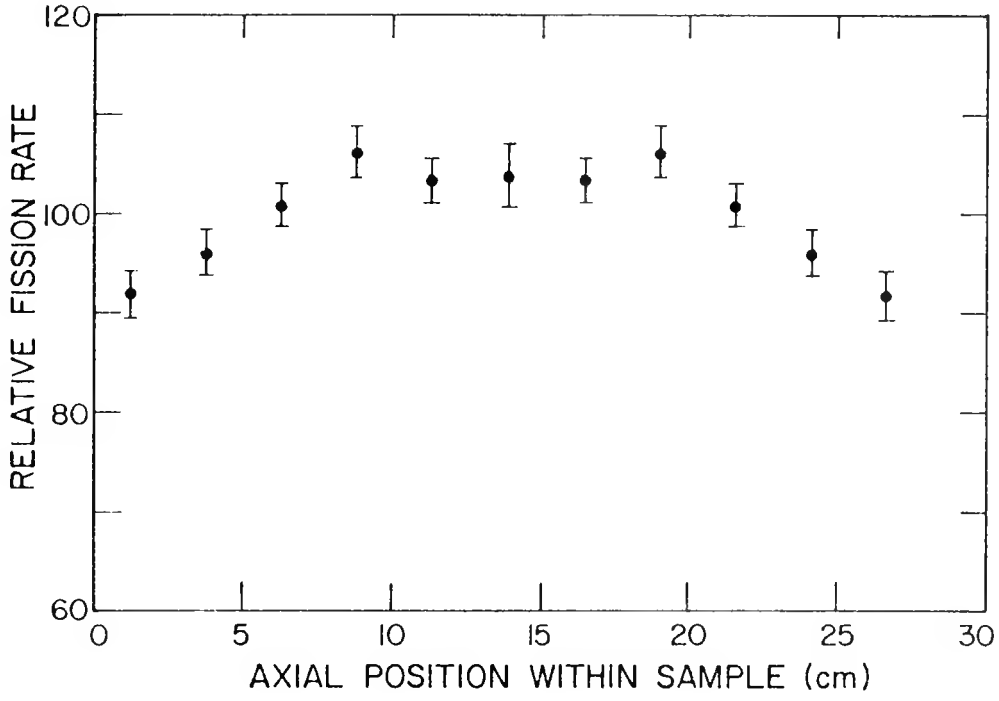


Fig. 27.
Induced fission rate in a 27.94-cm-tall cylindrical sample. The position is measured vertically from the bottom of the sample.

and Title II (construction) drawings are being made at the Ralph M. Parsons Co., Pasadena, California. The purpose of ICPP is to reprocess spent fuel containing high-enrichment uranium received from noncommercial sources. The Shuffler will determine ^{235}U content by making a neutron interrogation both of spent fuel packages before dissolution and of waste canisters after reprocessing operations.

The neutron interrogator will consist of a ^{252}Cf source, a source Shuffler mechanism, neutron detectors, and fission chambers. The instrument will be housed in a cubicle located below the fluorinel dissolution cell. Waste canisters and spent fuel packages will be lowered by crane into the interrogator through tubes that penetrate the cell floor and extend below the Shuffler. The cubicle is isolated from the dissolution cell and requires remote operations. Additional equipment needed to operate the interrogator will include a data processor and nuclear electronics (with associated equipment) located in the building control room. In addition, a remote terminal will be placed in the crane operation area near the dissolution cell to enable the crane operator to perform interrogation operations.

General design criteria for the neutron interrogator system have been specified and reviewed by Parsons Co. and Allied Chemical Corp. Specifications for assay of waste canisters call for the capability to determine ^{235}U content to 100 ± 30 g (95% confidence level). The ^{235}U content of fuel packages is to be determined within ±5% at a 95% confidence level, with a total transfer and analysis time not to exceed 30 min per assay. Specifications of Shuffler component function, space requirements, power, and cabling interconnection have also been submitted to Parsons Co. and are being incorporated into the Title II design.

C. Moisture Effects on the Assay of Waste Canisters for ICPP (D. A. Close, H. O. Menlove, and G. Eccleston)

One of the questions concerning the assay of waste from the fluorinel process at the ICPP has been about the effect of moisture in the waste canister.

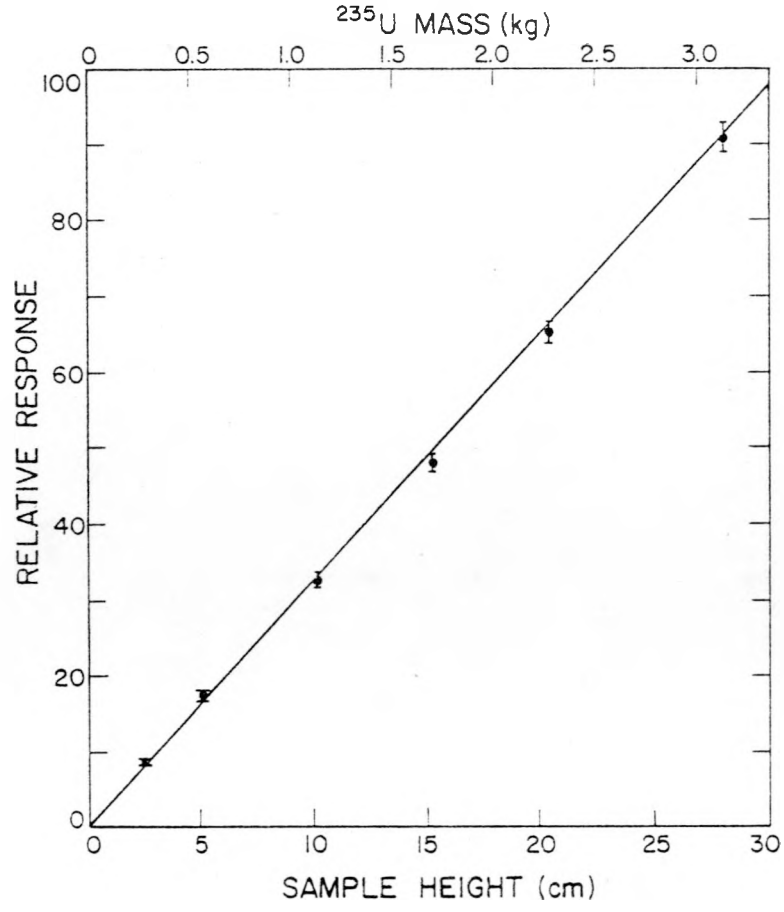


Fig. 28.
Delayed-neutron response as a function of sample height and ^{235}U mass.

Monte Carlo calculations were performed to study this effect on measurements made with the ^{252}Cf Shuffler. We used concentric cylindrical shells for the geometry mockup. The waste was contained in a cylindrical canister, 12.7 cm in diameter and 60.9 cm high, surrounded by a thin layer (5×10^{-6} cm) of ^{235}U to simulate a fission monitor containing 100 mg of ^{235}U. Then followed (1) an 0.8-mm-thick layer of cadmium, (2) a 15.2-cm-thick shell of lead, (3) an 0.8-mm-thick layer of cadmium, (4) two layers of polyethylene, between which was an 0.84-cm-thick shell of ^{3}He at 4 atm pressure. The inner layer of polyethylene was 2.54 cm thick; the outer layer was 6.78 cm thick. This arrangement gave the same amount of ^{3}He as found in the actual case where 27 ^{3}He tubes 2.54 cm in diameter and 30.48 cm long are filled to a pressure of 4 atm. For the neutron interrogation, a point ^{252}Cf source was located 15.32 cm from the center of the waste canister.

The waste from the ICPP results from dissolving spent fuel elements in a fluorinel process. The highly radioactive material contains ZrO_2, iron, and uranium, as well as some fluorine. Also present are some boron and cadmium which act as poisons for criticality control. For the calculations, we assumed the mass of ZrO_2 plus the mass of iron to be 45 kg, 97% of which was ZrO_2 and 3% of which was iron. We assumed that the waste canister contained 100 g of ^{235}U and 5.26 g of ^{238}U.

Two series of calculations were made. The amount of water in the canister was the only variant in the first series. There the waste always contained 176 g fluorine, 13.9 boron, and 34.8 g cadmium, which corresponds to 4*M* fluorine, 6 g/ℓ boron, and 15 g/ℓ cadmium in a canister having 30% water by volume. The first calculation incorporated 30% water by volume; subsequent calculations assumed less water, thus simulating the drying of the waste.

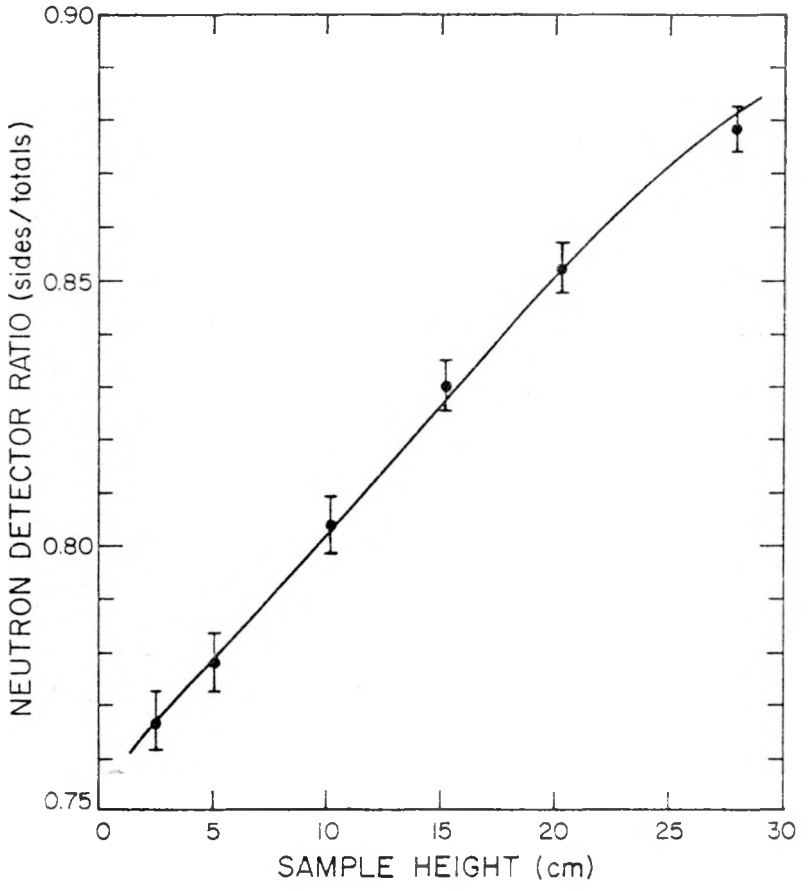


Fig. 29.
Delayed-neutron ratio defined by (counts in side detectors)/(counts in side plus bottom detectors) as a function of sample height.

The events tallied by the Monte Carlo code were the number of fissions occurring in the waste and in the ^{235}U fission monitor. The number occurring in the monitor was applied as a correction factor to the number occurring in the waste. The relative number of fissions in the waste as a function of the amount of water in the waste canister is shown in Fig. 30. Also shown are the fission counts in the waste corrected by the fission monitor events.

The number of fissions in the waste differs by more than a factor of 10 as the amount of water in the waste canister varies from 0 to 30%. When these counts are corrected by the fission monitor counts, the factor is lowered to 1.8. Thus, the fission monitor corrects for much of the effect of the water, but there is still a noticeable effect on the corrected response of the assay instrument when >5% water by volume is in the waste canister. As long as the waste canister contains <5% of water by volume, the fission monitor adequately corrects for the effect of water.

In the second series of calculations, the boron and cadmium were removed from the waste. The maximum effect of these two poisons can be seen by comparing results of the first and second series of calculations, shown in Fig. 31.

The number of fissions in the waste increases by more than a factor of 20 as the amount of water in the waste canister increases from 0 to 30%. When these counts are corrected by the fission monitor, the factor becomes 2.5. The factors are 10 and 1.8, respectively, for the case where the neutron poisons are present. For moisture contents <5% by volume, the boron and cadmium have little effect on the assay when fission monitor corrections are applied. In general, over the range of water concentrations considered here, the fission monitor does not correct

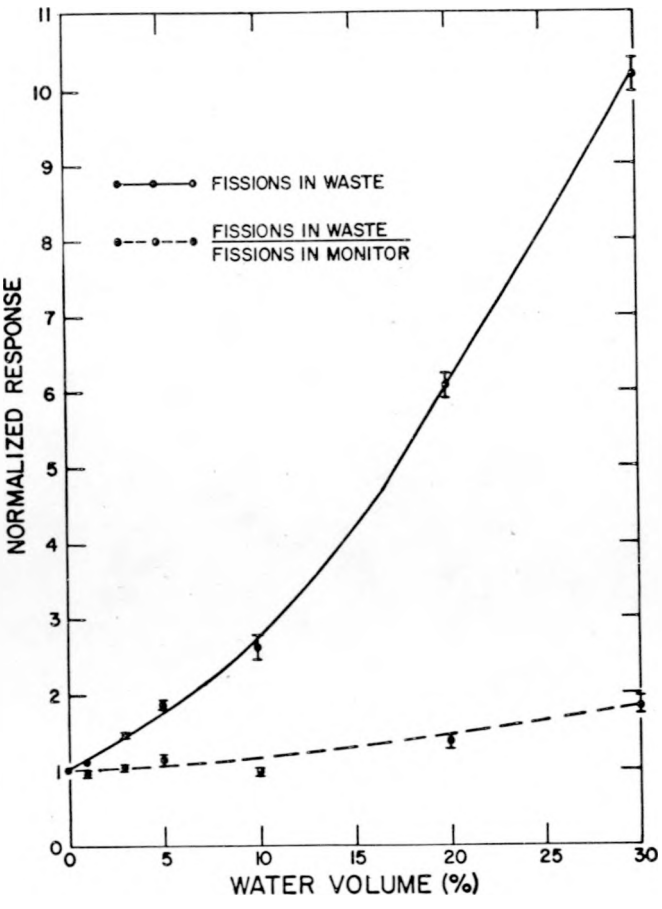


Fig. 30.
Relative number of fissions in the waste canister (solid curve) and relative number of fissions in the waste canister corrected by the fission monitor (dashed curve) as a function of the amount of water in the canister. In all cases the canisters contained 176 g fluorine, 13.9 g boron, and 34.8 g cadmium.

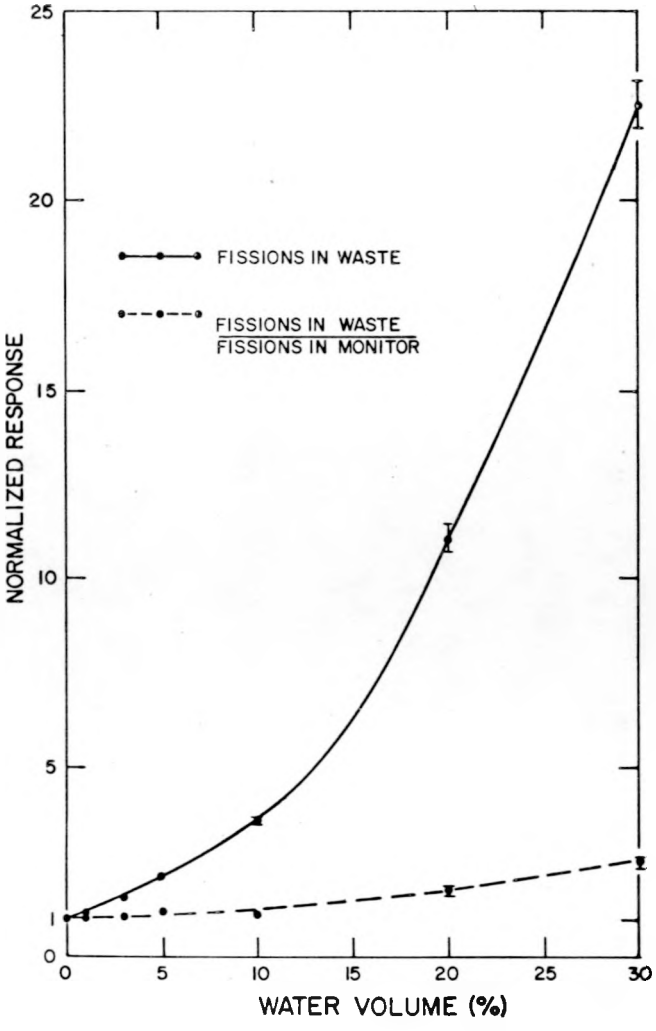


Fig. 31.
Relative number of fissions in the waste canister (solid curve) and relative number of fissions in the waste canister corrected by the fission monitor (dashed curve) as a function of the amount of water in the canister. The canister had a 4M fluorine concentration and no boron or cadmium.

as well for water in the absence of boron and cadmium. Therefore, the poisons are useful not only for criticality control but also for reducing the sensitivity of the assay instrument to water.

D. High Performance Fuel Laboratory (HPFL) Fuel Pin Scanner (M. S. Krick and H. O. Menlove)

The Hanford Engineering Development Laboratory (HEDL) is buying an intermediate fuel pin scanner for mixed-oxide, e.g., FFTF-type fuel, from a commercial source (delivery in 1979) and is planning to buy an advanced fuel pin scanner (delivery in 1981). Performance objectives for these systems are shown in Table IV. If the tighter specifications on total fissile accuracy and fuel column length accuracy are met for the intermediate scanner, they will also be applied to the advanced scanner.

TABLE IV

PERFORMANCE OBJECTIVES FOR THE INTERMEDIATE AND ADVANCED FUEL PIN SCANNERS

Item	Intermediate Scanner	Advanced Scanner
1. Scanning rate	20 pins/h	50 pins/h
2. Fissile content per pin		
Precision (2σ)	0.4%	---
Accuracy (2σ)	0.65%	0.75%
3. Fissile Uniformity		
Probability of false detection of nonuniformity per 0:64 cm	0.0002	0.0002
Relative deviation of fissile content per 0:64 cm with 95% detection probability (with 97% acceptance probability for nondeviant pin)	5%	2-3%
4. Accuracy of fuel column length measurement at 95% confidence level	0.038 cm	0.25 cm
5. Minimum detectable gap at 95% confidence level	0.025 cm	0.025 cm

LASL will assist HEDL in the evaluation of the commercial proposals for the intermediate scanner and will perform specific measurements and evaluations requested by HEDL in connection with the advanced scanner. In particular, LASL will study detector stability ^{252}Cf source strength requirements, and novel scanning techniques.

V. DETECTOR AND ELECTRONICS DEVELOPMENT

A. Multiplicity Measurements with Shift Register Coincidence Counter Electronics (M. S. Krick, P. Collinsworth, N. Ensslin, and J. E. Swansen)

The assay of plutonium by neutron coincidence counting is complicated by the presence of fission multiplication and by fissions induced by (α,n) neutrons. Because these induced-fission chains have a higher average neutron multiplicity than spontaneous fission, a measure of the multiplicity would provide information useful in making multiplication corrections.

The improved shift register coincidence counter (Ref. 8, p. 4) breadboard has been modified to allow scaling on multiplicities (m) from m = 1 to m = 6 in both the accidental and real-plus-accidental gates.

The multiplicity distribution in the accidental gate should be a Poisson distribution with a mean

determined by the total counting rate. Good agreement has been obtained between calculated and measured multiplicity distributions in the accidental gate. Multiplicity distributions in the real and accidental gates are being measured for ^{252}Cf, plutonium, and AmLi sources to study the potential usefulness of these distributions for analyzing multiplication effects in plutonium samples.

B. Data Processing Equipment for Development of Prototypical ^{252}Cf Shuffler Systems (G. Eccleston, T. Crane, and S. Bourret)

A minicomputer-based system for the data accumulation, analysis, and system control of prototypical ^{252}Cf Shuffler assay systems is being developed. This data-processing equipment will be used on Shufflers being designed for field test and evaluation programs. The initial equipment consists of a Nova 1200-type computer with 32-K core of 16-bit words. Additional features include a realtime clock, hardware multiply-divide, power fail/auto restart, and automatic program load. Hardware difficulties have been corrected and software is being developed.

Shuffler software is being developed using FORTRAN IV rather than using machine-dependent assembly language so that the software system will be usable with a minimum of modifications on most standard computers. In addition, the powerful features of FORTRAN will permit more sophisticated and useful coding.

The FORTRAN operating system has been written and several FORTRAN subroutines added to permit communication with CAMAC equipment. Such communication requires assembly language drivers. Interrupt driver assembly language software that is FORTRAN-callable has been written and checked for the Nova computer. Similar software has been received for the Digital Equipment Corp. computers. Development of the FORTRAN-based operating system permits wide user application by incorporating additional or replacement FORTRAN subroutines specific to a user's needs. However, for communication to new and nonstandard devices some assembly language drivers may still have to be written.

C. Electronic Engineering Developments (K. Johnson, D. Cramer, R. Dooley, R. Kuoppala, and J. Parker)

1. DYMAC Plutonium Solution Assay System (PUSAS)

The PUSAS system[18] has been developed for on-line analysis of plutonium solution samples for the Dynamic Materials Control (DYMAC) project. Each of the five assay stations has its own detection system and its own data reduction computer, which transmits assay results to the Eclipse accounting and materials control computing system. A movable control cart containing a Pace microprocessor analysis system with a terminal, paper-tape reader and monitor scope will be used to program the stations. The requirement that the control cart effect changes in the calibration and software for each station provides a level of tamper-resistance. The design of the system is essentially completed and software development is about to begin. Figure 32

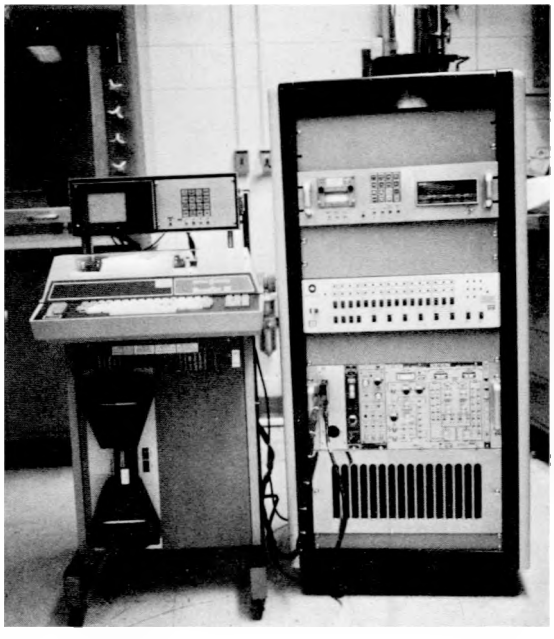


Fig. 32.
Electronic components for the PUSAS system. The portable cart used to load programs into the PUSAS minicomputer is shown on the left.

shows the electronic components, including the movable control cart, for the PUSAS system.

