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Progress Report

MASTER

**Nuclear Safeguards
Research and Development
Program Status Report
January—April 1978**



University of California



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NUCLEAR SAFEGUARDS RESEARCH AND DEVELOPMENT

Program Status Report

January-April 1978

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.

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NUCLEAR SAFEGUARDS RESEARCH AND DEVELOPMENT EXECUTIVE SUMMARY

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The Nuclear Safeguards Program at the Los Alamos Scientific Laboratory (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.

The continuing program of nondestructive analysis and verification of spent fuel has included a series of Monte Carlo calculations designed to demonstrate the feasibility of using measured neutron flux to infer burnup in pressurized water reactor (PWR) assemblies stored under water. The calculations have also shown that neutron measurements are sensitive to interior fuel pins. Additional gamma-ray and neutron measurements have been carried out on irradiated materials testing reactor (MTR) fuel assemblies at LASL's Omega West reactor and similar measurements are scheduled for the Big Rock Point Nuclear Plant in Michigan. These measurements will provide information for optimizing methods of determining cooling time, burnup, and activity profiles for irradiated boiling water reactor (BWR) fuel assemblies. Measurements of neutron and gamma-ray dose rates from high-enrichment spent fuel elements were made at the spent fuel storage facility at Idaho Falls to provide design information for the ^{252}Cf Shuffler neutron interrogation system; the Shuffler system is being designed to measure the ^{235}U content in waste

canisters from the fluorine dissolver and also in spent fuel assemblies before dissolution. These and other studies indicate the promising potential of active neutron interrogation techniques for assaying fissile material content in irradiated reactor fuel.

A number of advances have been made during this report period in the area of neutron assay technique development. In support of studies on plutonium-uranium coprocessing, in-process NDA measurements have been carried out on wet and dry plutonium samples using a SNAP-II total neutron counter and a thermal-neutron coincidence counter. With suitable standards and appropriate correction for neutron self-multiplication, neutron coincidence counting is expected to provide a reasonable NDA method for assaying wet plutonium oxalate cake.

In support of IAEA inspection and verification requirements, both passive and active neutron assay methods are being evaluated for verification of plutonium and uranium in fast critical assemblies (FCA). Active neutron interrogation methods, e.g., using the ^{252}Cf Shuffler or the active well coincidence counter (AWCC), are required for measuring FCA fuel drawers and plate storage canisters containing mixtures of plutonium and uranium because passive neutron methods are not applicable to ^{235}U metal. The combination of passive neutron and gamma-ray techniques (achievable in the same instrument) can provide rapid quantitative verification of plutonium in FCA fuel.

In addition to neutron methods, passive gamma-ray techniques are being evaluated for the quantitative verification of plutonium in FCA fuel drawers. The ^{239}Pu and ^{241}Pu content of an entire drawer can be measured simultaneously; typical accuracies of 1.5% (1σ) for ^{239}Pu and 1% for ^{241}Pu were obtained. In addition to fuel drawers, certain types of storage canisters can be assayed rapidly by gamma measurements. The main advantages of

gamma assay are that it uniquely identifies the fissile isotopes present, can be used to determine isotopic ratios, and, with proper calibration, the isotopic content of fuel containers. The unique gamma signature (albeit a "surface" signature) of various plutonium isotopes makes plate substitution difficult. For verification of storage vault inventories an array of ^3He neutron detectors on the walls and ceiling of a vault is being investigated experimentally. More detailed measurements are required to determine the sensitivity of such an array to plutonium movement or removal, but with strict entry/removal control, excellent sensitivity (e.g., <1%) may be possible for plutonium.

Neutron coincidence counting of large plutonium oxide or metal samples is strongly affected by self-multiplication within the sample, e.g., for 1- to 2-kg plutonium metal buttons the response may be increased by a factor of 2 or more. Simple correction factors based on the measured doubles/singles ratio have been derived and appear useful for removing the effects of self-multiplication on neutron coincidence counting.

IAEA inspectors have found the portable high-level neutron coincidence counter (HLNCC) to be useful for plutonium assay in field applications. The HLNCC has not been applied to the assay of ^{235}U or ^{233}U because of their extremely low spontaneous fission yields. To provide assay capability for ^{235}U and ^{233}U a small AmLi neutron interrogation source has been combined with a ^3He thermal-neutron well counter that detects coincidence neutrons from the induced-fission reactions. The source-detector-moderator-reflector configuration of this AWCC has been optimized to detect the induced-fission spectrum neutrons with maximum detection efficiency while at the same time having a low efficiency for counting the α, n neutrons from the AmLi interrogation source. The performance data obtained from these studies are being used to design a lightweight, highly stable, ruggedized version of the AWCC for field use by the IAEA.