2. MEGAS Box Counter

A MEGAS-type box counter for the measurement of low-level radioactive waste was built. It differs from previous units in that it has a box-handler table. The counter is a low-profile, front-loading unit, shown in Fig. 33. The unit was thoroughly tested before it was delivered to TA-55 for use in the new plutonium facility.

3. SGS Barrel-Handler for the IAEA

A barrel-handler was added to the segmented gamma scanner (SGS) with various modifications to allow contiguous operation of the barrel-handler and the can-handler. The barrel-handler is shown in Fig. 34. To obtain high sensitivity measurements for low-activity samples, we modified the SGS so that the entire barrel could be viewed without the need for segmented scanning. The unit is now complete, tested, and ready to be shipped to the IAEA. The complete SGS system, with barrel-handler, is

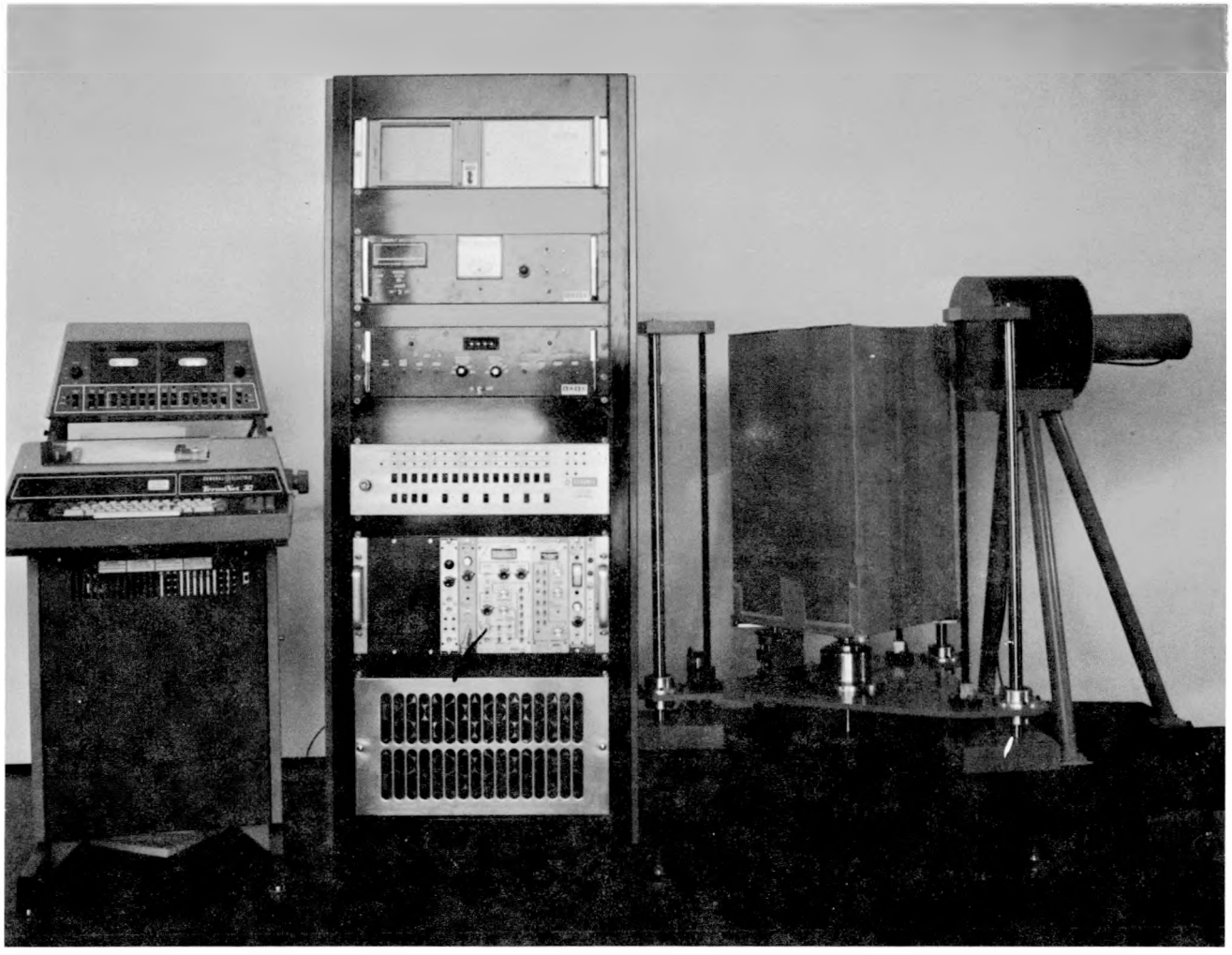


Fig. 33.
MEGAS-type box counter for the measurement of low-level radioactive waste. This unit was recently installed in the new LASL Plutonium Facility (TA-55).

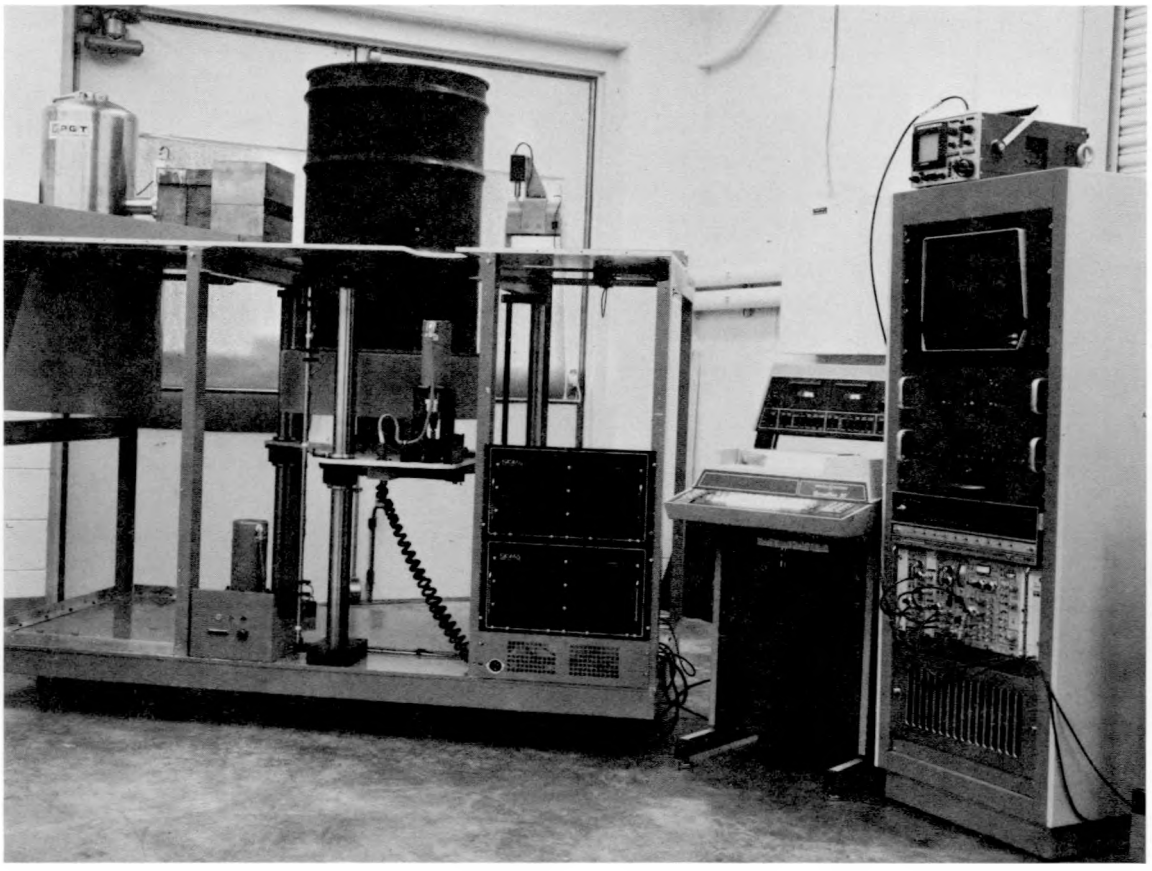


Fig. 34.
SGS with barrel-handler incorporated. (Sides of barrel-handler have been removed to show the interior.) The unit is being evaluated by the IAEA.

scheduled to be delivered in January 1978 to Casaccia, Italy, for in-plant tests by IAEA staff members.

4. Computer System for Safeguards Research

The computer system that has been ordered is a DEC 11/60 with 64-K memory, 17.5 Mbytes of disk, dual floppy disks, Decwriter, GE Terminet, and a VT-11 (vector graphics CRT display). We expect delivery at the end of February 1978.

The site for the computer is to be TA-35, Building 2, Room C-160. Group ENG-4 has initiated the paperwork and specifications to have the room modified and environmentally controlled. The projected completion date is February 15, 1978.

VI. STANDARDS AND MEASUREMENT CONTROLS

A. Increasing the Measurement Accuracy of the CMB-8 Random Driver (D. G. Langner, N. Ensslin, D. Lee, and H. R. Baxman*)

We have tried to determine whether the measurement uncertainties could be reduced for three material categories routinely assayed by the RD. Two of the three material types—burned metal turnings and casting residues—consist of pure U_3O_8 and from the standpoint of material accounting are the most important samples now measured by the RD. The third material type is hydrofluoric slag (HFS) waste.

*Group CMB-8.

Formerly, a single calibration curve, based on standards of U_3O_8 mixed with graphite, was used to assay all material types (Ref. 4, p. 9). To improve the measurement uncertainties for the three material types in question, new and separate calibrations were made for pure U_3O_8 and the HFS waste. For U_3O_8 the calibration (Fig. 35) was made with U_3O_8 standards containing 250, 500, 1000, 1500, and 2000 g of uranium. Over this mass range the assay accuracy is 2 to 3%, which is adequate for accounting purposes. For samples containing >2000 g, assay uncertainties increase significantly because of neutron absorption and self-multiplication effects; consequently, we decided to limit the sample size to 2000 g for all assays of these materials.

Because of the complex nature of the HFS waste, calibration could not be accomplished by simply building standards. Instead, representative waste samples were selected and assayed by segmented gamma scanning (see Part 1, Sec. VI-D). The SGS results were then used as the nominal "standard" uranium content of the HFS samples. The response of the RD to these low-fissile mass samples was increased to a statistically acceptable level by placing the samples in a polyethylene sleeve for measurement. The resulting HFS calibration curve is given in Fig. 36. Over this uranium mass range, a 5 to 10% measurement accuracy is possible.

Other changes included updating of the RD's data reduction software to include these new calibrations. Also, the automatic-calibration sequence (Ref. 3, p. 43) now occurs quarterly instead of monthly. Through a series of checks, the user is warned if his assay is outside the permissible mass range. The printed output shows which calibration is being used to reduce the RD data, when that calibration was last updated, and whether the polyethylene sleeve is required for a valid assay.

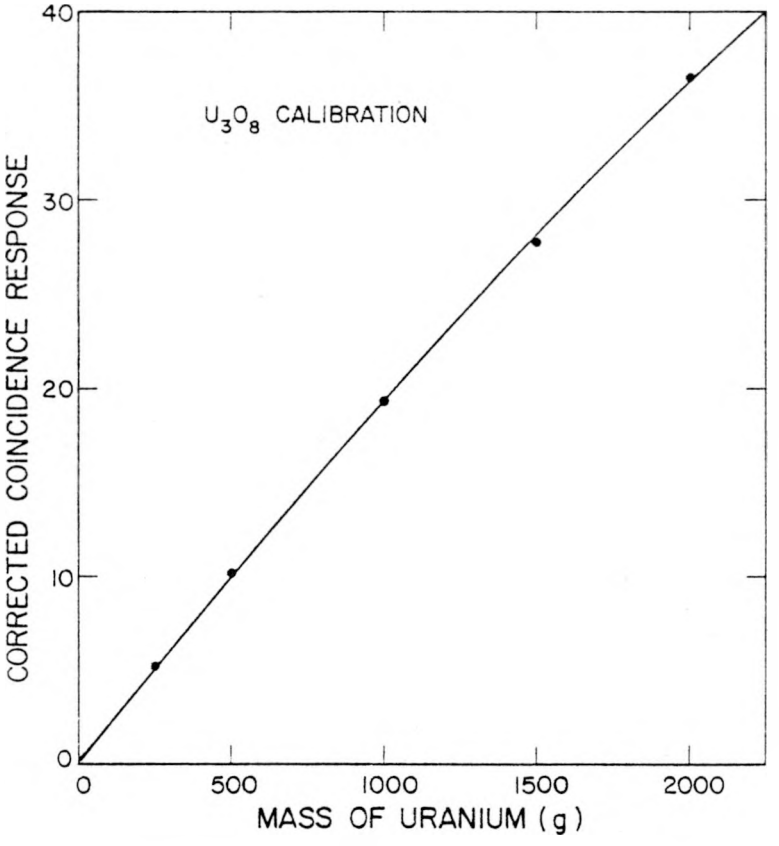


Fig. 35.
Calibration curve for measurement of U_3O_8 with the CMB-8 random driver.

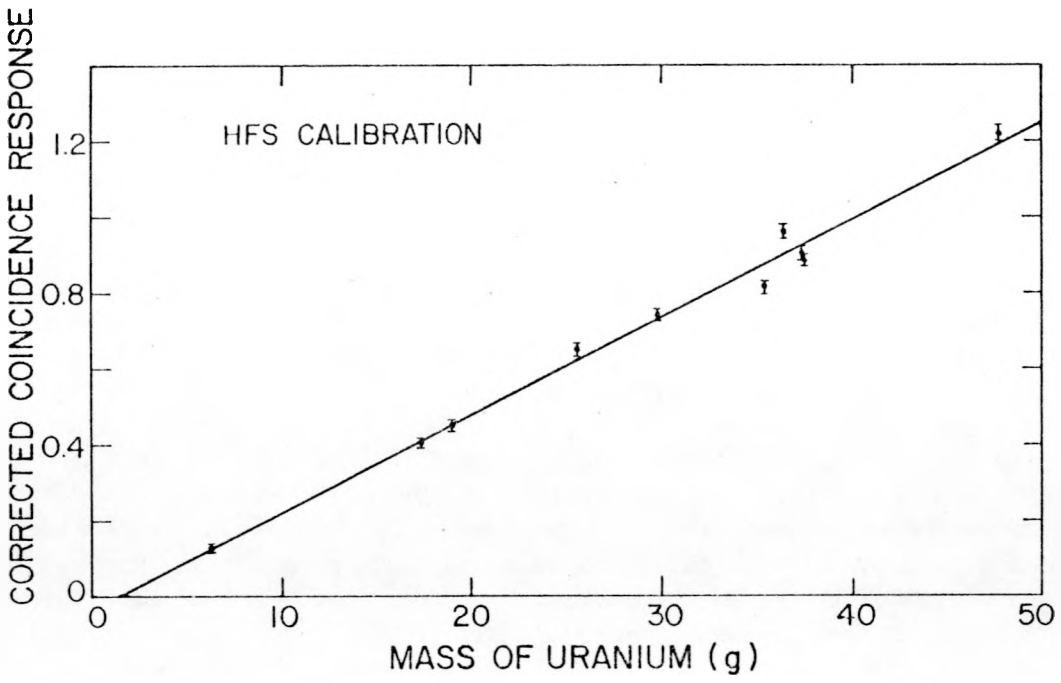


Fig. 36.
Calibration curve for measurement of HFS with the CMB-8 random driver.

B. Enrichment Verification of USAS Type III Solutions (D. G. Langner)

At the request of CMB-8, we made a study to determine whether the enrichment could be verified for four Type III solutions assayed between March and May 1977. Type III solutions (50 to 500 g/ℓ) are assayed on the uranium solution assay system (USAS) by a K-edge densitometry technique. This method depends only on the elemental uranium density of the solution and cannot distinguish between a solution containing depleted uranium and a solution containing enriched uranium.

For all assays, USAS measures the ^{235}U 186-keV and the (^{169}Yb transmission source) 110-, 130-, 177-, and 198-keV gamma-ray peak areas. Only the 100- and 130-keV transmission data are used to deduce a Type III result, but all the information is stored and thus was available for the study.

The concentration of ^{235}U in the solutions in question was deduced from the 186-keV peak area and the transmission data at 177 and 198 keV. The transmission data were used to determine the sample self-absorption correction factor, which for the USAS Type III geometry is approximately equal to that for a far-field slab geometry, i.e.,

$$CF = -\ln T_{186} \Big/ (1 - T_{186}) \quad ,$$

where T_{186} is the transmission measured through the sample at 186 keV. T_{186} is determined by a linear interpolation between the values measured at 177 and 198 keV.

The correction factor was determined with standard solutions assayed concurrently with the four samples in question. The results are summarized in Fig. 37, where the 186-keV peak area per unit solution density is plotted against the self-absorption correction factor. The equation given in the figure is a linear least squares fit of the data.

Figure 38 shows the assay results for the four samples plotted against the concentration of ^{235}U predicted on the basis of the densitometry assay and nominal 93.15% enrichment. For all the samples, the transmission-corrected assay agrees with the K-edge densitometry assay to within estimated errors, thus verifying the nominal enrichment. Further, these results suggest that this enrichment verification can be performed routinely to within ±5% for this range of concentrations.

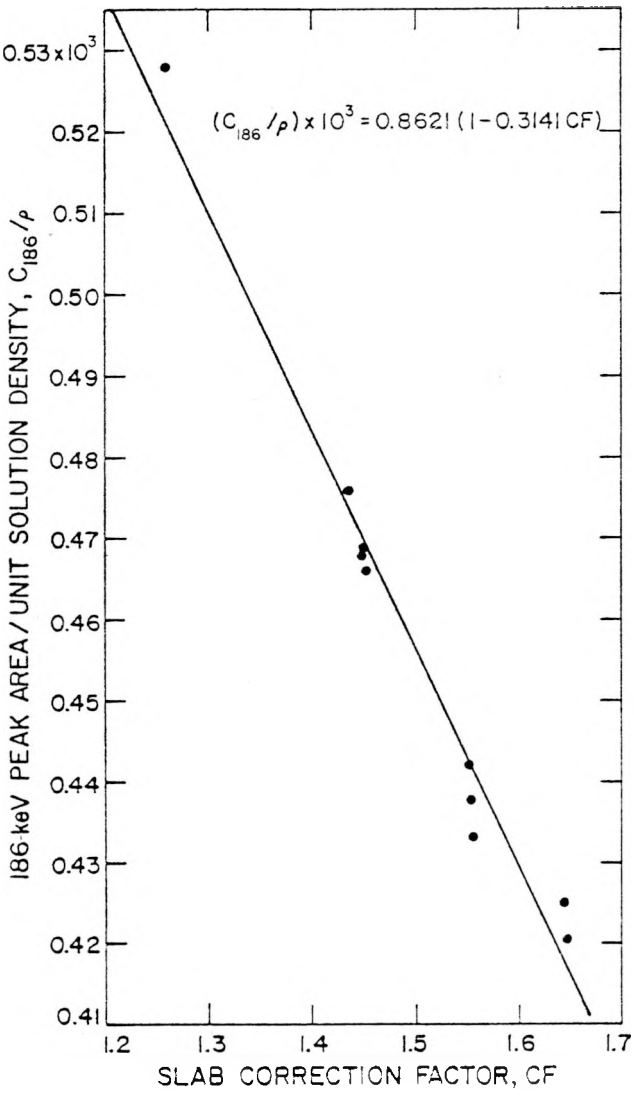


Fig. 37.
Experimental data used to derive the sample attenuation correction for USAS Type III solutions. The equation given is a least squares fit of these data.

C. Evaluation of the CMB-8 USAS (D. G. Langner, T. R. Canada, and H. R. Baxman)

The CMB-8 USAS has been operating for almost 2 yr, during which time a two-phase program has been under way to determine USAS's performance in the measurement of Type II (1 to 50 g/ℓ) and Type III (50 to 500 g/ℓ) solutions. In phase I, assay results from the USAS were compared with chemical analyses, and daily performance was monitored by measuring known solution standards. In phase II, we had a tighter system of daily measurement controls based on secondary standards.

During phase I, 30 Type II samples were sent to CMB-1 for chemical assay. Figure 39 shows the relative difference between the USAS assay and the chemistry results for these samples. The figure also shows 10-day averages of the relative difference between the USAS assay and the nominal concentration of known solution standards. The solution standards used for these measurements were prepared by CMB-1 to a concentration accuracy of ±0.3%.

An average bias of −4.3% exists between the chemistry and USAS assay values; however, the ±4.6% average deviation in this bias is large. In comparison, the ±1.2% average deviation of the USAS-standards data is small. Still, this deviation is 1.5 to 2 times larger than predicted from counting statistics and calibration errors. There is also a nonrandom behavior around zero.

To evaluate the USAS's performance for Type III samples, 28 Type III samples were analyzed chemically. Figure 40 gives the relative difference between the USAS assay and the chemistry results. Ten-day average relative differences between USAS and the nominal concentrations of standards are also plotted. Group CMB-1 prepared the solution standards for the Type III evaluation to ±0.1% accuracy.

As with the Type II solutions, the bias with chemistry, 1.9 ± 3.2%, is large in comparison to the USAS-standard data bias, −0.9 ± 1.0%. However, after the USAS was recalibrated with fresh standards in December 1976, the bias relative to the chemistry data decreased. The USAS-standard data from December 1976 to March 1977 showed a nonrandom decreasing trend.

The USAS-standard data for phase I indicate that the large chemistry-USAS biases cannot be explained by instrumental drifts or measurement reproducibility. Therefore, these biases must be caused by problems either with the original calibration standards or with chemistry procedures.

Changes in the solution standards may have caused some of the nonrandom behavior in the USAS-standard data, particularly the Type III behavior evident between December 1976 and March 1977. Visual inspection of the solution standards revealed that they were evaporating during the measurement period. Chemical analysis of a

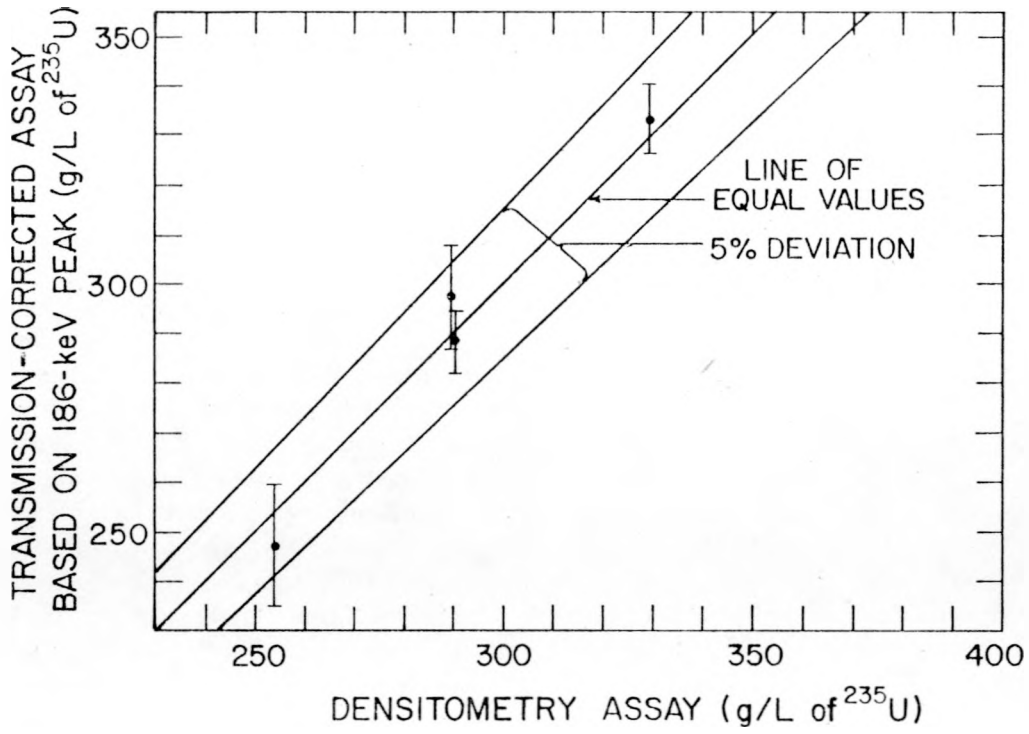


Fig. 38.
Comparison of the transmission-corrected assay results with the densitometry assay results for four Type III USAS samples.

number of these standards showed an average concentration 20% greater than the prepared nominal value.

In phase II of the USAS evaluation, we instituted new daily measurement-control procedures and abandoned the daily measurement of solution standards because of the evaporation observed during phase I. Secondary foil standards encased in plastic were measured daily, and four measurement parameters were monitored. Periodically these secondary standards were referred to freshly prepared solution standards as a calibration check.

For Type II materials, the pulser rate, the transmission factor, and the 186-keV peak area were monitored during the daily foil measurement. Figure 41 gives 10-day averages for these parameters plotted against date. These data demonstrate that systematic uncertainties for this material type approximately meet the 0.5% design criterion. No chemistry comparisons were made during phase II because of a reduction in the CMB-8 staff.

For Type III samples, we monitored the assay value for the daily foil measurement. Ten-day averages of this assay value are given in Fig. 42. Also plotted is the relative difference between USAS and chemistry analyses for five samples. The scatter in the chemistry-USAS data is quite large compared to the scatter in measurement-control data. The Type III foil assay results show excellent (±0.4%) long-term reproducibility of USAS for Type III measurements.

The overall agreement between USAS and chemistry results is poor. The scatter in these data is larger than the ±2% accuracy claimed by the chemistry laboratory. The data in phases I and II indicate that the bias and scatter observed in the chemistry-USAS comparison data may be due to the complex chemistry involved in the sample preparation as well as the chemical analysis procedures.

The bias between chemistry results and USAS Type III results during April to December 1976 probably was due to evaporation of the solution standards before calibration. Figure 43 shows the chemistry-USAS data re-evaluated with the

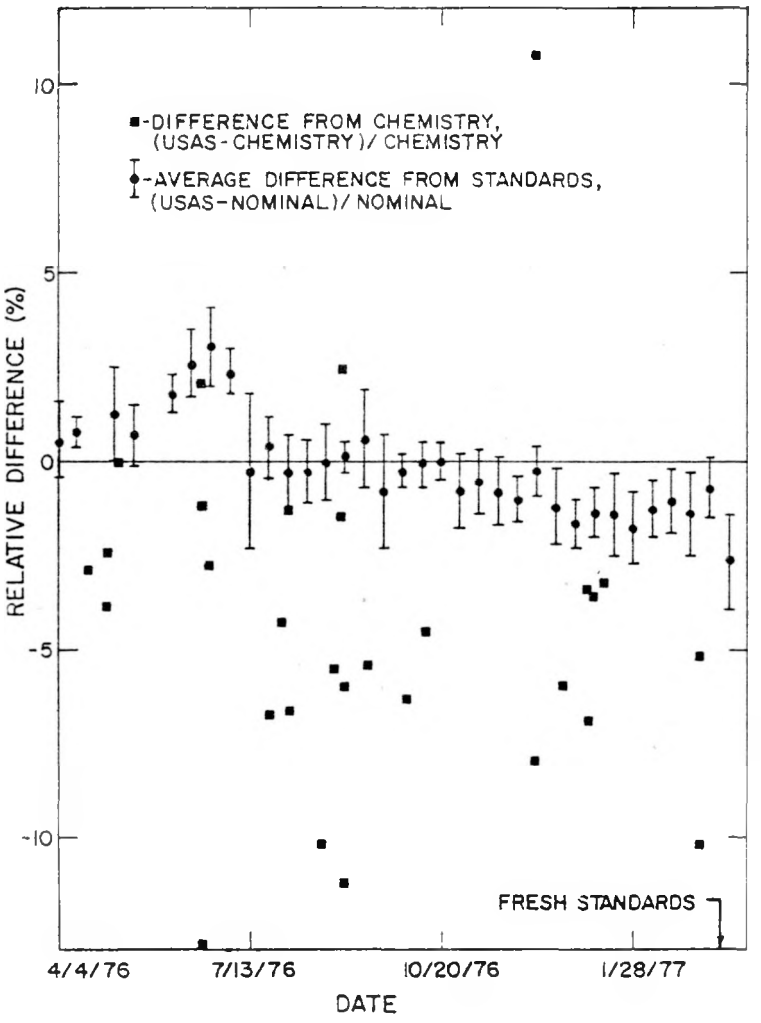


Fig. 39.
USAS performance for Type II samples during phase I.

calibration constants obtained from freshly prepared standards in December 1976.

Some of the USAS-chemistry scatter may be attributable to variations in pipetting techniques. All USAS assays assume that the sample container holds 20 mℓ of solution. Although we have been assured that pipetting can be performed routinely to an accuracy of <0.5%, sample volume variations have been identified as a potential source of bias and scatter undetectable by the measurement controls.

Phase II data show that the USAS routinely performs within its design parameters. On the assumption that the samples are pipetted to ≤0.1% accuracy, the USAS performance ratings are:

Type II

Assay range:	1-50 g/ℓ of uranium
Assay accuracy:	0.5 to 1.0%
Assay time:	400 to 800 s

Type III

Assay range:	50 to 500 g/ℓ of uranium
Assay accuracy:	0.5 to 1.0%
Assay time:	1000 to 2000 s.

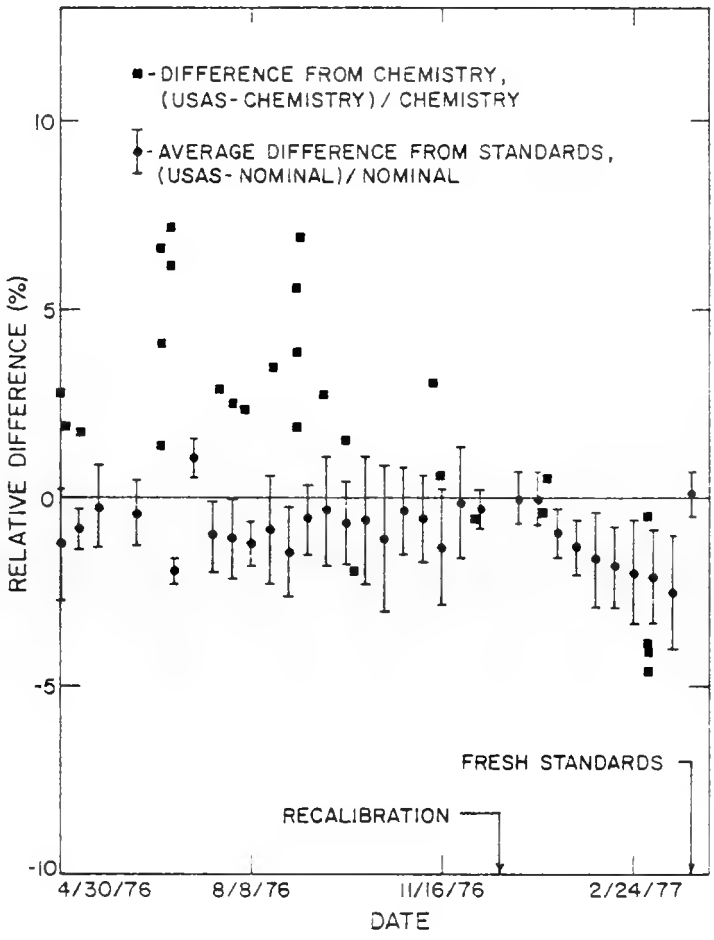


Fig. 40.
USAS performance for Type III samples during phase I.

Figures 44 and 45 summarize these results for both a new and a decayed ^{169}Yb transmission source.

Although the USAS's performance meets its design criteria, certain areas still need improvement. Recommended hardware improvements include changing the single-point gain stabilization to two-point stabilization and redesigning the sample holder. The former will reduce those variations in the Type II 186-keV count rate that are caused by drifts in the high-energy gain. The new sample holder should be designed to make the USAS measurements independent of pipetting errors.

Desirable software/analysis enhancements include verifying Type III enrichment using 186-keV peak area information, checking Type III samples for large-density matrix contamination by extrapolating transmission information to the K-edge and updating the Type II calibration daily based on the foil secondary standard. The daily updating will improve the measurement accuracy by correcting for long-term drifts.

D. Passive Gamma-Ray Assay in Support of Random Driver Calibration Studies (J. L. Parker)

For several years an RD in the LASL uranium recovery facility has been used successfully to determine nondestructively the ^{235}U content in containers of scrap and process residues. Container volumes have been as large as ~8 ℓ, containing ≥500 g of uranium in as much as ~8 kg of matrix material.

Currently, we wish to assay the ^{235}U content of a process residue (in the usual containers) containing

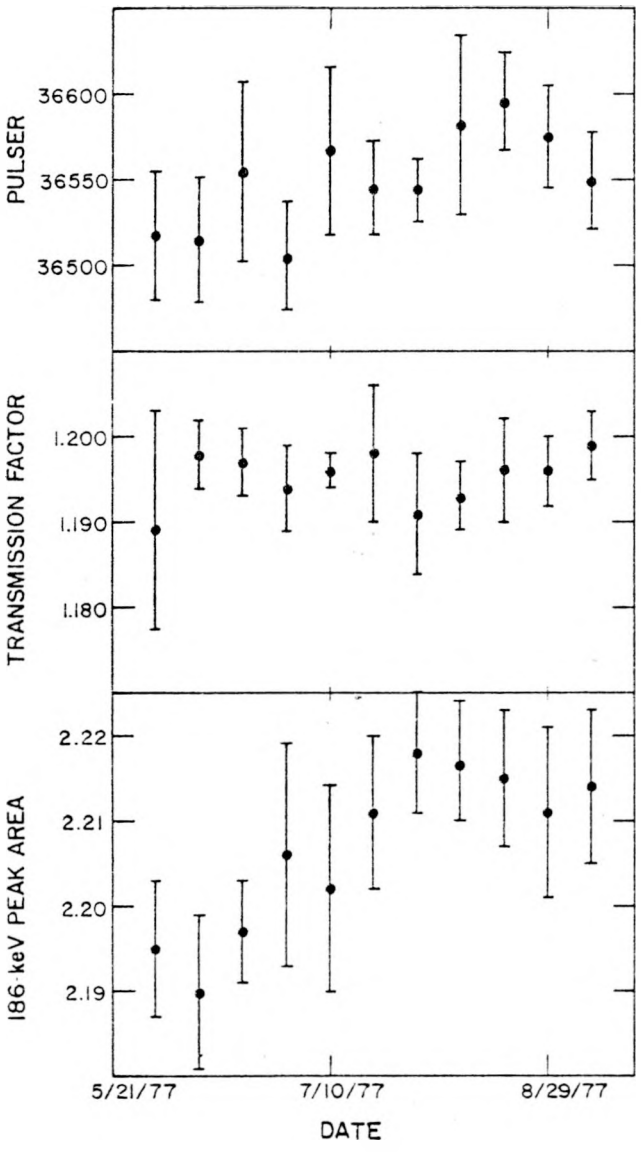


Fig. 41.
USAS performance for Type II samples during phase II.

only 10 g of uranium. Because of the large differences in uranium concentration and chemical composition, we could not extrapolate directly from the calibration of the earlier system to the assay of the HFS residue. Consequently, we made passive gamma-ray assays of HFS residues to provide secondary standards for the calibration of the HFS material in the RD. In that way, RD assay can be used in the recovery facility for another category of material. The gamma-ray assays also indicate the attainable accuracies of passive gamma-ray assay on suitable materials.

In the first step of the gamma-ray assay, four cans of the HFS residues were milled and blended, then repackaged in fifteen 2-ℓ polyethylene bottles. (A transmission-corrected gamma-ray assay produces highly accurate results with such a configuration.) The 15 bottles of HFS residue were assayed by a transmission-corrected segmented gamma-scan procedure. The results indicated that we could have assayed the material in the original cans, which have an 18.7-cm diameter instead of the 11.5-cm diameter of the 2-ℓ bottles. Hence, we put the material into the original four cans and assayed again.

Because the segmented scans indicated that the milled and blended material was of uniform density and composition, we also performed a far-field assay in which the detector was located ~1 m from the cans and segmentation was not used. This procedure precluded systematic errors that could result from the dissimilar diameters of the sample cans and the 2-ℓ bottles and from small end-effect errors in the segmented scanning procedure. The values of four batches of HFS material measured by three types of passive gamma-ray assay are summarized in Table V. Precision of all measurements was 1 to 1.5% at the 1-σ level. The excellent consistency of the results indicates that the predicted precision was attained and that the corrections made for different-size containers and different assay geometries were correct.

We had the opportunity to check the absolute accuracy of the assay results as well because Group Q-1 was requested to assay a set of small samples from the same four batches of milled and blended HFS material on which we had made the passive gamma-ray assay. Wet chemical analysis of the small samples had shown scatter in the results. The NDA was made to determine whether a sampling problem existed despite milling and careful blending. Samples were taken from the top, middle, and bottom of the four HFS batch cans into both 3-dram and 1-dram vials—a total of 24 small samples of mass varying from ~0.5 to ~6.5 g and of uranium content varying from ~3 to ~30 mg. Each of the 24 samples was assayed by an active interrogation delayed-neutron method; in addition, each of the twelve 3-dram-vial

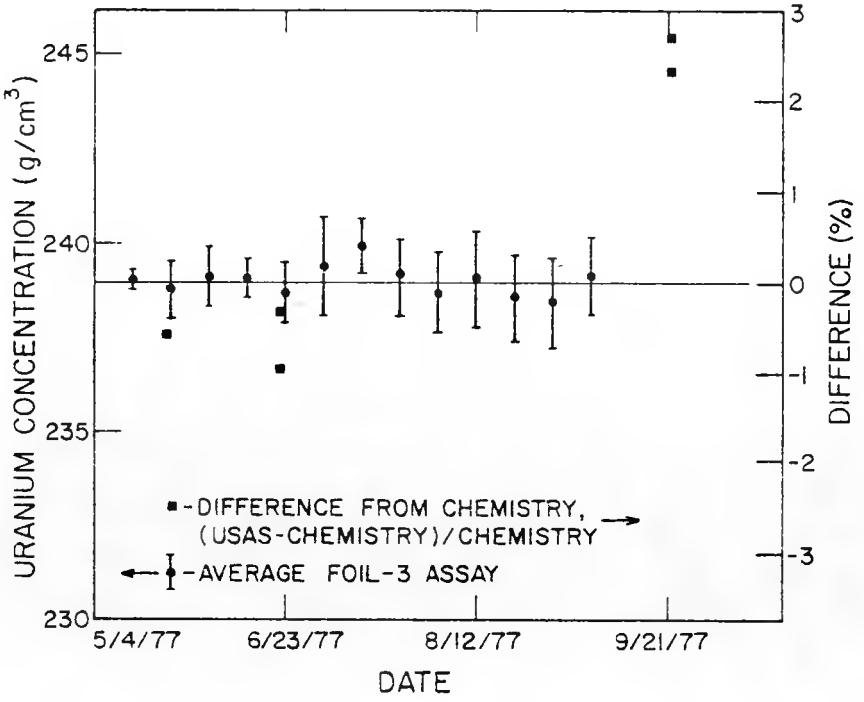


Fig. 42.
USAS performance for Type III samples during phase II.

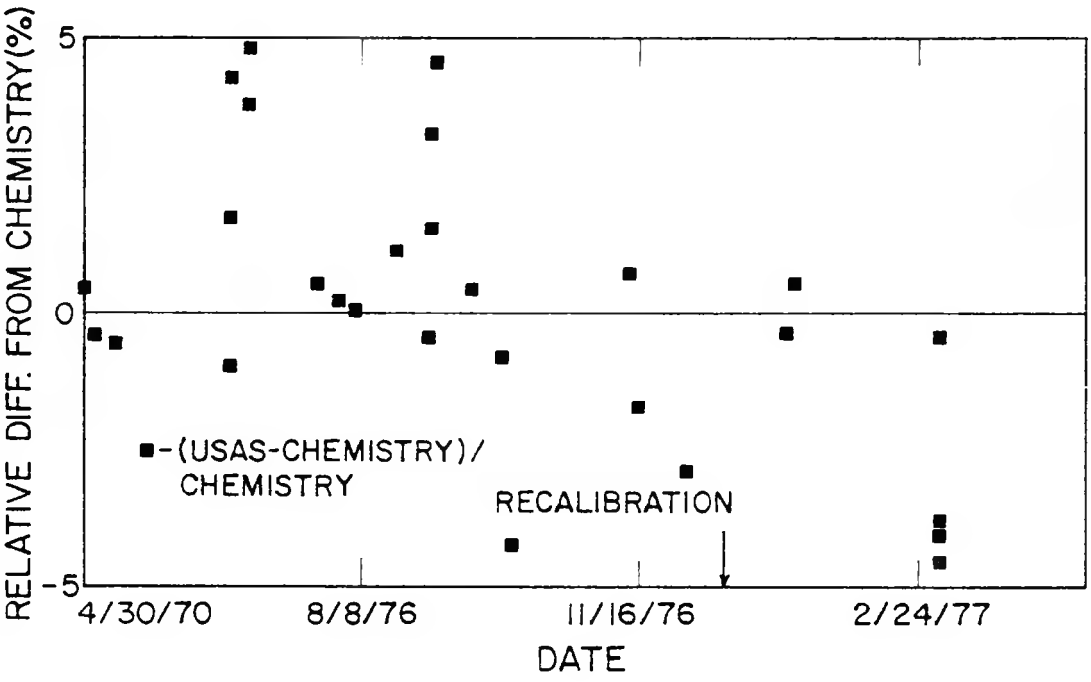


Fig. 43.
Phase I USAS (Type III)-chemistry comparison data corrected for calibration error.