As indicated in earlier reports, a Bragg-reflection wavelength dispersive spectrometer should be useful as a narrow "band-pass" filter for fission-product background suppression in absorption edge densitometry and x-ray fluorescence measurements on hot solutions. Preliminary tests using a LiF crystal indicate that with suitable arrangement of source-slit geometry such a system will perform ade-

quately as a band-pass filter for low-energy photons in conjunction with either densitometry or x-ray fluorescence measurements on hot solutions.

The L-edge densitometer designed to measure multiple concentrations of special nuclear material (SNM) in solution has undergone full test, evaluation, and calibration at LASL and has been installed at the Savannah River Laboratory (SRL) for assay of uranium and plutonium in coprocessing demonstrations there. The LASL test and evaluation results on mixed solutions of uranium and plutonium (concentration ratio 10:1) indicate assay accuracies of >5% for plutonium and >1% for uranium. Two other systems being developed at LASL for test and evaluation at the Savannah River Plant (SRP) are (1) a ^{252}Cf Shuffler neutron interrogation system for assay of highly enriched ^{235}U materials at the reactor fuel (UA1) fabrication facility at SRP and (2) a Shuffler-type active assay system for nondestructive assay (NDA) of waste canisters and spent fuel packages at the proposed Fluorinel Reprocessing Facility in Idaho.

A number of instruments have been identified that could form the basis of an international perimeter safeguards system around a large-scale centrifuge facility, such as the Portsmouth Gas Centrifuge Enrichment Plant (GCEP). Conceptual designs have been developed for five basic instruments that can be applied to the verification of the GCEP declared material balance and the possible detection of high-enriched uranium production. These include in-line enrichment monitors for feed, product, and tails assay, a trap material enrichment meter, and area radiation monitors. This capability should provide the equivalent of a continuous inspector presence at the feed and withdrawal stations and a 100% sampling plan.

An initial conceptual design of a large-vehicle monitor for a facility such as GCEP has been completed; UF_6 is detected via passive neutron emission with estimated sensitivity of ~ 1 kg UF_6 unshielded. Studies are also under way in the areas of UF_6 cylinder verification meters, shipping dock, trap, waste, and plumbing monitors. The total measurement, monitoring, and verification capability envisaged in our present conceptual designs should provide an effective overall perimeter safeguards system for a modern, high-throughput enrichment facility such as GCEP.

The Intel 8748 microcomputer shelf monitor has been debugged and interfaced with a Nova 3 minicomputer. Sixteen such monitors are being fabricated for the Real Time Inventory shelf monitoring system to be tested and demonstrated at TA-18. On the basis of results from this test program, a 200-detector system, now being designed, has been proposed for one room of the plutonium storage vault at TA-55.

During this reporting period, the DYMAC program began formal operation at the new Plutonium Processing Facility. The first shipment of plutonium entered the TA-55 facility on February 9, and regular daily shipments have been received since. The Metal Fabrication wing began full operation in April and the Advanced Carbide Fuels work in the ^{239}Pu R&D wing, currently involved in a formal shakedown program using depleted uranium, will start working with plutonium in June.

The DYMAC computer currently has 40 ports for use by terminals and measurement instruments. At the close of the present reporting period, 24 interactive terminals and 10 electronic balances were coupled to the computer. To date, three NDA instruments have been installed in the TA-55 counting room. The Metal Fabrication process has begun using two thermal-neutron coincidence counters (TNCs) in the casting receipt area; 16 TNCs are to be installed at the new plutonium facility. By April 30, 1978, the DYMAC inventory contained 661 items, most of them located in the TA-55 storage vault. During this period the DYMAC system operated 45 processing days and handled 2178 transactions. No SNM differences occurred during comparison of the physical inventory with the DYMAC general ledger nor with the laboratory-wide LASL accountability system (LAS) for nuclear materials.

As part of our ongoing effort to establish design and performance requirements for cost-effective, integrated safeguards systems for nuclear fuel cycle facilities, the report, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility," is being published as LA-7011. This conversion report is the third of a series intended to treat each of the process facility modules characteristic of a complete national fuel cycle. The other two modules, mixed-oxide fuel fabrication and fuel reprocessing, were addressed in the reports LA-6536 and LA-6881, respectively.