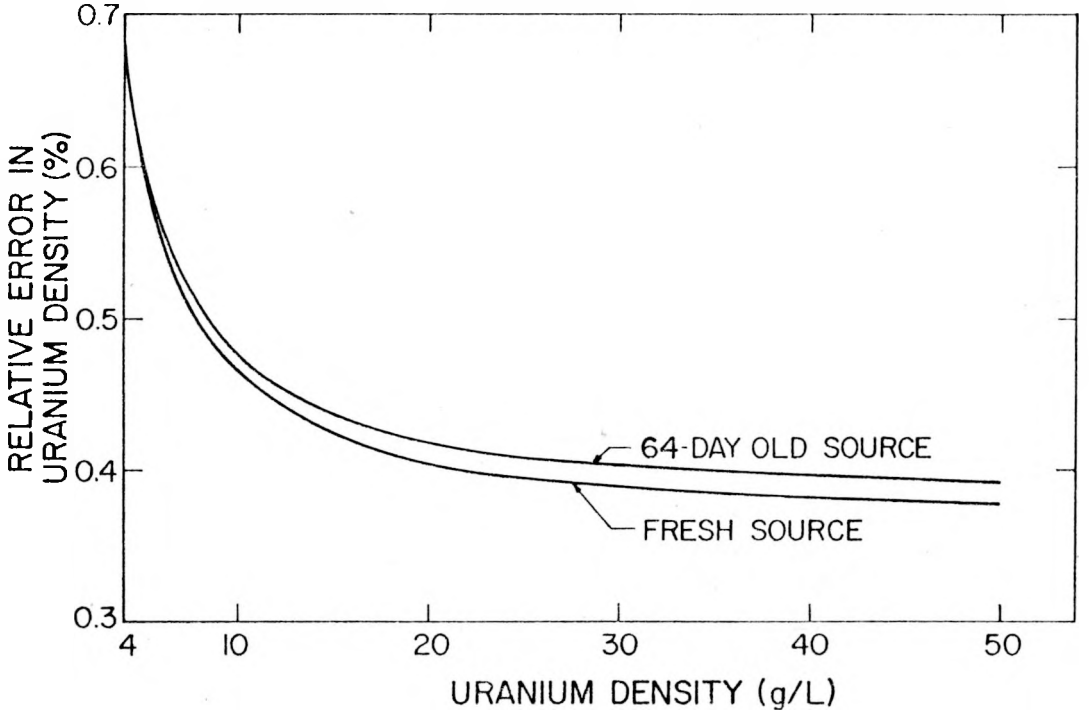


Fig. 44.
Relative error in a Type II assay as a function of uranium density ρ_u for a 400-s count.

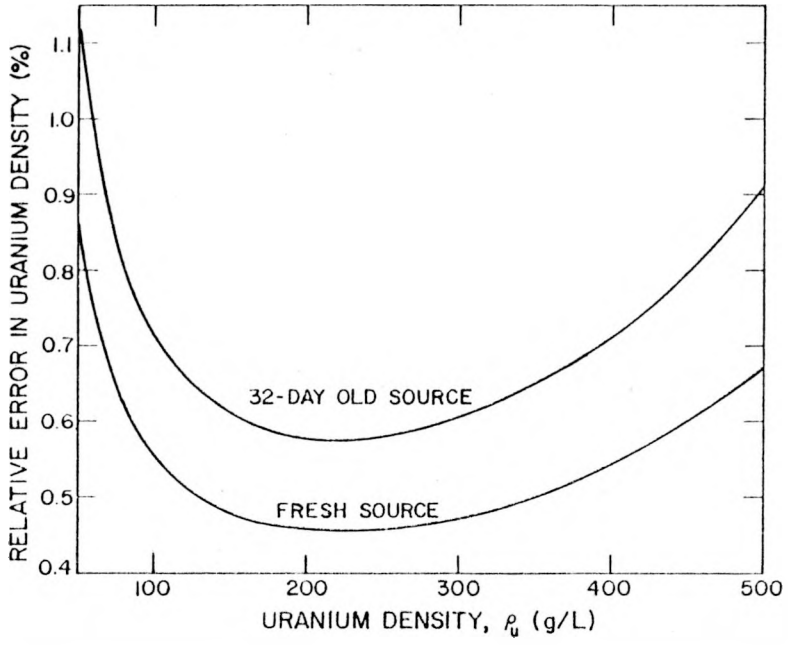


Fig. 45.
Relative error in Type III assay as a function of uranium density ρ_u for a 1000-s count.

TABLE V

^{235}U VALUES DETERMINED IN HFS MATERIAL BY THREE KINDS OF PASSIVE GAMMA-RAY ASSAY

Batch	Results on 2-ℓ Bottles (g ^{235}U and 1-σ Precision[a])	Results on Large Cans (g ^{235}U and 1-σ Precision[a])	Far-Field Results on Large Cans (g ^{235}U and 1-σ Precision[a])
HFS435-203	6.02 ± 0.09		
	5.88 ± 0.08		
	5.88 ± 0.09		
	5.99 ± 0.09		
Total	23.77 ± 0.18	23.96 ± 0.31	23.69 ± 0.28
HFS435-205	5.87 ± 0.09		
	5.67 ± 0.09		
	6.01 ± 0.10		
Total	17.55 ± 0.16	17.37 ± 0.26	17.87 ± 0.27
HFS435-244	8.58 ± 0.10		
	8.97 ± 0.11		
	8.75 ± 0.11		
	8.40 ± 0.11		
Total	34.70 ± 0.20	35.07 ± 0.39	35.01 ± 0.35
HFS435-248	8.06 ± 0.11		
	8.27 ± 0.12		
	8.50 ± 0.13		
	7.79 ± 0.12		
Total	32.62 ± 0.24	33.39 ± 0.37	33.05 ± 0.33

[a]Estimated precisions for all measurements are based on counting statistics only.

samples was carefully assayed by a transmission-corrected passive gamma-ray procedure. The two measurement methods are based on radically different physical principles, the only point of contact in the two methods being the use of the same physical standard, a 3-dram vial containing 4.209 g of graphite and 0.1184 g of U_3O_8 at 93.18% enrichment. The two sets of results are given in Table VI. We estimate a ±3% precision (1-σ level) for the delayed-neutron assay results and ±2% to ±4% precision (1-σ level) for the gamma-ray results. The uncertainties of the two sets of results overlap in 10 of the 12 cases. The excellent agreement in total ^{235}U masses for the two sets of assays is persuasive (but not conclusive) evidence of the accuracy of both methods in small-sample assays of the HFS materials.

Knowing the masses of all the small samples as well as the masses of the batches from which they came, we could compute the uranium concentration for both the small samples and the batches. The concentrations are compared in Table VII. First, the uranium concentrations of the three samples from any given batch agree within the estimated precisions, indicating that the uranium concentration of the milled and blended material was ±3% uniform

TABLE VI

SMALL-SAMPLE ^{235}U VALUES DETERMINED IN HFS MATERIAL BY PASSIVE GAMMA-RAY ASSAY AND BY ACTIVE DELAYED-NEUTRON ASSAY

Batch	Sample	Passive Gamma Results (mg ^{235}U and 1-σ Precision[a])	Delayed-Neutron Results (mg ^{235}U)[b]
HFS435-203	Top	26.54 ± 0.53	25.8
	Middle	8.60 ± 0.33	8.85
	Bottom	15.06 ± 0.41	16.2
HFS435-205	Top	13.08 ± 0.39	12.7
	Middle	24.81 ± 0.50	23.8
	Bottom	8.42 ± 0.33	8.42
HFS435-244	Top	11.72 ± 0.35	11.7
	Middle	26.77 ± 0.54	26.8
	Bottom	40.32 ± 0.60	40.3
HFS435-248	Top	12.47 ± 0.34	12.5
	Middle	41.05 ± 0.62	41.2
	Bottom	23.34 ± 0.47	23.4
	Total	252.2 ± 1.6	251.6

[a]The 1-σ precision is based only on counting statistics.

[b]The delayed-neutron results have an estimated probable error of ±3%.

throughout a batch and thereby eliminating concern about a major sampling problem. Second, the agreement between the batch concentrations and sample concentrations is additional evidence of the accuracy of passive gamma-ray methods in assaying both small and bulk samples. Taking into account the possible calibration errors and known systematic errors, we claim an accuracy no worse than ±3% at the 1-σ level for all the passive gamma-ray assays reported here.

The foregoing study, particularly the assay results of the milled and blended HFS material in the original 18.7-cm-diam cans, indicated that as-canned HFS material might be assayed successfully if it were not too heterogeneous. Because we demonstrated that the differences in diameter and in density between the standards and the cans of HFS material could be corrected for, several cans of the as-canned HFS material were test-assayed. Although the gross sample density and uranium concentration varied somewhat, the variations could be accommodated by the transmission-corrected, segmented gamma-ray assay procedures. Hence, we assayed 10 cans of as-canned HFS material, ranging in uranium content from <6 to >134 g of ^{235}U, to assist in establishing the RD calibration over that broad ^{235}U mass range. Although the accuracy cannot be as good as for the milled and blended material, it probably is no worse than ±5% (1-σ level), which is adequate.

E. Passive Gamma-Ray Assay Applications (J. L. Parker)

During this reporting period, Group Q-1 assisted other LASL groups by making the passive gamma-ray assays listed in Table VIII. We made ^{235}U assays

TABLE VII

^{235}U CONCENTRATIONS DERIVED FROM SMALL-SAMPLE MEASUREMENTS AND FROM BATCH MEASUREMENTS

Batch	Sample	Small-Sample Concentrations (mg ^{235}U/g & 1-σ Precision[a])	Batch Concentrations[b] (mg ^{235}U/g & 1-σ Precision[a])
HFS435-203	Top	4.02 ± 0.08	
	Middle	4.02 ± 0.15	4.01 ± 0.04
	Bottom	3.88 ± 0.10	
	Av	3.97	
HFS435-205	Top	3.88 ± 0.12	
	Middle	3.76 ± 0.08	3.70 ± 0.04
	Bottom	3.80 ± 0.15	
	Av	3.81	
HFS435-244	Top	6.01 ± 0.18	
	Middle	5.98 ± 0.12	5.98 ± 0.06
	Bottom	5.88 ± 0.09	
	Av	5.96	
HFS435-248	Top	5.72 ± 0.16	
	Middle	5.74 ± 0.09	5.70 ± 0.06
	Bottom	5.66 ± 0.11	
	Av	5.70	

[a]All precisions are estimates from counting statistics only and, in particular, do not include possible error in the reported sample and batch masses.

[b]Batch concentrations are based on the average of the batch-assay values obtained from the three procedures used.

of uranium-bearing scrap, waste, and ash for Group CMB-8 and a ^{241}Am assay for CMB-11. The ^{235}U assays required the use of several different geometries because the materials varied so widely in size, shape, density, chemical composition, and uranium concentration.

F. Use of the Van de Graaff Small Sample Assay Station (J. D. Brandenberger and E. L. Adams)

The Van de Graaff small sample assay station (SSAS) was used recently for uranium assays needed by CMB and AP Divisions. Group Q-1 received from CMB-8 a total of 24 samples weighing <10 g each—6 samples from each of 4 large containers. The samples were highly enriched uranium in the form of UO_2 or U_3O_8 in a powdered ZrO lattice. With the SSAS, we measured the ^{235}U content of the samples within an estimated probable error of ±3%. The results are shown in Table IX. The passive gamma measurements made on 12 of the samples are given in Part 1, Sec. VI-D, and are in excellent agreement with the SSAS assays.

Group AP-3 requested a set of small-sample assays on seven vacuum pumps and a bag of filters thought to contain significant amounts of ^{235}U. The SSAS assay (see Table X) showed a ~1-g total of ^{235}U in all the pumps and filters.

TABLE VIII

PASSIVE GAMMA-RAY ASSAYS MADE FOR OTHER LASL GROUPS

No. Packages	Sample Type	Isotope Assayed
12	30-gal drums containing furnace parts and process residues	^{235}U
2	28-ℓ boxes containing rags and paper	^{235}U
8	1- and 2-gal cans containing rags	^{235}U
6	1- and 2-ℓ bottles of ash	^{235}U
6	250-ml bottles containing $\leq$50 ml of high-concentration ash	^{235}U
1	1-gal cans containing miscellaneous waste	^{241}Am

These examples demonstrate the Van de Graaff's usefulness in performing accurate assays of small amounts of fissionable material in a variety of sample configurations.

G. Van de Graaff Assay of Large U_3O_8 Standards (E. L. Adams and M. S. Krick)

Experimental Van de Graaff assays were performed on large, 93%-enriched U_3O_8 standards to develop a backup assay system for the CMB-8 RD (see Part 1, Sec. VI-A). The active assay technique used fast neutron interrogation and delayed-neutron counting. The fast neutrons (~400 keV) were produced from the $^7Li(p,n)$ reaction with a 3.7-mg/cm^2 lithium metal target and a 25-μA beam. The delayed neutrons were counted with the superslab detector.[19]

Because the U_3O_8 containers were too large (~18-cm diam) to be handled in the interrogation chamber of the superslab detector, the assays were performed with the geometry shown in Fig. 46. The superslab was partially disassembled by removing the front doors and the sample-handling components; it was then positioned 1.5 m from the lithium target. The sample was placed midway between target and detector.

To flatten the neutron flux interrogating the sample, the sample was placed in a lead cavity,. The cavity, with interior dimensions of ~30 by 30 by 40 cm and ~10-cm wall thickness, provided a response having an average measured deviation of 3.2% throughout the cavity.

U_3O_8 standards with ^{235}U masses of 250, 500, 1000, 1250, 1500, and 2000 g were assayed in the lead-walled cavity using a sample rotator. Measurements were made with and without cadmium covers on the samples. The sensitivity of the assay system as described was 0.26 counts/s/g of ^{235}U. The background counting rate was ~3 counts/s.

The results of the measurements on the U_3O_8 standards are given in Fig. 47. Weighted least squares fits were performed with the response function $R = Am/(1 + Bm)$, where R is the detector response, m is the sample mass, and A and B are calibration parameters. The 1250-g sample is packaged in a container of smaller diameter and shows larger self-absorption effects than the other samples, so it was not used in the least squares fitting procedure. With the response R normalized to 250 g for the 250-g sample, the fitting parameters for measurements on the cadmium-covered samples are $A = 0.949$ and $B = -8.81 \times 10^{-6}$; for the measurements with no cadmium cover, the parameters are $A = 0.984$ and $B = 8.15 \times 10^{-5}$. Figure 47 shows the two calibration curves and the straight line $R = Am$, where A is the parameter for the measurements of cadmium-covered samples. The rms per cent deviation of the measurements relative to the calibration curve for the cadmium-covered samples (including the 1250-g sample) is 2.6%. Assay accuracies could approach 1% if the calibration curve were produced more carefully and if the container dimensions were constant.

TABLE IX

ASSAY VALUES OF ^{235}U IN SAMPLES FROM GROUP CMB-8

CMB-8 Container No.	Bottle No.	Net Weight of Sample (g)	CMB-8 Estimate[a] of ^{235}U (mg)	SSAS Assay ^{235}U (mg) (±3%)
435203	148	2.7865	15	12.4
	146	1.7925	10	8.1
	145	1.0261	5	4.5
	110[c]	6.5987	30	27.7
	111[c]	2.1400	10	9.5
	109[c]	3.8805	20	17.4
435205[b]	136	1.2851	5, 20	5.3
	143	3.4122	10, 50	10.3
	147	0.8311	3, 15	3.2
	105[c]	3.3760	10, 50	13.6
	104[c]	6.5906	25, 100	25.5
	102[c]	2.2182	10, 40	9.0
435244	137	0.8305	10	3.0
	135	1.7925	25	18.6
	138	3.0681	45	9.8
	047[c]	1.9505	30	12.6
	107[c]	4.4767	65	28.8
	061[c]	6.8578	100	43.3
435248	139	0.4699	5	5.6
	149	2.9828	40	11.8
	150	1.5373	20	19.8
	112[c]	2.1776	30	13.4
	---[c]	7.1486	100	44.2
	103[c]	4.1215	50	25.1

[a]Estimates based on RD assay of complete containers, assuming uniformity of contents.

[b]Two separate CMB-8 estimates were provided.

[c]J. L. Parker assayed these samples by detection of 186-keV gamma rays. The results are in Part I, Sec. VI-D of this report.

H. Comparison of Gamma and Neutron NDA Techniques for 30-gal Waste Barrels (T. W. Crane, J. L. Parker, and C. A. Spirio)

We were asked by J. A. Kircher of CMB-8 to assay a group of 30-gal barrels containing highly enriched uranium intermixed with furnace parts and reduction residues. Only a rough estimate of the uranium content was available, and a measurement was necessary to close the record on material balances.

The barrels were assayed by two techniques. First, the SGS was used for passive counting of 185-keV gamma rays from ^{235}U. Second, a californium Shuffler was used for active neutron interrogation and delayed-neutron counting (Ref. 3, p. 20). Both techniques used a single standard containing 46.4 g of ^{235}U in a 30-gal barrel intermixed with paper to approximate the waste barrels. The SGS used a

TABLE X

ASSAY VALUES OF ^{235}U IN PUMPS AND FILTERS FROM GROUP AP-3

Pump No.	^{235}U (mg) ±3%
1	127.3
2	107.9
3	126.8
4	35.4
5	129.7
6	46.9
7	379.6
Filters	156.9

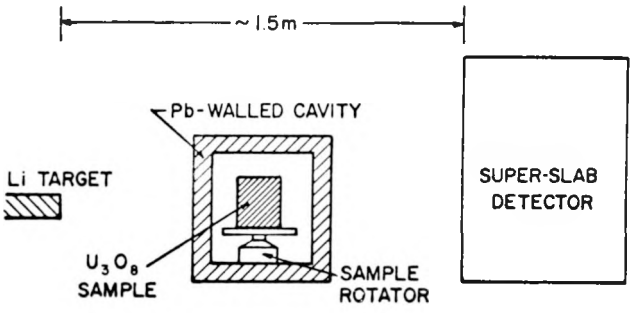


Fig. 46.
Experimental arrangement for Van de Graaff assay of large U_3O_8 samples (not to scale).

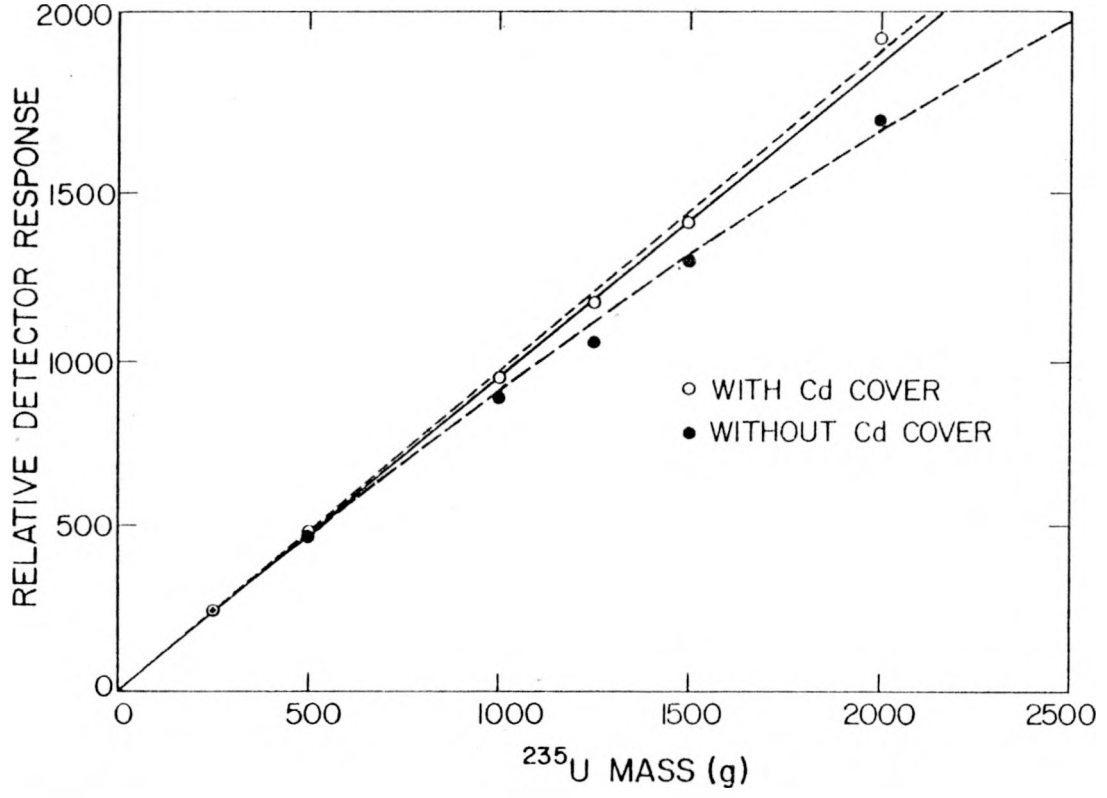


Fig. 47.
Relative detector response vs ^{235}U mass for the Van de Graaff assay of U_3O_8 standards. The upper curve is a least squares fit to data acquired with cadmium-covered standards; for the lower curve, the standards had no cadmium covers. The assumed response function is $R = Am/(1 + Bm)$. The straight line represents $R = Am$ for the case where standards had cadmium covers.

gamma-ray transmission measurement to correct for the matrix differences between the standard and the samples; however, no matrix corrections were made for the Shuffler unit.

Table XI shows the results of the two assay techniques. The estimated overall accuracy of these measurements is ±20% (1-σ level). For the 10 barrels assayed by both the SGS and Shuffler, the total inventory was 177.6 g and 165.9 g, respectively, a reasonable agreement for this type of material. Two barrels were not assayed by the Shuffler because they were too heavy to transport to the Shuffler.

The higher count rate of the Shuffler gave it better precision than that of the SGS. For the standard (30-gal barrel), the Shuffler count rate was 7.4 counts/s/g ^{235}U, whereas the SGS count rate was 1.8 counts/s/g ^{235}U. However, because the waste barrels were more dense than the standard, the SGS count rate was reduced by about an order of magnitude.

I. Calibration of a Commercially Produced MEGAS Box Counter (T. W. Crane)

A copy of the MEGAS box counter[20,21] was purchased by HEDL from Jomar Systems. HEDL delivered the instrument to LASL, where it was calibrated using waste standards prepared by CMB-11. The calibration constants determined for MEGAS were applicable to the HEDL unit. During the calibration, representatives of the Nuclear Materials and Equipment Company (NUMEC) were trained in the operation of the box counter and shown the MEGAS box counter at DP Site. NUMEC will use the HEDL box counter to sort combustible waste at their plutonium facility in Leechburg, Pennsylvania. Boxes above the 10-nCi/g fiducial will be sent to HEDL for disposal or recovery, while boxes below the 10-nCi/g fiducial will be disposed of at NUMEC.