The pivotal role of the conversion process is underscored by development of the concept of the bonded crucial facility (BCF), wherein the conversion plant function is expanded to include storage of the product and feed inventory for the adjacent chemical separations and fuel fabrication plants, respectively. Such an expanded complex could then incorporate all inventory and control functions for the most diversion-attractive materials at a key monitoring and control point in the overall fuel cycle.

LASL is providing technical support to the DOE Alternative Fuel Cycle Technology (AFCT) programs that are being administered by the Savannah River Operations Office. The first alternative being studied as a follow-on replacement for the conversion process reported in LA-7011 is the coconversion of coprocessed, mixed uranium-plutonium nitrate solutions via the General Electric COPRECAL process. Modeling of the COPERECAL process is complete to the degree required for materials accounting. The measurements model for COPRECAL is generally similar to that employed in the conversion process. However, the material forms and compositions and the process equipment are sufficiently different to require appropriately modified measurement techniques and materials control capabilities.

Another ongoing major effort in support of AFCT programs is the design of safeguards systems for fuel cycle facilities operating under the $^{233}\text{U}/^{232}\text{Th}$ fuel cycle. The reprocessing of thorium-uranium fuels differs significantly from reprocessing in the uranium-plutonium cycle. Although the head-end chop-leach process is similar to that for LWR fuels, the dissolution of $\text{UO}_2\text{-ThO}_2$ fuels is more difficult, requiring treatment with HF. The resultant presence of fluorides must be considered in subsequent analytical procedures.

The draft report "Preliminary Concepts for Materials Measurement and Accounting in Critical Facilities," has been published and the draft of companion report, "Concepts for Inventory Verification in Critical Facilities," is nearing completion.

During this reporting period, the program of development of safeguards design and evaluation methodology has included significant contributions in the areas of modeling and simulation for evaluating safeguards system effectiveness, system

security threat evaluation, vulnerability assessment, and decision analysis applications.

In our continuing commitment to training and technology transfer, the LASL safeguards staff has trained, briefed, and transmitted technical design

information and data to representatives from various US nuclear industry and contractor facilities, as well as domestic, foreign government, and international agencies.

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 briefing, 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

A. Passive Neutron Assay of Spent PWR Fuel Assemblies (J. E. Stewart, S. T. Hsue, and K. Kaieda)

Previously we reported that the neutron emission rate of spent fuel pellets is proportional to burnup from 18 000 to 27 000 MWD/MTU.¹ Typically, safeguards inspections of spent fuel are performed on complete assemblies submerged in water. We report here on calculations that demonstrate the feasibility of correlating neutron measurements with burnup in a spent fuel assembly—in this case a pressurized water reactor (PWR) assembly stored under water. Our calculations also confirm the sensitivity of neutron measurements to the interior fuel rods of an assembly.

Two complications may arise when neutron assays are performed on whole assemblies submerged in water: neutron moderation and neutron multiplication. A Monte Carlo neutron transport code, MCNP, was used to calculate these two effects (see Pt. 1, Sec. II-D). For ease in computation, we assumed the assembly to be infinite in length and assumed that the fuel was mixed with the fuel pin cladding. These assumptions should not affect final results in an important way. The neutron sources within the fuel rods were determined from our previous calculations¹ (based on postirradiation destructive examinations), assuming a cooling time of 1 yr.

Table I shows the calculated values of (1) the multiplication factor (total neutrons per source

TABLE I
CORRELATION OF NEUTRON FLUX WITH
SOURCE STRENGTH AT VARIOUS BURNUPS

<u>Burnup</u> <u>(MWD/MTU)</u>	<u>Multiplication</u>	<u>Φ_{th}/S</u> <u>(MTU/cm²)</u>	<u>Φ_{epith}/S</u> <u>(MTU/cm²)</u>
12 859	1.57	5.52E-5	6.05E-5
15 170	1.54	5.44E-5	5.57E-5
19 208	1.52	4.89E-5	5.79E-5
20 602	1.54	5.01E-5	5.92E-5
23 557	1.49	4.94E-5	5.53E-5
25 258	1.50	4.71E-5	5.20E-5
26 884	1.51	4.89E-5	5.78E-5
S.D. (%)	1.8	6.0	5.0

neutron), (2) the thermal flux per source neutron, and (3) the epithermal flux ($0.5 \text{ eV} < E < 100 \text{ eV}$) per source neutron for a range of burnups. Neutron fluxes were calculated for a point in the midplane of the assembly, 12.5 cm normal from the surface. The table shows the system multiplication factor to be rather constant (S.D. 1.8%) for burnups ranging from 13 000 to 27 000 MWD/MTU. Columns 3 and 4 in Table I indicate that both thermal- and epithermal-neutron flux per source neutron are also fairly constant. Source neutrons have been shown to be proportional to burnup,¹ and Table I indicates that neutron flux measurements in underwater PWR assemblies can be used as a burnup monitor with an accuracy >10%.