TABLE XI

^{235}U CONTENT OF TWELVE 30-GAL BARRELS MEASURED BY THE SHUFFLER AND THE SSAS

Drum No.	Contents	Net Weight (kg)	Estimated ^{235}U (g)	Measured ^{235}U (g)[a]	
				SGS	Shuffler
70	Reduction residues	92	65	59.1 ± 2.2	---
79	Furnace parts	31	9	53.8 ± 1.4	37.50 ± 0.10
80	Furnace parts	23	6	1.1 ± 1.0	1.239 ± 0.016
81	Furnace parts	56	16	4.8 ± 1.4	3.699 ± 0.030
82	Furnace parts	47	12	2.1 ± 1.0	2.314 ± 0.026
83	Reduction residues	126	61	55.9 ± 1.4	---
84	Reduction residues	69	36	14.8 ± 0.8	19.72 ± 0.06
85	Reduction residues	68	35	19.2 ± 1.0	18.89 ± 0.06
86	Reduction residues	68	35	20.8 ± 0.9	24.84 ± 0.08
87	Reduction residues	69	30	21.6 ± 1.0	21.67 ± 0.07
88	Reduction residues	69	28	21.3 ± 0.9	20.46 ± 0.07
89	Reduction residues	39	33	18.8 ± 1.0	15.54 ± 0.06

[a]Errors indicate statistical precision only (1-σ level).

J. INMM Committee 9.5 Projects (T. W. Crane and G. W. Eccleston)

The Institute of Nuclear Materials Management (INMM) supports committees that prepare standards through the American National Standards Institute (ANSI) for the nuclear industry. INMM Committee 9.5 prepares written standards for NDA techniques. To assist the committee in preparing a guide, "Non-Destructive Assay of Low Enrichment Uranium Feed Material," we contributed descriptions of available NDA techniques with their limitations because several of the instruments were initially developed at LASL. INMM Committee 9.5 also is about to accept as an ANSI standard (ANSI-N15.23) a guide based primarily on the LASL-developed fuel rod scanner.[22]

VII. TRAINING AND TECHNOLOGY TRANSFER

A. Safeguards Technology Training Program (T. R. Canada, J. W. Tape, and staff)

Two courses, "Fundamentals of Nondestructive Assay of Fissionable Material Using Portable Instrumentation" and "In-Plant Nondestructive Assay Instrumentation," were conducted at LASL during the weeks of October 17 and December 5, 1977. These sessions marked the fifth and second offerings, respectively, of the two courses. Lists of the 33 domestic and 28 foreign attendees and their organizational affiliations are shown in Tables XII and XIII. Demand for those courses continues to be high.

Over the years, the "Fundamentals" curriculum has been modified to meet the current needs of the domestic and international safeguards community while continuing to emphasize basic concepts and the use of portable, inspector-oriented instrumentation. This year we introduced a laboratory session on the calibration and use of the HLNCC that LASL is providing to the IAEA. Traditional neutron assay principles using the SNAP detector and the SAM were also taught. Exercises on gamma-ray assay techniques using the SAM and the NaI(Tℓ) detectors covered plutonium-ash measurements and the enrichment principle. CdTe detectors were demonstrated as an example of a potential gamma-ray detector for portable applications. Lectures and demonstrations during the week included an introduction to passive NDA techniques, an oveview of the DOE Safeguards Technology Training Program, and a demonstration of the SGS that is to be supplied to the IAEA.

The instruments presented during the "In-Plant" course are typical of the NDA tools that are, or will be, found in nuclear facilities. They are generally computer-based and capable of routine operations with high assay accuracy. The following instruments (and the basis for their assay) were studied during the December course: the SGS (transmission-corrected passive gamma counting), the USAS (transmission-corrected gamma-ray counting and densitometry), the RD (active-neutron interrogation with fast coincidence counting of induced-fission neutrons), and neutron well coincidence counters (passive coincidence counting of spontaneous-fission neutrons). Attendees spend a full day with each instrument, calibrating and operating it as well as discussing assay principles.

In addition to the laboratory sessions, lectures were presented on "^{252}Cf Shufflers and Fuel Rod Scanners," "In-Plant Holdup Measurements," "Absorption Edge Densitometry," "Applications of In-Plant Instrumentation to a High-Throughput Mixed-Oxide Fuel Fabrication Facility," and "Implementation of Dynamic Materials Control."

B. Visits and Technical Data Transmittals (Q-1 Staff)

Table XIV is a summary of Group Q-1 interactions with other organizations in which substantive technical data transfers or planning transpired. The visits include both those at LASL and travels by Q-1 representatives to host facilities. Group Q-1's responses to written requests for design data and reports are included. Many interactions were part of coordinated efforts of all LASL safeguards groups to respond to the organizations listed.

TABLE XII

FUNDAMENTALS OF NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS USING PORTABLE INSTRUMENTATION: COURSE ATTENDEES
October 17-21, 1977

Organization	Representative
British Nuclear Fuels Ltd.	Arthur N. Spencer
Brookhaven National Laboratory	Alan M. Bieber, Jr.
IAEA	W. T. Battrick Clarence Breskovic Anatoly Filatkin A. E. Matolczy G. Rabot Edward G. Selleck Valentin Slizov D. Strelrov Masayori Tsutsumi William I. Winters Elke Wolfl Lars Wredberg
Japan AER Institute	Keisuke Kaieda
Lawrence Livermore Laboratory	Stephen Chin
LASL	John Gonzales Matthew N. Hykel
National Lead Co. of Ohio	Allen R. Diehl
Science and Technology Agency, Japan	Takeshi Someya
Texas Instruments	Fred Sherman
Union Carbide Corp., N.D.	Robert W. Jackson John W. Ruthven
Union Carbide Corp., ORNL	Charles W. Benson Donald R. Stallions
United Nuclear Corp.	Thomas Ashley
USDOE	James Fowke James K. Reilly
USNRC	Paul H. Bissett Ira Cohen Daniel Holody G. Dan Smith Robert J. Summers

TABLE XIII

IN-PLANT NONDESTRUCTIVE ASSAY INSTRUMENTATION: COURSE ATTENDEES
December 5-9, 1977

Organization	Representative
Argonne National Laboratory	Alan E. Proctor Nancy Trahey
USDOE, New Brunswick Lab.	William Ulbricht
DuPont, Savannah River Plant	Lou M. Papouchado P. N. Sand William C. Scotten
Goodyear Atomic Corp.	Atsushi Sugiura
IAEA	Djali Ahimsa Elmir Dermendjiev Ricardo Diaz-Duque Roine Ekarv Herminio Gonzalez-Montes Miroslav Krivanek Leo Oudejans Robert Schaer Svein Thorstensen Emanuel Yellin Mohamed Yousiv
Japan AER Institute	Keisuke Kaieda
Oak Ridge National Laboratory	Alan M. Krichinsky Whitney H. Tipton
Rockwell Hanford Operations	George Westsik
Science and Technology Agency, Japan	Takeshi Someya
Westinghouse Nuclear Fuel Div.	Hassan J. Ahmed
United Nuclear Corp.	Barry Snitzer
USNRC	Frank P. Gillespie James Patterson G. Dan Smith

TABLE XIV

VISITS AND TECHNICAL DATA TRANSMITTALS

Date (1977)	Organization	Subject
9-15	Goodyear Atomic Corp.	Drawings—enrichment monitor.
10-4	HEDL	Engineering evaluation of commercial MEGAS combustible waste assay unit for HEDL.
10-4	Canberra Industries	Circuit diagrams of the high count rate shift register coincidence unit.
10-17	Argonne National Lab.	Information on techniques and equipment for gamma scanning of fast reactor fuels.
10-19	Argonne National Lab./West	Drawings—HEDL-FFTE rod scanner.
10-19	General Atomic	Drawings—RD.
11-18	IRT, Inc.	Design drawings and specifications for RD.
11-18	Westinghouse	Design drawings of dual range coincidence detector, glovebox neutron well counter, the SNAP detector, and excerpts from progress reports describing these instruments.
11-22	National Nuclear Corp.	Design information on the RD.
11-22	Consumer Power	Information on the gamma scanning of spent fuel assemblies.
12-8	HEDL	Blueprints of HP-97 interface to coincidence counter.
12-9	IRT, Inc.	Shuffler design and performance.
12-9	National Nuclear Corp.	Shuffler design and performance.
12-14	National Nuclear Corp.	Design drawings, specifications, and software listings for RD.
Visitors to LASL, Domestic		
9-15	HEDL	HPFL pin scanner.
11-8	Sandia Labs.	NDS instrumentation for assay of advanced fuels.

TABLE XIV (cont)

Date (1977)	Organization	Subject
11-10	DOE/SR SRL-DuPont	LASL alternate fuel cycle.
11-15/11-16	Westinghouse	LWR waste assay systems.
11-16/11-18	DOE/SS	Program planning.
12-7/12-8	NBS	Safeguards standards.
12-7	ISPO/BNL	IAEA tasks.
12-8	Argonne National Lab.	NDA instrumentation.
Visitors to LASL, Foreign		
9-16	Representative from Norway and Denmark	Safeguards bilateral discussions.
10-5	Representatives from Japan	Safeguards bilateral discussions.
10-12	UKAEA-Harwell	Plutonium isotope determination by gamma-ray spectroscopy/NDA problems.
10-21	Representatives from Greece	Safeguards bilateral discussions.
10-25	Harwell	NDA techniques.
10-25/12-3	[Japan]	NDA techniques.
10-28	Representatives from Belgium	Safeguards bilateral discussions.
11-14/11-18	IAEA	IAEA neutron coincidence counters.
11-16	Representatives from Japan	Safeguards for Tokai reprocessing facilities.
11-22	Representatives from South Korea	Safeguards bilateral discussions.
12-5/12-6	IAEA	Neutron collar.
12-12/12-13	IAEA	Spent fuel assay.

TABLE XIV (cont)

Group Q-1 Travel

Date (1977)	Name	Destination	Subject
9-19	H. Menlove D. Close G. Eccleston	Allied Chemical, Idaho Falls	ICPP fluorinel sludge assay system.
9-26	C. Hatcher	Seattle, Wash.	ISPO program coordination.
9-28	S. Hsue	Mol, Belgium Vienna, Austria	IAEA.
10-3	T. Crane J. Tape	Gatlinburg, Tenn.	Paper presented at Analytical Chemistry in Energy Technology conference.
10-4	R. Walton	DOE & SRP, Savannah River	Safeguards test and evaluation projects.
11-1	T. Canada J. Tape L. Cowder	SRP	LASL/SRL densitometry project.
11-2	S. Hsue J. Phillips	Vallecitos, Cal. CA/GE	NDA—irradiated fuels.
11-14	K. Johnson	New Orleans	NDA Standards Committee.
11-16	G. Eccleston H. Menlove	Los Angeles	Review of draft of design criteria for neutron interrogator.
11-28	N. Ensslin S. Hsue	ANS, San Francisco	Safeguards papers presented at meeting.
11-28	S. Bourret	San Diego	Paper presented at Fall DECUS Symposium.

PART 2

DETECTION, SURVEILLANCE, VERIFICATION, AND RECOVERY

GROUP Q-2

Carl Henry, Group Leader

E. J. Dowdy, Alternate Group Leader

The activities of Group Q-2 are directed toward the development of compact and highly sensitive instruments for the detection and surveillance of SNM. The group participates actively in the Nuclear Emergency Search Team (NEST), DOE's program to provide immediate response to nuclear emergencies involving accidents, lost or clandestine materials, and terrorist threats. Q-2 provides personnel, techniques, and procedures for NEST exercises, and design data for new and/or improved portable and mobile nuclear search systems in support of the NEST program. Because much of the effort in this area is classified or sensitive, the work is reported separately in classified progress and topical reports.

Group Q-2 also applies nuclear detection and surveillance techniques to the problem of perimeter safeguards at domestic and international nuclear facilities. This effort includes the design of handheld personnel and vehicle monitors for searches at exits from SNM access areas, and the continued development, testing and evaluation of SNM portal monitors and associated test methods and standards to evaluate performance of portal installations.

A substantial program is also under way to develop techniques for automatic and continuous monitoring of SNM in storage vaults. For long-term storage of significant quantities of SNM, such techniques may provide assurance in real time that no diversion has occurred and reduce the frequency or eliminate the need for periodic physical inventories.

I. PERIMETER SAFEGUARDS

A. Personnel and Vehicle Monitors

1. Delta Rate Personnel-Vehicle Monitor (PVM) (D. R. Millegan)

Specifications are complete for the delta rate logic for the PVM and requests for quotations on fabrication costs have been made. The new model PVM incorporating this logic will be identified as HSS-1050 MOD IV.

2. Protective Force Training (W. E. Kunz)

The DOE Protective Force supervisory personnel were given a lecture series on hand-held SNM monitor operation. The effects of shielding, source-to-detector distance, exponential attenuation, background radiation, and the statistical variation of count rates were discussed and demonstrated with a conventional NaI(Tℓ) detector system with an audible-count system and ^{235}U and ^{239}Pu sources.

B. Portal Monitors

1. DOE Doorway Monitors (P. E. Fehlau and J. M. Bieri)

A standard operating procedure for periodic performance testing of doorway monitors was written and sent to CMB-11 as a suggested routine for use at the present TA-21 portal monitor installation and future TA-55 portal monitor installations.

Maintenance of the TA-21 portal monitor was limited to replacement of the amplifier module, which had developed an intermittent open circuit. The other service calls apparently were a result of short-term background variations.

The IRT PRM-110 doorway monitor, which we will be installing in TA-55, has been operating for 3 months in a laboratory environment at TA-18. There have been intermittent problems with the high-voltage supply and replacement parts have been difficult to obtain.

The final positions for the doorway monitors at TA-55 were established and appropriate fixtures for installation were designed and fabricated.

Two monitoring problems were examined. One involved the use of portal monitors at accesses to the TA-18 Kivas which house the critical assemblies. The other involved a proposal from Albuquerque Operations Office for a standard for SNM instrumentation.

2. Nuclear Regulatory Commission (NRC) Portal Monitors (P. E. Fehlau, T. E. Sampson, and C. N. Henry)

We completed the Task 5 report which discusses procedures that NRC inspectors might use to monitor portal monitor operation at license installations.[23]

Our conclusion on substitute sources for doorway-monitor evaluation was that exact substitutes could not be found and that because the ^{239}Pu 1-g standard has a higher gamma activity than the ^{235}U 10-g standard, the ^{235}U could be used alone. This conclusion was incorporated into the draft revision of Regulatory Guide 5.27 that we had previously sent NRC, and the use of a standard ^{235}U source will be assumed in the remainder of the project.

Preliminary work for Task 6, which is a trial run of the inspection techniques proposed, is being completed and the trial will be completed during the next reporting period.

C. Realtime Inventory (N. Nicholson, C. D. Ethridge,* and T. H. Kuckertz*)

1. Shelf Monitors

The Mod-4 shelf monitor printed-circuit-board hardware (design drawing #4Y-168742) has been checked out. The software for the single-component microcomputer, Intel 8748, is being debugged. Intel is developing an in-circuit-emulator (ICE-48) to aid in debugging 8748 software programs, but it is unavailable at present and the software checkout is being done with conventional test equipment.

The Mod-5 design hardware is similar to the Mod-4 design except that microcomputer software now replaces the relatively expensive ($6) universal asychronous transmitter/receiver in the Mod-4 to accomplish the transmit/receive function. The cost becomes significant when some 5000 shelf monitors are considered for a large storage vault. The Mod-5 hardware has been fabricated and the software has been written and assembled. Checkout of the Mod-5 design will begin upon completion of the Mod-4 software.

2. Proposed TA-55 Shelf-Monitoring System

There has been discussion about the installation of a demonstration shelf system in the TA-55 storage vault, where the realtime inventory system can be tested under realistic conditions and can be more effectively evaluated. The existing shelves will not need modification, but some redesign of the shelf monitors may be necessary because there may be as many as four containers per location.

The DYMAC system will include a teletype and a line printer outside the main vault door, where material entering and leaving the vault will be monitored, thus making the portal security system unnecessary.

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*Group E-5.

3. Sandia/Rockwell Phase II Verification Station

The Phase II Verification Station for the Sandia/Rockwell (formerly ARHCO) demonstration will be developed during the coming months. The instrumentation will include a balance capable of >1-g accuracy, reduced variance equipment capable of measuring the quantity of ^{240}Pu present in the sample, and possibly a gamma detector designed to detect gammas emitted by ^{239}Pu.

4. Computer System

The Data General NOVA 3/12 is nearly operational. The ORTEC CAMAC crate controller, which was incompatible with the NOVA 3/12, has been replaced with a BiRa crate controller. We are integrating various CAMAC modules into the system.

A panel containing status lights has been interfaced to the computer through two CAMAC modules—a parallel input gate and an output register driver. The lights indicate the condition of doors on the verification chamber and will inform the user whether material can be entered or removed. This status panel also indicates errors in the logic flow or sequence for entering or withdrawing material.

We are developing software that will allow the computer to communicate with a number of CAMAC modules, which are tied to detectors, instruments, and devices. The software is available upon request.

D. Reduced Variance Measurements of Pu Samples (E. J. Dowdy)

The reduced variance technique of detecting time correlations in the pulses coming from neutron detectors in the presence of plutonium (Ref. 3, pp. 53-55) has been extended to a wider range of samples, including metals, oxides, oxides with additives, and a carbide. The samples were placed in a typical well counter used for sample assay. The pulses from the amplifiers/discriminators were processed in parallel by a neutron well coincidence counter and the reduced variance logic modules. Simultaneous measurements of the coincidence counts R, the totals count T, and the variance-to-mean ratio 1 + Y, were made on the 18 samples described in Table XV. The data plotted in Fig. 48 show a correlation between the variance-to-mean ratio and the ratio of coincidence counts to total counts, implying that equivalent information is obtained in both data analysis techniques.

Another correlation has been made between the data obtained with the reduced variance technique and the coincidence counting technique. The correlation is between $\dot{R}$, the real coincidence count rate from the coincidence counter, and $\overline{C^2} - \overline{C}^2 - \overline{C}$, an expression involving the moments of the count distribution which are obtained by using the reduced variance logic module. Measurement data for the samples listed in Table XV were used to make this correlation. Figure 49 is a plot of the data for some nonmetallic samples, and Fig. 50 is a similar plot for the metallic samples. Attempts to prove a single correlation for all the samples (metals and nonmetals) have been unsuccessful.

E. HgI_2 Detector Characterization (E. J. Dowdy)

We have considered using HgI_2 detectors in place of NaI(Tℓ) in certain safeguards applications. These detectors would allow a reduction in weight and volume for instruments such as the PVM.

We received from EG&G/Santa Barbara one HgI_2 "high-energy" detector (with integral preamplifier) which is said to have a measurable efficiency for full-range detection of gamma rays of energy as high as ~1 MeV. The detector is 570 μm thick, with a ~1-cm^2 area. It operates at a stated maximum bias of +1500 V. A Tennelec TC 203BLR amplifier with 2-μs shaping constant was satisfactory for the typical count rate of $\sim 8 \times 10^3$ cps used in this study. We investigated the effect of gamma-ray energy on detector efficiency and resolution. The results are shown in Figs. 51 and 52. When we checked the count rate dependence of the resolution for the ^{57}Co 122-keV line, we found a 30% degradation in resolution as the count rate increased from 1×10^3 cps to 2.5×10^4 cps. Because of their exhibited low efficiency compared with NaI detectors, the available HgI_2 detectors seem unsuitable for our applications.

TABLE XV

NEUTRON CORRELATION STUDY DATA

Sample No.	Sample Description	Coincidence (counts)[a]	Totals (counts)	Coincidence / Totals	1 + Y
STD-29424	Carbide	38455 ± 240	535283	0.0718 ± 0.0004	1.183 ± 0.002
STD #9	PuO_2	327499 ± 2600	4993102	0.0656 ± 0.0005	1.142 ± 0.002
STD #8	PuO_2 + MgO	143363 ± 2100	4042886	0.0355 ± 0.0005	1.072 ± 0.002
STD #7	PuO_2 + MgO	66428 ± 1500	2992977	0.0222 ± 0.0005	1.047 ± 0.002
STD #6	PuO_2 + DE[b]	57857 ± 800	1495895	0.0387 ± 0.005	1.095 ± 0.002
STD #5	PuO_2 + MgO	30173 ± 1000	1947560	0.0155 ± 0.0005	1.031 ± 0.002
STD #4	PuO_2 ; DE	27374 ± 460	847166	0.0323 ± 0.0005	1.076 ± 0.002
STD #3	PuO_2 + DE	34097 ± 550	1020818	0.0334 ± 0.0005	1.081 ± 0.002
705-003	PuO_2 + DE	8358 ± 180	300203	0.0278 ± 0.0006	1.070 ± 0.002
PEO 385	PuO_2	372496 ± 2800	5437964	0.0685 ± 0.0005	1.147 ± 0.002
PEO 447	PuO_2	750383 ± 5100	9833772	0.0763 ± 0.0005	1.146 ± 0.002
PEO 382	PuO_2	478746 ± 3600	6932052	0.0691 ± 0.0005	1.143 ± 0.002
643-01	Pu metal	3309560 ± 5700	10647524	0.3108 ± 0.0005	1.668 ± 0.002
643-003	Pu metal	2683290 ± 6100	11541739	0.2325 ± 0.0005	1.479 ± 0.002
643-004	Pu metal	512951 ± 2300	4221034	0.1215 ± 0.0005	1.283 ± 0.002
643-005	Pu metal	1299671 ± 5200	10021881	0.1297 ± 0.0005	1.262 ± 0.002
620-000	Pu metal	8069407 ± 8600	16216078	0.4976 ± 0.0005	2.055 ± 0.002
621-000	Pu metal	1991628 ± 3600	6131014	0.3003 ± 0.0005	1.692 ± 0.002

[a]Determined using inner ring of the well counter, 64-μs coincidence gate; corrected for deadtime effects.

[b]Diatomaceous earth.

F. Irradiated Fuel Monitors (E. J. Dowdy, J. T. Caldwell, S. W. France, W. E. Kunz, C. E. Moss, N. Nicholson, J. C. Pratt, A. A. Robba, and T. H. Whittlesey)

Eight nuclear radiation detection systems were studied to determine the suitability of each for surveillance of spent LWR fuel assemblies in storage ponds. The study was made for the IAEA under the International Safeguards Project Office (ISPO) program, and a report is being written. The eight systems were: Cerenkov radiation detectors, scintillation systems, neutron and gamma-ray ion chamber detectors, thermoluminescent detectors, solid state (CdTe and HgI_2) detectors, time domain reflectometers, monoenergetic neutron systems, and thermal radiation detectors. Of those, the Cerenkov detection systems, scintillators, and thermoluminescent detectors were selected for further study.

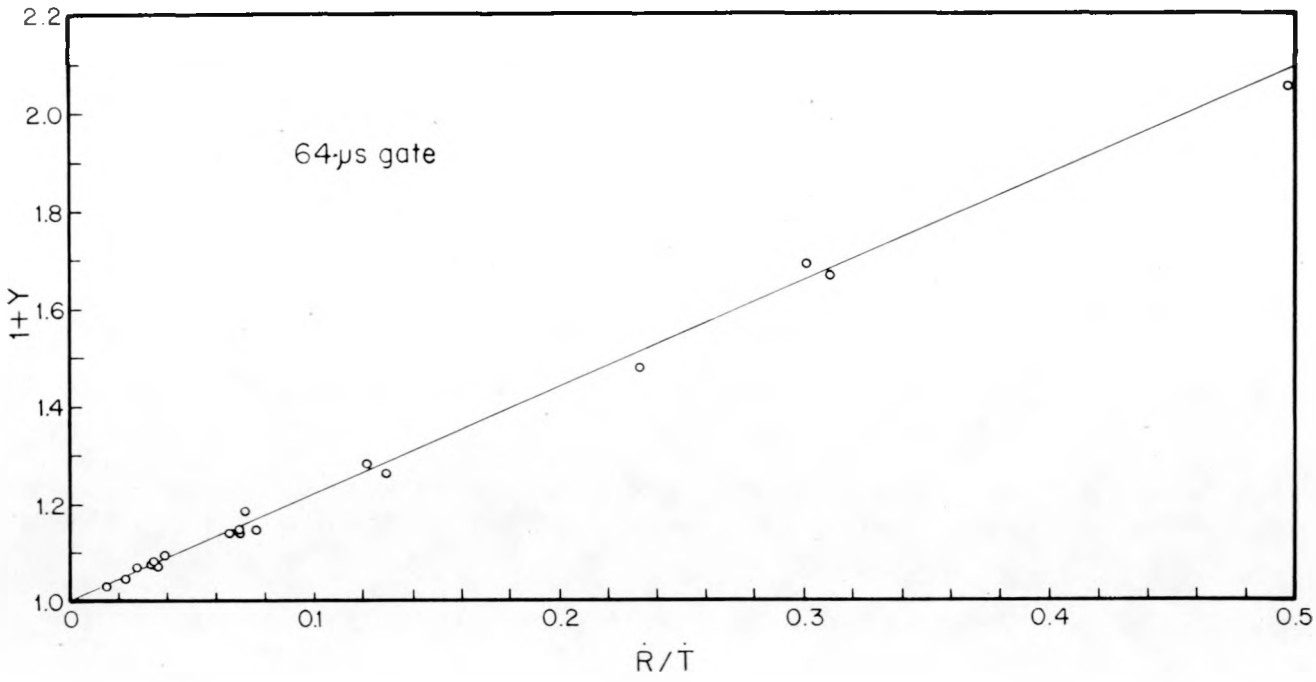

Fig. 48.
Results of measurements made on plutonium samples (Table I), using the reduced variance logic module and a neutron coincidence counter, showing the correlation between 1 + Y and $\dot{R}/\dot{T}$.

G. Reactor Power Monitor (A. A. Robba, E. J. Dowdy, G. M. Worth, and R. D. Hastings)

The IAEA desires a continuous independent record of the operating power of all nuclear reactors under surveillance. We have proposed a monitor that uses a thermal neutron detector for a sensor and a microprocessor-based data acquisition and analysis system. It provides an hourly record (in memory and on magnetic tape) of the Julian data, time, and operating power (in per cent of rated capability). The system operates from rechargeable batteries which are on trickle charge from host power. The hardware is nearly completed and the software is being written.

II. SUPPORTING RESEARCH

LASL-LLL Photofission Data (J. T. Caldwell, H. F. Atwater, E. J. Dowdy)

Photofission data for ^{236}U, ^{238}U, and ^{232}Th were reanalyzed using calculated multiplication values. Resulting changes in the centroid and width of the multiplicity distribution ($\bar{\nu}$,σ) and cross sections for the three isotopes were small. (^{235}U data were reanalyzed earlier.)

Comprehensive tabulation of data for ^{235}U, ^{236}U, ^{238}U, and ^{232}Th is under way, and LASL and LLL personnel are collaborating on a paper to be submitted to Nuclear Science and Engineering.

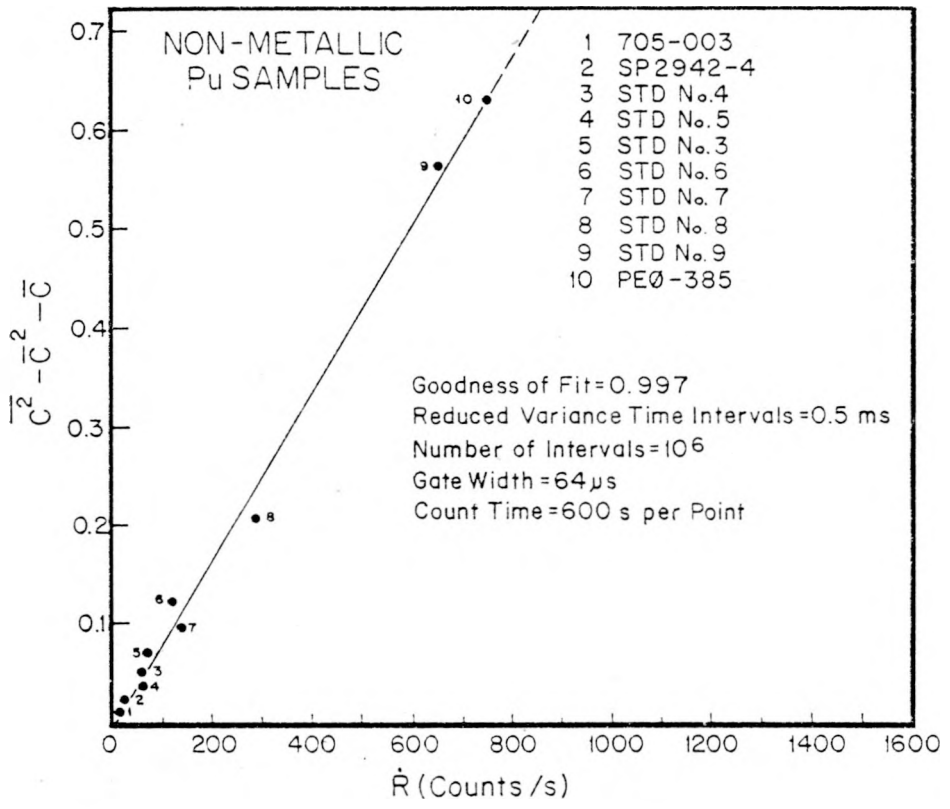

Fig. 49.

Results of measurements made on some nonmetallic samples (Table I), showing correlation between coincidence rate R from the coincidence counter and an expression involving moments of the count distribution from the reduced variance logic module.

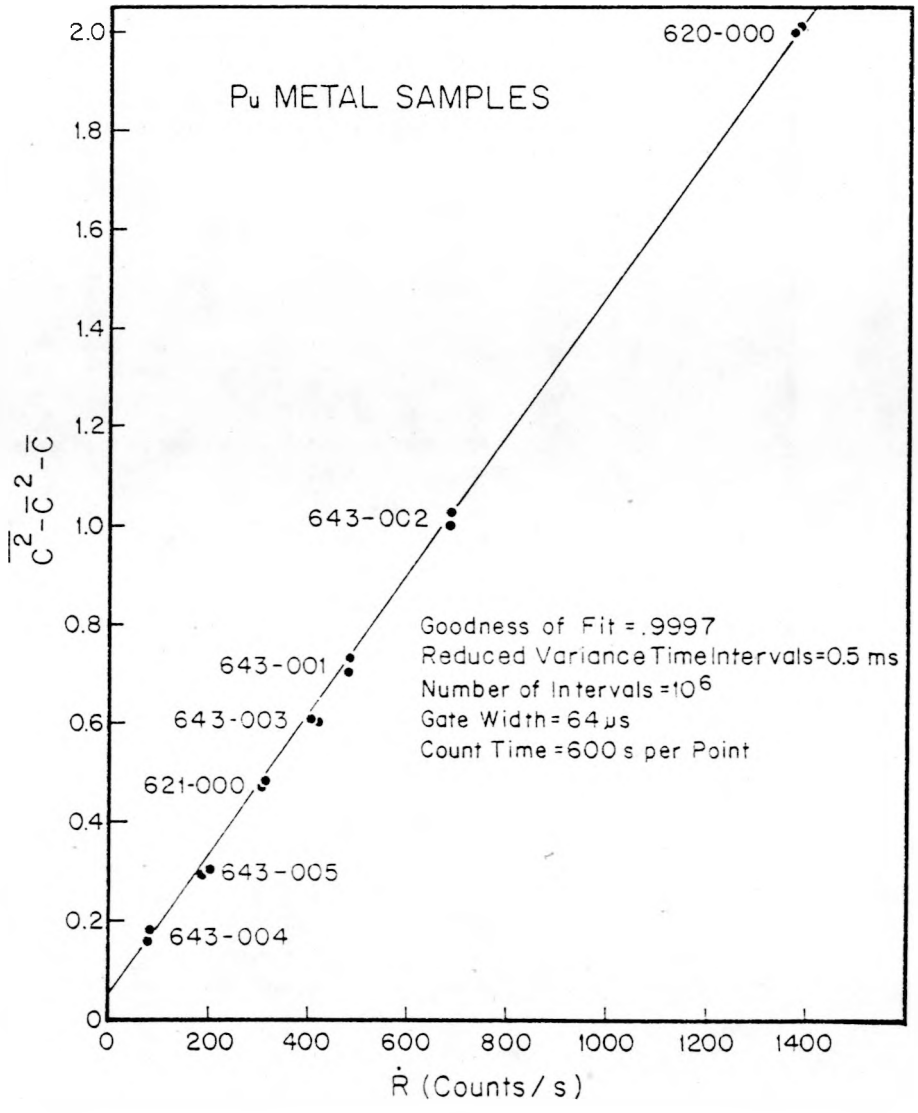

Fig. 50.
Results of measurements made on some metallic samples (Table I), showing correlation between coincidence rate $\dot{R}$ from the coincidence counter and an expression involving moments of the count distribution from the reduced variance logic module.

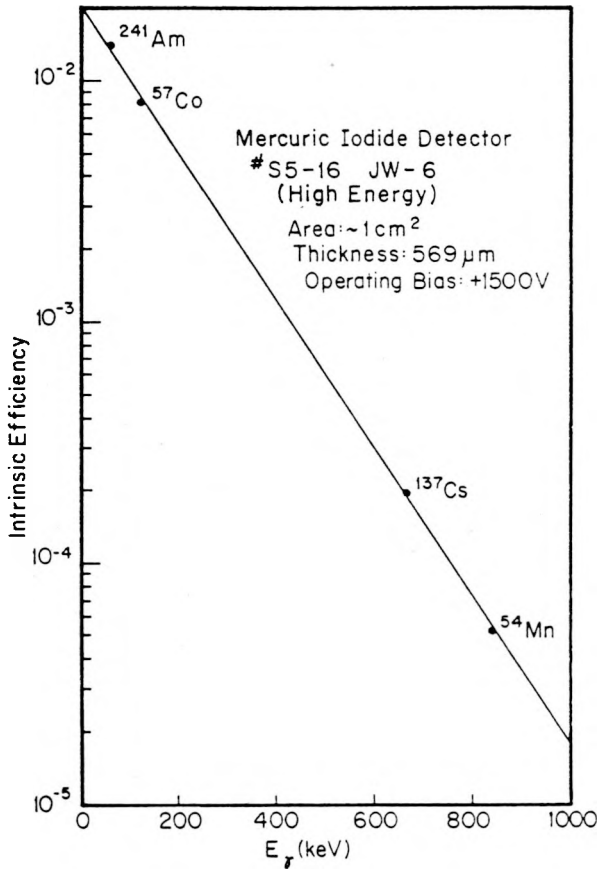

Fig. 51.
Efficiency vs energy for a HgI_2 high-energy detector.

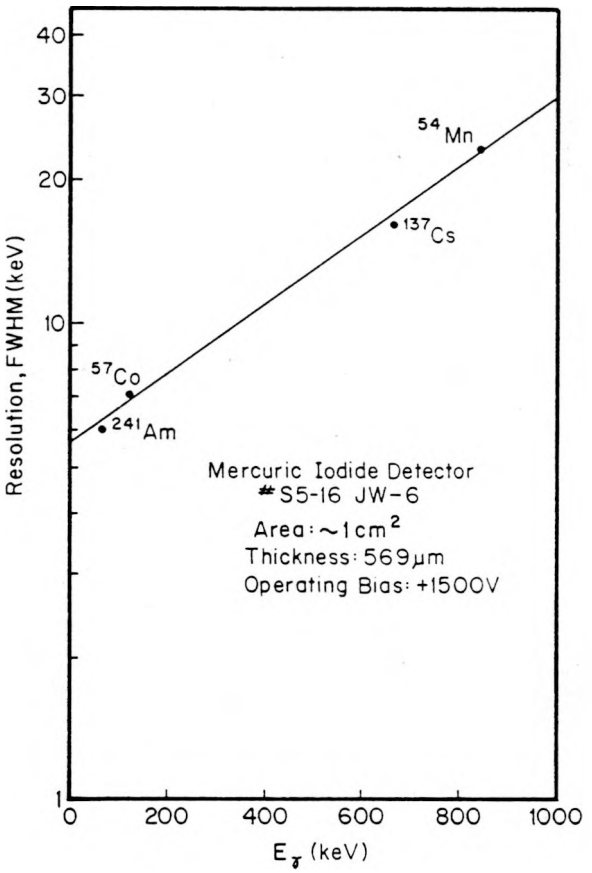

Fig. 52.
Resolution vs energy for a HgI_2 high-energy detector with integral preamplifier.

PART 3

SAFEGUARDS SUBSYSTEM DEVELOPMENT AND EVALUATION

GROUP Q-3

Ronald H. Augustson, Group Leader

Group Q-3 is responsible for developing, implementing, demonstrating, and evaluating the DYMAC program. DYMAC is an in-plant materials control system that uses a plantwide network of in-line NDA instruments coupled with automatic data-processing equipment to continuously pinpoint the quantity, form, and location of SNM throughout an operating nuclear plant.

DYMAC subsystems are: NDA instrumentation, data acquisition, data base management, and realtime accountability. The NDA instrumentation subsystem provides rapid, quantitative on-line measurement for SNM as it moves from one unit process accounting area to another. The data acquisition subsystem provides accurate and reliable material control information to the central computer. The data base management subsystem accepts and organizes incoming data into files for efficient retrieval of specific information. The realtime accountability subsystem draws on the data base for continuous status monitoring of the nuclear material within the facility. On detecting an abnormality, it signals the condition and provides inventory data to aid plant personnel in determining the appropriate response.

A DYMAC system is being integrated into the new LASL plutonium processing facility at TA-55 to serve as a working demonstration (DYMAC/TA-55). DYMAC/TA-55 goals are to demonstrate the reliability and operational feasibility of in-line NDA instrumentation in a production environment, accurate and efficient data generation, sensitivity to missing material, and compatibility with production control and quality assurance in a cost-effective manner.

The DYMAC/TA-55 program is divided into three phases. Phase I was a pilot program of research and development and in-plant testing of NDA instruments coupled with an interactive computer system at the LASL DP-Site facility. This phase has been completed.

In Phase II, currently under way, the technology and equipment developed in Phase I will be integrated into the new plutonium facility. DYMAC/TA-55 will be a full-scale, plantwide DYMAC system with NDA measurement stations in each unit process accounting area, interactive computer terminals, and realtime SNM accountability and control. In Phase III the data collected during operation of all subsystems will be evaluated in depth, focusing on effectiveness of material control, reliability, operational ease, costs, and applicability to other facilities.

Because installation of the DYMAC system at TA-55 has been slowed by slippage in the building construction schedule, plutonium did not enter the facility during this reporting period. Installation did proceed far enough in the advanced carbide fuels laboratory to allow extensive testing of the accountability system as well as to provide a basis for training the process operators. All DYMAC instruments should be installed by July 1, 1978.

I. DYMAC IMPLEMENTATION

A. DYMAC Communication System (J. Lopez*, D. A. Woodwell*, and K. A. Lindsey)

The DYMAC communication system is basically complete and operational. At present there are 28 on-line computer ports; another 36 can be connected as soon as the remote equipment is installed. Fifteen terminals are in working order in the facility. Figure 53 shows a terminal in use in the advanced carbide fuels laboratory. The caps lock key on all Teleray terminals has been modified to remain in the locked position so that the user cannot inadvertently enter a string of lower case characters, which the system does not recognize. All modified terminals have the label: CAPS LOCKED PERMANENTLY. The contractor will make a few additions and changes to

*Group E-2.

Fig. 53.
DYMAC terminal in the advanced carbide fuels laboratory.

communication lines, which DYMAC personnel will check out after installation.

The jackfield and its associated diagnostic equipment are installed in a 0.5-m- (19-in.-) wide rack located behind the DYMAC computer (see Fig. 54) and they are operational. A log has been set up for recording all port assignments and changes. It also includes a description of the jackfield and an outline

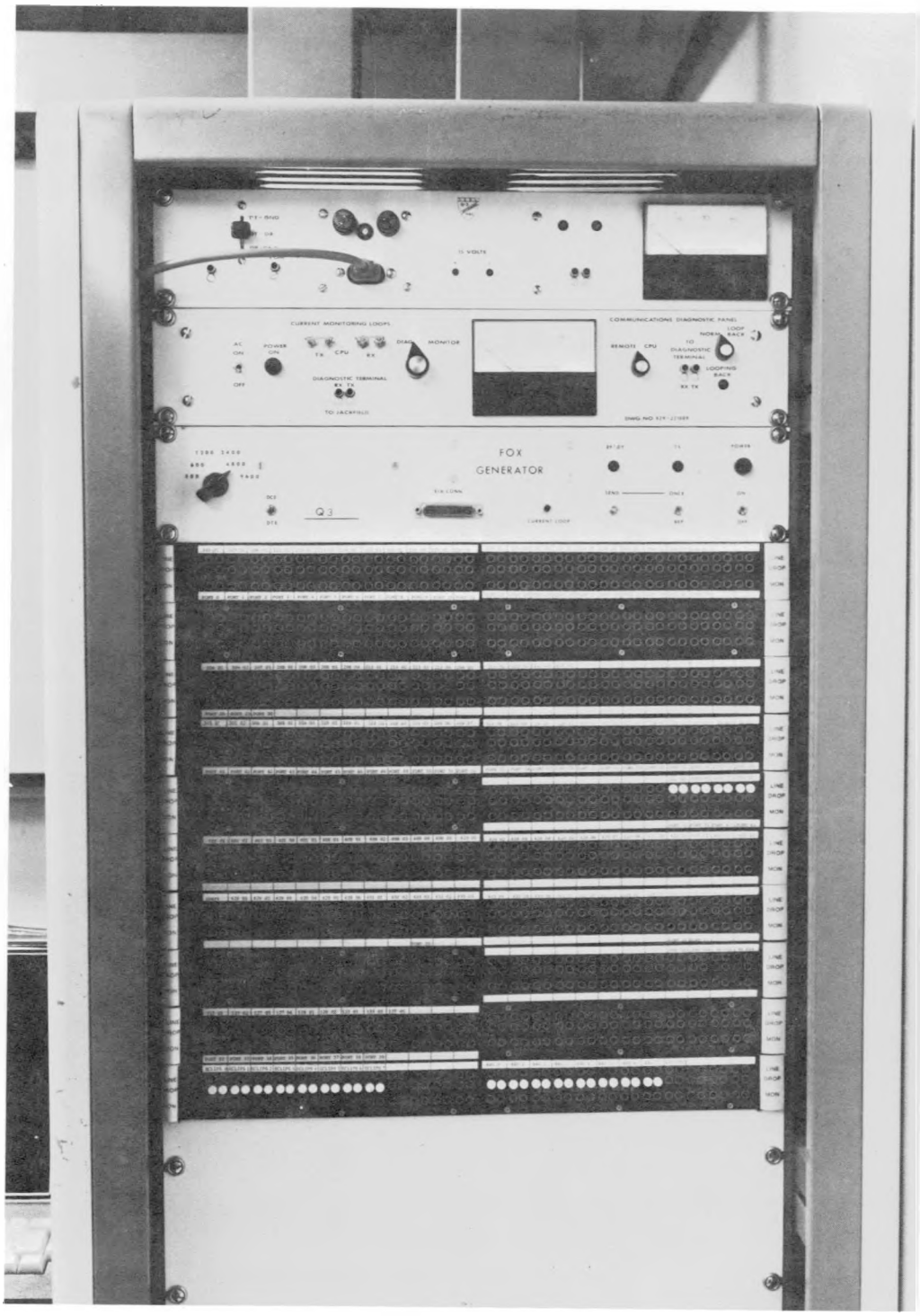

Fig. 54.
Communications patch panel and diagnostic electronics located in DYMAC computer room.

of trouble-shooting procedures. A portable diagnostic Fox generator serves as a backup to the generator on the communications rack.