Next we selected an assembly with medium burn-up (20 602 MWD/MTU) and calculated the flux level to be expected at various distances from the surface and corner in a spent fuel assembly. The results are shown in Figs. 1 and 2. The thermal flux is $\sim 10^5 \text{ n/cm}^2\text{-s}$ at the surface of the assembly and decreases by an order of magnitude at 7.5 cm from the surface. The flux level can be measured easily with a small fission chamber (counting rate 1000 cps).

The next set of calculations was made to determine whether the fuel-pin location within the assembly contributes to the neutron flux; in other words, how strong is the self-shielding effect and

how sensitive is a neutron detector to neutrons originating from interior rows of fuel pins?

Table II is a summary of the relative flux contribution from source neutrons started from various fuel pin rows with the average contribution normalized to 1. Row 1 represents the first row of pins from the surface of the 15 x 15 PWR assembly; the center of the assembly is a water pin; row 7 consists of pins next to the water pin. The results of Table II indicate a relatively flat contribution and 10-12.5 cm along midplane normal has the smallest standard deviation. A neutron detector located at this position detects all fuel pins about equally well.

In conclusion, Monte Carlo calculations demonstrate the feasibility of using measured neutron flux to infer burnup in PWR assemblies stored under water. The calculations also show that neutron measurements are sensitive to interior fuel pins.

B. Omega West and Big Rock Point Spent Fuel Measurements (J. R. Phillips and S. T. Hsue)

1. Omega West Reactor Exercise (J. R. Phillips, S. T. Hsue, K. Kaieda, and E. G. Medina)

Additional gamma-ray and neutron data were obtained in January 1978 to verify and extend the

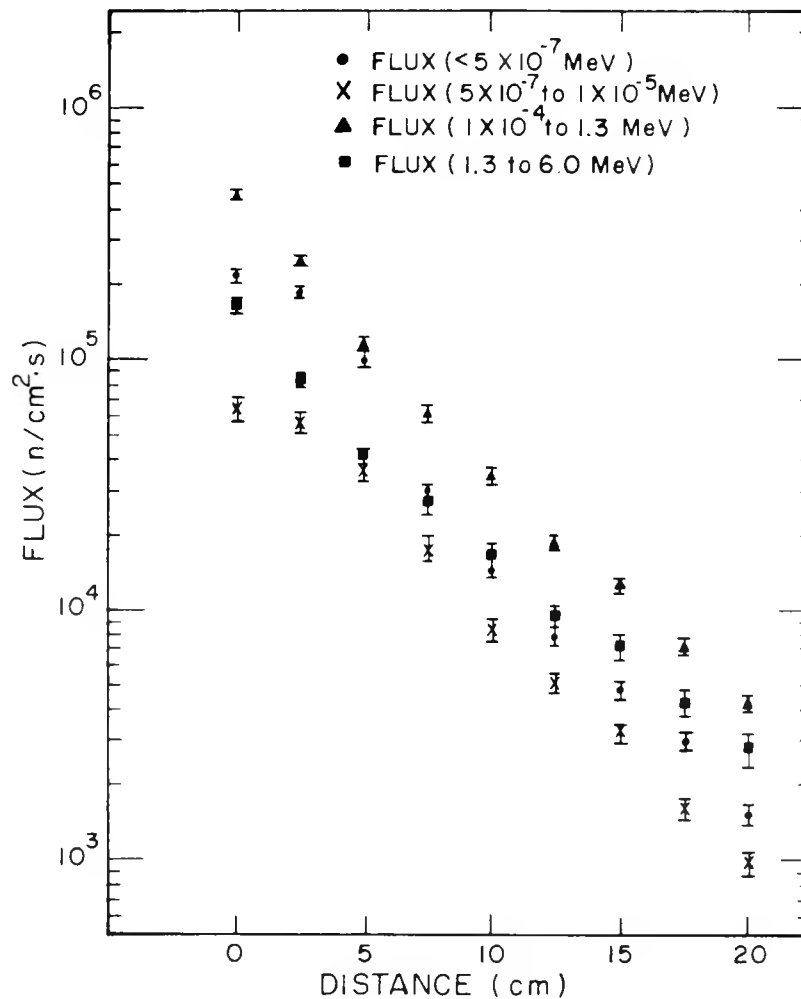


Fig. 1.