The DYMAC system continues to be plagued by problems that stem from design deficiencies of the computer interface boards, which provide 20-mA current loops for in-plant data collection from NDA instruments and for material transactions from CRT terminals. These communication channels are now vulnerable to several line faults related to the plant environment. When a fault occurs, it knocks out 16 channels at a time. Data General has agreed to correct the problem.

The existing DYMAC Communication System documentation is up to data and is being reviewed. It will be distributed soon to project personnel.

B. Instrumentation (R. S. Marshall, D. G. Shirk, M. M. Stephens, V. S. Reams, N. Baron, W. R. Severe, and R. Siebelist)

1. Balances

Of twenty-one 5.5-kg-capacity electronic balances that have passed quality control inspection, seven have been installed in gloveboxes in the ^{239}Pu R&D area at TA-55 (as shown in Fig. 55). The performance of these units is being evaluated.

2. Load Cells

We suspended the development of a load-cell system for weighing tanks containing SNM solutions. A problem with the tank stirrer shaft subjected the load cell to interfering side loads. No simple mechanism exists to decouple the stirrer shaft from the tank. Q-3 personnel will evaluate an alternative system in which a dip tube transmits the solution pressure from the bottom of the tank to a pressure transducer above the tank.

3. Thermal-Neutron Coincidence Counter (TNC)

The DYMAC system will use 16 TNCs. The necessary mechanical components have arrived for nine of them. Of these nine counters, three are installed in the ^{239}Pu R&D area and two are being installed—one in the ^{239}Pu R&D area and one in the ^{238}Pu R&D area. Five TNCs await component fabrication, and two are in final design stages.

Two TNCs in the ^{239}Pu area are operational. They now use older shift-register electronics that will be replaced when more advanced shift-register units become available in February 1978. Figure 56 shows a counter installed on top of a glovebox.

4. Solution Assay Instrument (SAI)

Sample and transmission source holders, the in-glovebox components for four SAIs, have been machined and will be installed, along with the other SAI components, after construction is completed in the recycle wing.

5. On-Line Holdup Measurements

A NaI gamma detector will be installed on top of one glovebox in the ^{239}Pu R&D area to monitor the buildup of PuO_2 in the glovebox exhaust filter. That glovebox will be the one in which PuO_2 and UO_2 are blended, preparatory to making breeder reactor fuel, and is expected to be "dusty."

This monitoring system includes a 5- by 5-cm NaI crystal (with integral photomultiplier tube), preamplifier, double-delay line amplifier, gain-stabilized amplifier, three single-channel analyzers, three printing scales, a master timer, and a printing unit. All modules are contained in two NIM standard bins. The gain-stabilized amplifier and automatic recycling master timer are included to provide essentially drift-free measurements and automatic monitoring. The gain-stabilized amplifier ensures minimum drift from count-rate variations and photomultiplier-tube aging. The master timer provides automatic recycling with a variable delay interval. Thus, one the system has been calibrated and an initial measurement started, operator intervention should be minimal.

The instrument is now being calibrated and the shielding and collimated housings are being constructed.

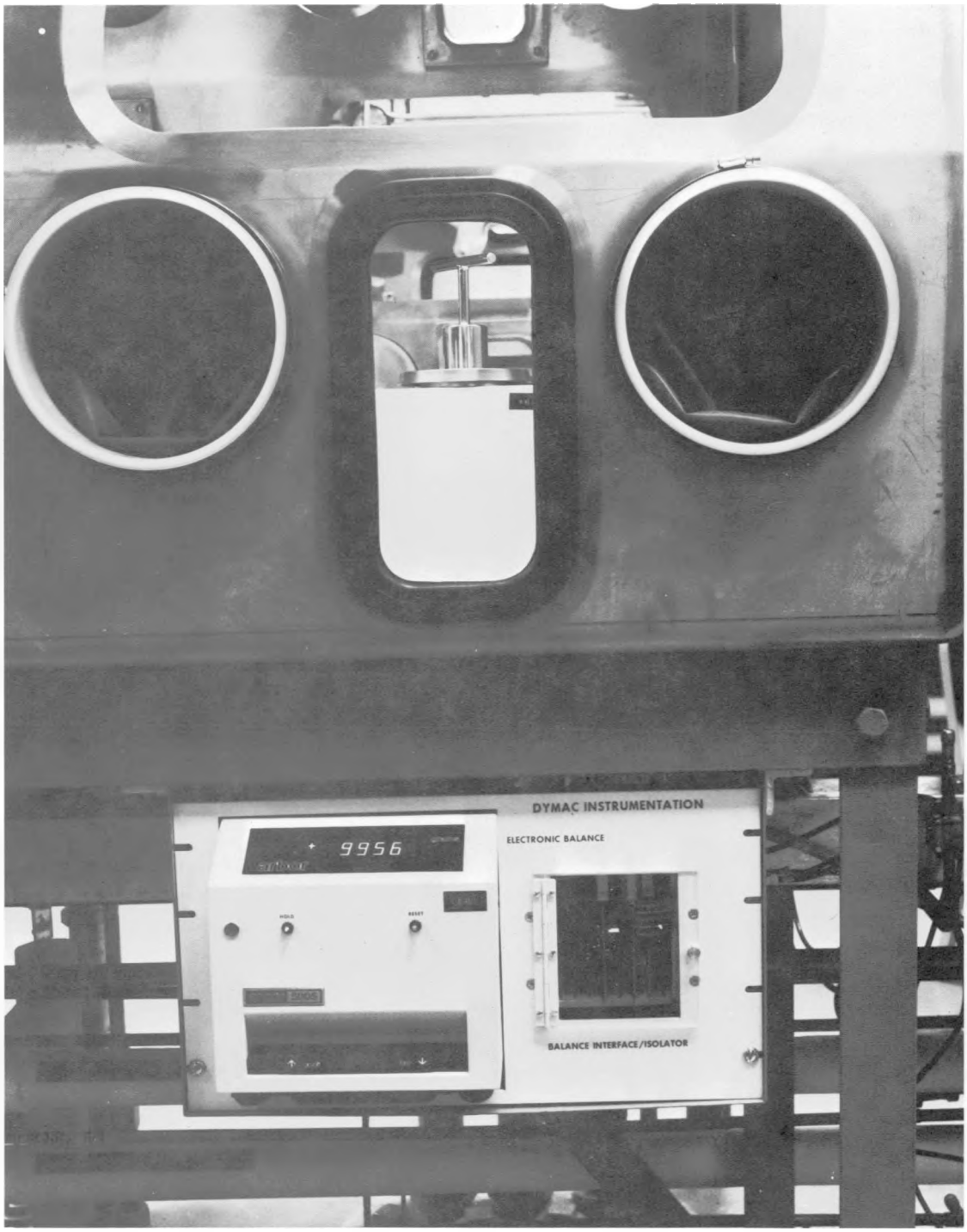

Fig. 55.
Electronic balance installed in glovebox in TA-55 as part of DYMAC system.

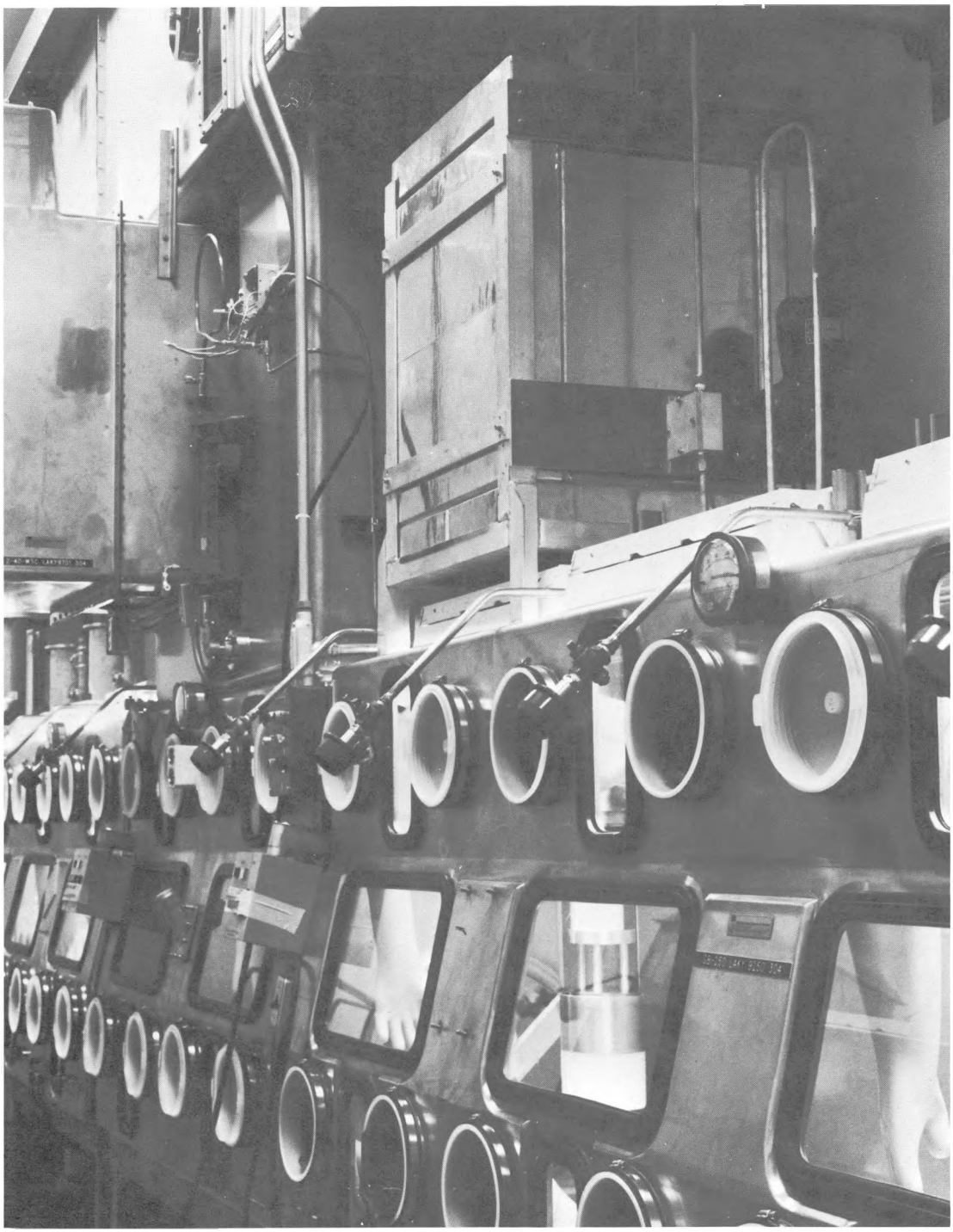

Fig. 56.
TNC installed in glovebox in TA-55 as part of DYMAC system.

C. DYMAC Software System (DYSS) (R. F. Ford*, J. Hagen, J. K. Wooten*, and T. M. Sandoski*)

The data base now contains files for the measurement control program. These files contain the history of each DYMAC instrument, including the date of the most recent precision, accuracy, and calibration checks. DYSS has been revised to increase its off-line reporting capabilities. The changes, which are specified in the Accountability Manual, accommodate requests from personnel in Group CMB-11 and the Nuclear Materials Department to provide the capability of producing special reports. In addition, the on-line reporting capability of DYSS has been expanded to include four new options, which the user selects from the option list displayed at a terminal.

ITEM STATUS enables the user to find out an item's current location and the quantity of nuclear material it contains. He need only provide the DYMAC name of the item, which consists of the account number, material type, and item identification.

LOCATION INVENTORY enables the user to obtain a list of all the inventory items present in one location and the amount of nuclear material in each item. He enters the location and the list is displayed.

ACTIVITY enables the user to find out all of the transactions associated with a particular item that have been made during the previous 45 days. He enters only the DYMAC name of the item.

ITEMS IN TRANSIT enables the user to display the entire file of items that are in transit in the facility.

DYSS also has been changed to store SNM amounts in whole accountable units to be consistent with the LASL Accountability System (LAS)** which interfaces with DYSS. For example, the DOE requires that depleted uranium be accountable in kilograms, so a measured quantity of 3575 g is now stored in DYSS as 4 kg, whereas previously it was stored as 3575 g. Plutonium is accountable to the gram, and a measured quantity of 23.42 g is now stored as 23 g.

*Group E-5.

**The Nuclear Materials Department, which accounts for all nuclear material at LASL, administers LAS.

Work began on the transactions for the metallography area in the metal fabrication wing. It involves designing the transaction sequences, receipt areas, types of measurements to be made, and how material balances will be determined. Transactions for the advanced carbide fuels area are still being modified.

The Eclipse version of DYSS will be altered to run on the NOVA 840 for training and also for backup. The programming team will recompile the FORTRAN source programs and rewrite and assemble the assembly language sources for the NOVA. They will also rewrite the INFOS-based routines used for data base searches and updates to run on the NOVA.

1. Nuclear Materials Officer (NMO) Transactions

The transactions for the NMO now branch off at the second decision point on the display screen. Only the NMO and his staff have the password to access these transactions. If the NMO wants to make an internal transaction, he selects one of the options given in Fig. 57a. If he wants to make an external transaction (a transaction that affects the station balance of the facility), he selects one of the options given in Fig. 57b.

Some of the NMO's established tasks have been programmed into DYSS. These tasks, shown below in order of performance, pertain to the procedure the NMO follows at the end of the processing day.

1) See that no unauthorized items are left in the in-transit file; if an authorized item is left, arrange with the appropriate custodian to make a RECEIVE transaction.

2) Review external transactions to make sure that amounts of shipments and receipts have been entered correctly.

3) See that the station balance is correct. (Station balance—the amount of nuclear material remaining in a facility—is the difference between the amount of nuclear material that has been received and the amount that has been shipped out.) DYSS maintains a general ledger on a disk to keep track of station balance. The NMO checks the current inventory against the entries in the general ledger. If the two agree, the station balance is

```
<<< SELECT OPTION >>>
  1...COUNT ROOM: RECEIVE INTERNAL SHIPMENT
  2...COUNT ROOM: SEND INTERNAL SHIPMENT
  3...VAULT: RECEIVE INTERNAL SHIPMENT
  4...VAULT: SEND INTERNAL SHIPMENT
```

a. Internal transactions.

```
<<< SELECT OPTION >>>
  1...COUNT ROOM: RECEIVE EXTERNAL SHIPMENT
  2...COUNT ROOM: SEND EXTERNAL SHIPMENT
  3...VAULT: RECEIVE EXTERNAL SHIPMENT
  4...VAULT: SEND EXTERNAL SHIPMENT
  5...CHANGE ENRICHMENT
  6...DECAY
  7...NORMAL OPERATING LOSS
  8...WRITE-OFF TO MUF
  9...DEGREDATION
 10...WRITE-OFF SHIPPER-RECEIVER DIFFERENCE
```

b. External transactions.

Fig. 57.
NMO option list.

correct. If they don't agree, the NMO must determine which account is in error and resolve the error with the account custodian.

4) Make a tape of the day's internal transactions for the Nuclear Material Department.

D. Project Sandbag (R. H. Augustson, W. R. Severe, D. C. Amsden)

To shake down the DYMAC system before actual production began at TA-55, Group Q-3 designed an exercise to test the transactions for the advanced carbide fuel process, the first to begin operation in the facility. (A description of the process appears in Ref. 3, pp. 59ff.) Originally, depleted uranium was to be used in the exercise, but slippage in the building schedule precluded its introduction into the facility at that time. Q-3 went ahead with its plans nevertheless, substituting cans of sand for depleted uranium. From October 3 through 11, 1977, "Project Sandbag" exercised every piece of code in DYSS.

The exercise was designed to test various aspects of the system:

- Accuracy of the computer software,
- Design of the accountability system,
- Reliability of the communications network, and
- Efficacy of making transactions in a production environment. The exercise also was intended to acquaint DYMAC personnel with the system.

Q-3 personnel prepared 80 cans of sand that were to undergo the entire sequence of processing from entry into the facility, through processing, to storage in the vault as product. On the basis of operating experience at DP Site, each of 40 cans was filled with 217 g of mock plutonium and each of the remaining 40 was filled with 868 g of mock uranium. The Sandbag procedure specified how much of each to blend, divide out, and remove as scrap and material in process (MIP) during processing. The cans did not enter the glovebox system, but were placed directly beneath the appropriate gloveboxes during processing. The only NDA instrument used during the exercise was the off-line electronic balance.

The cans entered the facility as two shipments. One shipment came through the count room, where it was entered into the DYMAC inventory and sent to the vault. The other shipment came through the vault, where it was entered in the inventory and where it remained until it was brought to the process area 16 cans at a time. Figure 58 shows the instruction sheet to be followed once the cans are brought from the vault to the material management room.

MANAGEMENT ROOM

- RECEIVE FEED STOCK FROM VAULT AT CHEMISTRY VALUE
- SEND TO PROCESSING AREA

OXIDE-BLENDING

- RECEIVE FEED STOCK FROM MANAGEMENT ROOM
- MAKE A BATCH FROM PART OF FEED STOCK, WEIGH BATCH WITH BALANCE
- ADD CARBON, UO_2, AND TRANSFER TO OXIDE PRESS

OXIDE PRESS (make briquettes from powder, some scrap will spill)

- MEASURE SCRAP WITH BALANCE
- SEND BRIQUETTES TO REDUCTION FURNACE

REDUCTION FURNACE (briquettes reduced to carbide)

- RECEIVE BRIQUETTES, WEIGH THEM WITH BALANCE, AND DETERMINE MIP FOR OXIDE PRESS
- WEIGH REDUCED BRIQUETTES WITH BALANCE, TRANSFER TO COMMINUTE, AND DETERMINE MIP

COMMINUTE (reduced briquettes are ground into powder, some scrap is generated)

- MEASURE SCRAP WITH BALANCE
- TRANSFER POWDER TO AUTOMATIC PELLET PRESS AND DETERMINE MIP

AUTOMATIC PELLET PRESS (press powder into pellets, some scrap will spill)

- MEASURE SCRAP WITH BALANCE
- WEIGH PELLETS WITH BALANCE, TRANSFER TO SINTERING FURNACE, AND DETERMINE MIP

SINTERING FURNACE (sinter pellets at high temperature)

- WEIGH SINTERED PELLETS WITH BALANCE, SEND TO GRINDING, AND DETERMINE MIP

GRINDING (grind pellets to specified diameter, some scrap will result)

- RECEIVE SINTERED PELLETS
- MEASURE SCRAP WITH BALANCE
- WEIGH MACHINED PELLETS WITH BALANCE, TRANSFER TO INSPECTING, AND DETERMINE MIP

INSPECTING (pellets inspected for conformance to specifications; some rejected as scrap)

- MEASURE SCRAP WITH BALANCE
- WEIGH INSPECTED PELLETS WITH BALANCE, SEND TO VAULT VIA MANAGEMENT ROOM, AND DETERMINE MIP

Fig. 58.
Instruction sheet for Project Sandbag. Each heading denotes a unit process; each circle denotes a transaction to be made for that unit process.

Once in the advanced fuels line, the can contents were blended, weighed, and divided into pellet lots. At each step the appropriate transaction was entered at the terminal and recorded in the DYMAC inventory. Material balances were calculated for each of the seven unit processes in the fabrication line. The information on current inventory, transaction activity, and in-transit monitoring was available to the process operator and NMO just as it will be when the DYMAC system is in full operation.

In all, 5233 g of mock plutonium and 20 928 g of mock uranium were blended and processed, producing 104 cans in all: 12 cans of product, 37 cans of scrap, 7 cans of MIP, and 48 cans of leftover feed stock. Of the original 80 cans entering the facility, 32 remained unprocessed in the vault.

The exercise was worthwhile for several reasons. It showed that the DYMAC system was well designed and functioned well in the processing area as predicted. It afforded DYMAC personnel the opportunity to learn how to interact with the DYMAC system. It brought to light user messages that were not easily understood, and these were reworded. Perhaps most important, the exercise allowed DYMAC personnel to find and correct bugs in the program and in the design of the system. Following are examples of the errors that were subsequently corrected:

- The program neglected to change the item description in one of the transactions, resulting in a miscalculation of the factored weight.
- When two users tried to access the same record simultaneously, it became permanently locked, and then was completely lost from the system.
- Terminal response time was slow because program development was going on in background mode. (This will not be the case when the system comes up for full-time operation.)
- The transaction for sending an item from the count room to the vault was not in working order.

During the exercise, DYMAC personnel learned how to correct operator-entered errors by using a general transfer to restore the inventory to what it should be. In the next few months DYSS will be changed so that only supervisors can correct erroneous entries in the inventory. The programmers imposed more diagnostics for data entry to reduce entry errors. They added a summary of the transaction results to the bottom of the full display, and they reformatted the full display to be more easily readable.

After incorporation of all the changes and corrections into DYSS, Project Sandbag was repeated in November on a smaller scale without using cans of sand; again, every transaction was exercised to verify that the system was working correctly. No problems were encountered, but a few more improvements were made to messages and formats.

Thus, Project Sandbag exercised all of the system features that pertain to shipment and receipt of nuclear material at TA-55 and to processes in the advanced carbide fuels laboratory. Similar exercises are planned for other processes.

E. Training (R. H. Augustson and D. C. Amsden)

Two training classes were held—one in November, the other in December—to familiarize process personnel with the DYMAC system. The course offered 6 h of lecture and 6 h of practical training. Lectures covered the principles of accountability: dynamic material balancing, DYMAC record structure, transactions, NDA measurements, on-line reports, and the functions of the NMO. Practical training consisted of 2 h of a get-acquainted exercise at the terminal in the training room and, the next day, a 4-h exercise in the plutonium facility in the advanced carbide fuels laboratory. During the exercise in the facility, the trainees followed a detailed sequence of instructions to guide them in processing cans of sand (much as in Project Sandbag), making transactions at the terminal for each step of the process, and evaluating their material balances at the conclusion of the exercise.

The practical training reinforced the lectures. At the end of the course the process operators and supervisors had learned to use the DYMAC system correctly and effectively. They were particularly quick in learning how to use the on-line reporting capability to find out the status of their processing.

Twenty-one people have taken the course (see Table XVI); nine work in the actual processing area and three are on the staff of the NMO. Others who have taken the course are in the Nuclear Materials Department or in the LASL safeguards groups.

In late December a special session was held to train nine people in the duties and capabilities of the NMO. After a short lecture session, the trainees learned how to use the terminal to perform the NMO's duties.

A User's Manual for the DYMAC system is being prepared. It will serve both as a text for the training course and a reference manual for the operating personnel in the facility.

TABLE XVI

DYMAC TRAINEES

Function	No. Persons Trained
NMO staff	3
Process operators and supervisors in CMB-11	9
Nuclear Materials Department	3
Safeguards scientists and technicians in Q-3 and Q-4	6

PART 4

INTEGRATED SAFEGUARDS SYSTEMS AND TECHNOLOGY TRANSFER

GROUP Q-4

R. J. Dietz, Group Leader

A major effort of Group Q-4 is to establish design and performance requirements for cost-effective, integrated safeguards systems for typical nuclear fuel cycle facilities and to assess the impact of these requirements on process selection and plant design and operating criteria. Functions of these facilities include uranium enrichment, spent fuel reprocessing, plutonium nitrate-to-oxide conversion, plutonium recycle-fuel fabrication, scrap recovery, and waste handling and disposal. Results of these studies provide facility designers and operators with working guides and performance criteria for materials control and accountability systems that can be integrated with physical protection and process control systems in fuel cycle facilities.

By maintaining a high level of participation in NDA and systems technologies through interactions with R&D organizations, industry, and government agencies, Q-4 staff members coordinate and disseminate safeguards technology throughout the nuclear community. Q-4 also provides technical information and assessments of both domestic and foreign fuel cycle facilities to DOE on a high-priority, as-needed basis.

During the past 4 months the Q-4 effort was divided evenly between two line items: Safeguards Concept Definition for Fuel Cycle Facilities and Technology Transfer and Support to DOE-SS, their subcontractors, and the safeguards industry. The efforts of Q-4 are reported in journals, consultations, briefings, and topical reports which are listed toward the back of this status report.

I. SAFEGUARDS CONCEPT DEFINITION FOR FUEL CYCLE FACILITIES

The safeguards concept-definition report, "Coordinated Safeguards for Materials Management in a Fuel Reprocessing Plant," has been published as LA-6881.[14] The report is in two volumes: the second volume contains the technical background in the form of a series of appended monographs for the safeguards analysis contained in the first volume.

The systems approach described in LA-6881 and in the previous status report[3] was used to assess the safeguards effectiveness of a materials management system for a nitrate-to-oxide conversion plant designed to couple the fuel reprocessing plant to the mixed-oxide fuel fabrication plant previously studied in LA-6536.[24] The conversion report (LA-7011) will be the third of a series that treats each of the process facility modules characteristic of a complete national fuel cycle. The report, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility,"[25] was completed on October 15, 1977, circulated in draft form, and favorably reviewed.

The conversion facility reported on in LA-7011 could receive the product from the 5-tonne/day nuclear fuel reprocessing plant described in Ref. 14 and convert it to plutonium oxide feed material for the mixed-oxide fuel fabrication plant described in Ref. 24. Under steady-state conditions, the conversion process effectively couples the chemical separations plant to the fuel fabrication facility and has enough overcapacity to make up for production transients and periods of decoupled operation. The materials management systems are compatible in all three facilities considered to date; they reinforce each other and, when combined, could share the safeguards responsibilities in a reprocessing complex where the components of more than one facility are collocated.

The conversion process plays a pivotal role in safeguarding the whole nuclear fuel cycle against subnational diversion or as part of a national nuclear nonproliferation policy. The conversion facility is unique in the civilian power economy because it invariably handles large amounts of concentrated, highly purified plutonium in a form that could be an attractive target for diversion or national misappropriation.

Many of the features that make the conversion process attractive for diversion also make it more amenable to materials management by using the concepts of unit process accounting and dynamic materials balancing previously developed for dynamic accountability systems. Low radiation levels and improved accessibility to the process facilitate the improved measurements necessary to implement these concepts.

The conversion process selected is based on a reference design provided by SRL. It involves the semicontinuous precipitation of plutonium (III) oxalate in four parallel process lines. The precipitate is dried and calcined to a plutonium oxide product that has the desired chemical and physical properties. The process is based on demonstrated technology, developed early in the weapons program at Los Alamos, and was selected for maximum ease of process control, product flexibility, and production scaling.

Two accounting strategies were devised for the materials management system; they involve treating each process line either as two contiguous unit processes or as a single unit process. In both strategies, dynamic material balances are drawn around the processing of each batch (~2 kg of plutonium), and both strategies incorporate the *same* set of accountability measurements. Key measurement points are located at the feed-solution receipt tanks (conventional volume measurement and on-line concentration measurement by absorption-edge densitometry) and at the product canister loading area (measurement by neutron counting or calorimetry). Concentration measurements require a companion determination of isotopic composition by gamma-ray or mass spectrometry. The strategies differ in that measurements of wet plutonium oxalate precipitate in filter boats are treated as material transfers in the first (two-unit-process) strategy and as part of the in-process inventory in the second (one-unit-process) strategy. Until an improved measurement technique is developed for wet plutonium precipitate, modeling and simulation studies based on projected production data and realistic measurement parameters clearly show that the single-unit-process strategy provides better sensitivity to both short-term and long-term, low-level diversion.

The two strategies are complementary in that the same measurement data are combined to form dynamic material balances in two ways. Diversion can be better localized by the first accounting strategy because it contains two unit processes in each process line. In the second strategy the wet boat measurements are combined into the in-process inventory (the boats constitute the furnace in-process inventory); therefore, the inaccuracy of the wet boat measurements tends to cancel, resulting in an improved diversion sensitivity.

The diversion sensitivity of the one-unit-process accounting strategy is shown in Table XVII. The sensitivities were determined from a series of computer-generated process histories for which decision-analysis algorithms were developed to handle large quantities of materials measurement data in near-realtime. They should be compared to the value required for conventional inventory-based systems—a 33-kg limit of error for material balances (LEMUF) determined every 2 months for the entire process area.

TABLE XVII

DIVERSION SENSITIVITY[a] IN THE CONVERSION PROCESS

Detection Time	Average Diversion per Batch (kg Pu)	Total at Time of Detection (kg Pu)
1 batch (1.35 h)	0.13	0.13
1 day	0.03	0.63
1 wk	0.01	1.24
1 month	0.005	2.65

[a]As determined for a single process line in the second accounting strategy in accordance with ERDAM Appendix 7401-C, "Nuclear Materials Management and Safeguards System Handbook."

The sensitivities reported in Table XVII are for the materials accountability part of a stand-alone safeguards system for the conversion process. Total installed cost is estimated at <$6M and the annual operating cost should be less than half that. Mean time between failures of the system should be >3 months, and a ~5-h mean time is estimated for repair (or replacement) of a vital component. Costs would be significantly lower if the conversion process dynamic accountability system were incorporated into the overall safeguards system of a larger collocated nuclear complex of the type suggested for both domestic and international commercial reprocessing facilities.

The utility and attractiveness of the material handled in conversion facilities make the facilities appropriate for special nonproliferation considerations in the so-called Bonded Crucial Facility (BCF) concept. In this concept, the crucial facility (the conversion plant) is expanded to include storage of the product and feed inventory for the adjacent chemical separations and fuel fabrication plants. The expanded complex can provide continuous monitoring of the production and use of plutonium on an industry-wide or national scale. The BCF could be operated under an umbrella of enhanced safeguards that could be extended to include multinational control.

The most important conclusions of the nitrate conversion study follow.

- Strategic quantities of SNM can be safeguarded in a high-throughput, nitrate-to-oxide conversion facility at reasonable cost with minimal process disruption. Field testing and evaluation of safeguards system components are required to validate and refine the system design.
- The conversion process is the keystone to safeguarding the back end of the fuel cycle. Studies of these and other processes must continue so that the most safeguardable and proliferation-resistant process can be selected.
- The interaction of dynamic materials accounting, process control, and materials containment and surveillance must be investigated further.
- Instruments for use at key measurement points must be well characterized and field tested. Testing of absorption-edge densitometers, neutron well counters, gamma spectrometers, and calorimeters is required.
- Techniques for measuring plutonium oxalate slurry in precipitators and filter boats should be demonstrated and evaluated.
- For each type of conversion process, arrangements of unit processes, process equipment, and operating procedures must be evaluated to exploit any inherent safeguards advantages.

II. SAFEGUARDS EVALUATION OF ALTERNATIVE FUEL CYCLE FACILITIES

Group Q-4 now is tasked to provide safeguards technical support to the Nuclear Production Division's (NPD) alternative fuel cycle technology programs. Two programs are involved: the Alternative Fuel Cycle Technology (AFCT) program and the Thorium Fuel Cycle Technology (TFCT) program, both administered by the Savannah River Operations Office under the technical guidance of SRL.

Our technical support will depend heavily on our completed and ongoing concept definition studies (described in Part 4, Sec. I), which provide a point of departure for evaluating the relative nonproliferation advantages (or disadvantages) of several alternative modes of operation for existing and proposed fuel cycle facilities and assessing their impact on existing fuel cycle facility designs. The first alternative, now being studied, is the co-conversion of coprocessed, mixed uranium-plutonium nitrate solutions via the suggested Coprecal process. LASL personnel have visited SRL and the Coprecal pilot line at the General Electric Company's Vallecitos Laboratory and are evaluating a reference process design for its safeguardability.

Another alternative, that of operating fuel cycle facilities under the ^{233}U-^{232}Th fuel cycle is also being studied. Details of this fuel cycle are not so well characterized as those for uranium-plutonium, and

we are developing reference processes and analytical methods jointly with other laboratories. To acquire a data base sufficient to characterize a typical thorium facility, LASL personnel have discussed the thorium fuel cycle with SRL personnel and the technical staff of the HTGR programs at Oak Ridge National Laboratory and the General Atomic Company.

Topical reports on the status of analytical techniques applicable to safeguarding the thorium fuel cycle and a preliminary evaluation of the safeguardability of the Coprecal process will be issued during the next reporting period.

III. DEVELOPMENT OF METHODOLOGY FOR SAFEGUARDS EVALUATION AND ANALYSIS

All safeguards effectiveness evaluations of fuel cycle facilities require an intimate knowledge of the processes and operational details of the facility as well as the unique characteristics of a dynamic materials management system that subdivides the process in time and space into individual unit process accounting areas. Thus large quantities of process and materials balance data must be analyzed efficiently and in a manner amenable only to computerized data handling. This is done by modeling and simulating plant operations and materials flows and by computerizing the routine aspects of the decision process so that the safeguards officer is not deluged with data from normal plant operations—data that might be used by an informed divertor to obscure abnormal operating conditions. Accordingly, the development and standardization of methodology for decision analysis and for effectiveness evaluation are increasingly important components of these studies and have helped immensely in defining areas where process and measurement improvements are indicated. The development of process and facility modeling and simulation methodology is a continuing effort that forms an integral part of the facility safeguards studies and that is reported separately. Decision analysis and safeguards systems reliability are common to all such studies and are therefore worthy of special attention.

Development and refinement of decision analysis as applied to nuclear materials accounting for safeguards continues. One now can process large amounts of data having various degrees of correlation, and several different decision algorithms can be used. The mathematical foundation is complete, and graphical display techniques are highly developed, as illustrated by a 16-mm motion picture film showing the results of the decision analysis code DECANAL being applied to data from a reprocessing plant.

The salient features of decision analysis (including the movie) were presented in an invited paper, "Decision Analysis in Safeguarding Special Nuclear Material,"[26] presented at the 1977 ANS Winter Meeting in San Francisco. The necessary computerized data acquisition, handling, and analysis raises another generic problem common to future safeguarded facilities, namely, that of digital systems reliability, security, and vulnerability.

We have written a computer program, RELSIM, to simulate the reliability of large-scale data acquisition and information system networks. The program is used to compute projected reliability performance, including downtime associated with individual hardware modules and the network as a whole. The program's modular design permits identification of major contributors to downtime at each hierarchical level, and a sensitivity analysis is conducted to determine changes of overall system performance with improvements in individual component reliability. The effects of incorporating redundant architectures on overall reliability provide the basis for cost/performance trade-off studies.

IV. INTERNATIONAL SAFEGUARDS

Safeguarding foreign fuel cycle facilities, either under the auspices of the IAEA or through a bilateral agreement with the host country, is an important aspect of the US nonproliferation policy. Q-4 has responsibility for three tasks in direct support of IAEA activities: 1) the definition of materials accounting data required by the IAEA for safeguarding modern fuel cycle facilities, 2) a review of dynamic materials control concepts for IAEA personnel, with emphasis on applications to international safeguards, and 3) techniques for materials control in critical-assembly facilities. Task 1 will be completed in FY 1978; Tasks 2 and 3 extend at least through FY 1979.

A report, "Materials Flow and Inventory Data for a 200-Tonne Per Year Mixed-Oxide Fuel Fabrication Plant,"[27] was submitted to the IAEA through the International Safeguards Project Office (ISPO) as part of the ISPO program plan for US Technical Assistance to IAEA Safeguards. Materials flow and inventory data derived from Ref. 24 formed the basis for the report. The IAEA has requested that such data be made available for use in planning safeguards for simular facilities worldwide and has identified mixed-oxide and highly enriched uranium fuel fabrication plants as being of special interest.

Task 3 was addressed by the publication of the draft report "Preliminary Concepts for Materials Measurement and Accounting in Critical Facilities,"[28] which is being circulated for comment. Large, fast-critical-assembly research facilities, used to simulate plutonium-fueled breeder reactors, are operating in many of the developed countries of the world. Two such facilities are the Zero Power Plutonium Reactor (ZPPR) at Idaho Falls, Idaho, used as the reference facility for this study, and the Fast Critical Assembly (FCA) at Tokai, Japan. These large facilities can simulate breeder reactors as large as 1000 MWe with cores containing as much as 3000 kg of fuel.

Large critical facilities present a unique safeguards problem. Inventories can include thousands of small fuel pieces constituting tonnes of fissile material in relatively pure form. The widespread use of such facilities in nonweapons states is a distinct proliferation hazard. Complete physical inventories at critical facilities can be performed only infrequently, perhaps annually, because of the excessive burden placed on facility personnel and operations. The major problem in safeguarding critical facilities is the timely verification of in-reactor inventories without serious disruption of reactor operations during the long periods between physical inventories.

We propose that when the reactor is initially loaded a reference configuration be established and the integral reactivity and possibly other reactor parameters be meaured. These measurements are sensitive to the total inventory of fissile material in the reactor, but the sensitivity is dependent on the reactor size, composition, and material distribution. Periodically the reactor would be returned to the reference configuration for remeasurement and verification of its characteristics. In addition, fuel-bearing drawers would be sampled periodically from the reactor inventory, and verification measurements would be made in the reactor area using NDA techniques. A sampling plan for fuel drawers can be developed that is tailored to the distribution of SNM in the reactor matrix and to the sensitivity of the integral measurements. The storage vault would be maintained as an item control area in which storage canisters would be stored under tamper-proof seal.

Specific NDA techniques to be investigated for measurements of fuel drawers and vault canisters are gross neutron and neutron coincidence counting, gamma-ray spectroscopy, and autoradiography in which images of individual fuel plates are produced on x-ray film. The autoradiography technique has been developed and applied successfully at Argonne National Laboratory to drawers and canisters containing plutonium fuel plates. Neutron and gamma-ray instruments developed at LASL have been fielded by IAEA inspectors at the Japanese FCA facility; however, further developmental effort is required to characterize and optimize instrumentation for the FCA application.

The proposed verification techniques are sensitive to different properties and characteristics of the fissile material. Although one technique might be subverted, a combination of these techniques and

related sampling plans provides increased effectiveness and reduced vulnerability. The detection sensitivity for a combination of techniques can be a very complicated function. The question of which measurement and accounting strategies afford the best assurance depends on a variety of factors including operational constraints. Computerized modeling and simulation methodology and decision analysis tools developed at LASL during previous safeguards systems studies can be used to address these complexities.

The ZPPR facility has been selected as a reference to maintain the necessary contact with reality. In future studies, the effectiveness of various measurement and accounting strategies against diversion will be evaluated and compared by use of the modeling and simulation approach. Our experience indicates that this approach yields reliable results and provides a credible basis for facility-specific recommendations.

V. TECHNOLOGY TRANSFER AND TECHNICAL SUPPORT

Through visits, briefings, and consultations, the Q-4 staff has presented the results and methodology of the DOE-SS/LASL Coordinated Safeguards Program to the staffs of DOE-SS, the SRP, SRL, and Savannah River Operations Office, the General Electric Company, General Atomic Company, Clemson University, Sandia Laboratories, Systems Planning Corporation, International Energy Associates Limited, Argonne National Laboratory (Idaho), Allied-General Nuclear Services, Oak Ridge National Laboratory, DOE-NPD, DOE-ISA, and the Arms Control and Disarmament Agency. At Los Alamos we have briefed representatives of several of the above agencies, plus those from Science Applications Incorporated, Lawrence Berkeley Laboratories, the Nuclear Regulatory Commission, Pacific Northwest Laboratory (Battelle), the IAEA, and Westinghouse Corporation.

Technology transfer briefings, under the bilateral safeguards information-exchange provisions of the nonproliferation treaty (NPT) were given to representatives of the governments of Belgium, Norway, Denmark, Greece, South Korea, and Japan. The information exchange with Japan was especially effective in view of the timeliness of the US-Japanese Tokai reprocessing plant interchange. Another group of Japanese was briefed subsequently on the safeguards implications of nitrate conversion processes, as a consequence of Q-4's previous interactions with the Japanese and our identification of the conversion process as being of special safeguards concern.[25]

Technical support was provided to DOE concerning the Tokai negotiations, foreign and international safeguards experiments, and the future utilization of the Barnwell Nuclear Fuels Plant (BNFP). A review article on the process features of the uranium-thorium fuel cycle was prepared for technical guidance.[29] A 3-day meeting was hosted with NRC and NRC contractor personnel to discuss and review their materials accounting study, NUREG-0290.[30] LASL's position, described in Ref. 3, p. 82, was successfully defended. DOE-SS has requested a similar review[31] of another NRC materials control study called Controllable Unit Accounting (CUA) performed by the Mound Laboratory. It will be completed in January 1978. We also reviewed a proliferation scenario for the thorium fuel cycle.

VI. MISCELLANEOUS

The most important requirements for meaningful technology transfer are timeliness and effective communication. Q-4 continues to develop in-house capability for effective response.

The major components of the Safeguards Modeling and Simulation Computer System have been installed and are undergoing acceptance tests. The system is based on a typical low-cost commercial

minicomputer (Prime Model 300) of the type available to any industrial-scale fuel cycle facility and will allow us to perform modeling and simulation exercises more nearly in realtime and to simulate effectiveness evaluations and decision analyses on modest equipment more nearly representative of what would be purchased as part of a coordinated safeguards system. Current codes are being converted to this machine, which still lacks its full complement of peripherals because of delays in delivery.

Reporting capabilities have been enhanced with the delivery of update equipment to convert the current Wang System 20 word processor to a System 30. The System 30 has a capacity larger by an order of magnitude, permits more flexible editing, and can provide word processing capability simultaneously to three Safeguards Groups: Q-1, Q-3, and Q-4.

Computerized color graphics are now available to Q-4 on a quick-response basis and are being used extensively as an effective teaching and communication aid. A 16-mm color motion picture clip was prepared as part of an ANS invited paper[26] to illustrate the decision analysis process and alarm-sequence displays characteristic of realtime accounting systems.

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GLOSSARY

AFCT	Alternative Fuel Cycle Technology
ANSI	American National Standards Institute
AWCC	active well coincidence counter
BCF	bonded crucial facility
BNFP	Barnwell Nuclear Fuels Plant
BWR	boiling water reactor
CUA	Controllable Unit Accounting
DOE	Department of Energy
DYMAC	dynamic materials control
DYSS	DYMAC software system
FCA	Fast Critical Assembly
FFTF	Fast Flux Test Facility
HEDL	Hanford Engineering Development Laboratory
HFS	hydrofluoric slag
HLNCC	high-level neutron coincidence counter
HPFL	High Performance Fuel Laboratory
IAEA	International Atomic Energy Agency
ICPP	Idaho Chemical Processing Plant
INMM	Institute of Nuclear Materials Management
ISPO	International Safeguards Project Office
LWR	light water reactor
MCA	multichannel analyze
MIP	material in progress
MTR	materials testing reactor
NDA	nondestructive assay
NMO	nuclear materials officer
NPD	Nuclear Production Division
NRC	Nuclear Regulatory Commission
NUMEC	Nuclear Materials and Equipment Co.
PSD	pulse shape discrimination
PUSAS	plutonium solution assay system
PVM	personnel vehicle monitor
PWR	pressurized water reactor
RD	random driver
SAI	solution assay instrument
SGS	segmented gamma scanner
SNM	special nuclear material
SRL	Savannah River Laboratory
SRP	Savannah River Plant
SSAS	small sample assay station
TC XRF	transmission corrected x-ray fluorescence
TFCT	Thorium Fuel Cycle Technology
TNC	thermal neutron coincidence counter
USAS	uranium solution assay system
XRF	x-ray fluorescence
ZPPR	Zero Power Plutonium Reactor

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