Calculated neutron fluxes along midplane normal from a PWR spent fuel assembly submerged in water.

results of experiments made during September 1977. The recent measurements included axial distribution of fission and activation products for determining burnup profiles of individual fuel elements. More than 20 isotopic ratios are being correlated with declared burnup and cooling time of irradiated materials testing reactor (MTR) fuel assemblies to determine the existence of linear and nonlinear relationships. Results of these experiments will be summarized in the next progress report and a formal report will be issued.

2. Nondestructive Measurement at Big Rock Point Nuclear Plant (J. R. Phillips, S. T. Hsue, S. E. Beach, C. R. Hatcher, D. M. Lee, E. G. Medina, and K. Kaieda)

A series of gamma-ray and neutron measurements are planned at Consumers Power Co., Big Rock Point Nuclear Plant, during May 14-24, 1978, to provide information for optimizing methods of determining cooling-time, burnup, and activity profiles for irradiated boiling water reactor (BWR)

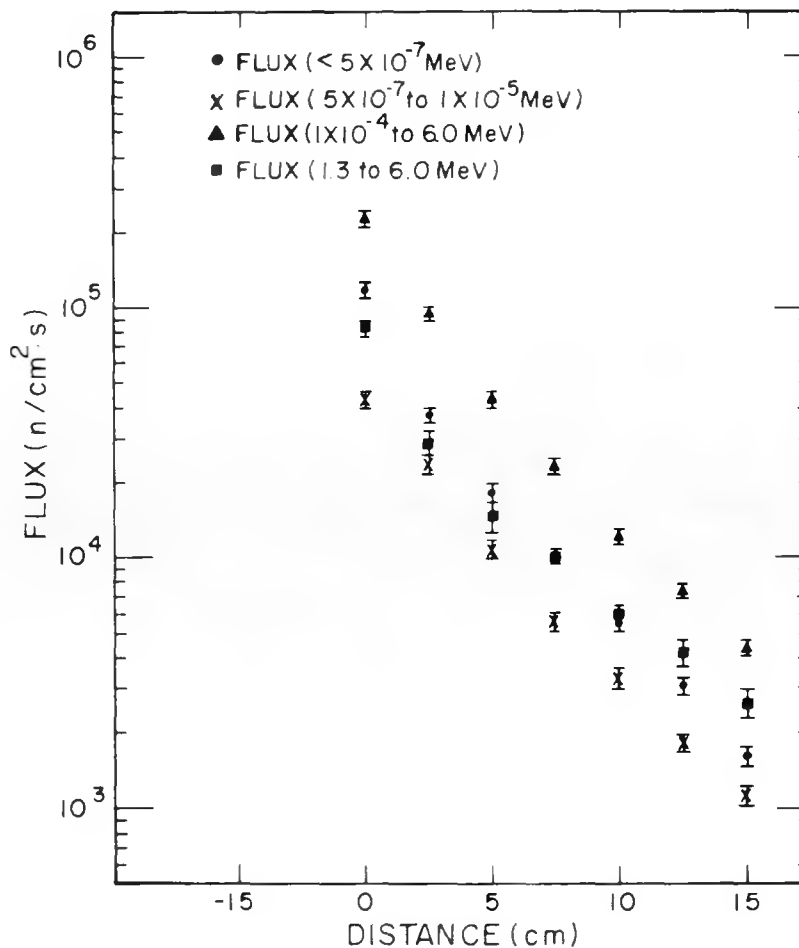


Fig. 2.

Calculated neutron fluxes along midplane diagonal from a PWR spent fuel assembly submerged in water.

fuel assemblies. Verification of the irradiation histories of fuel assemblies is essential in safeguarding storage facilities.

Negotiations with Consumers Power Co. were initiated in November 1977. A meeting was held at their facility in February 1978 to describe our proposed experimental plan, and two design reviews followed in March and April. Consumers Power personnel have been most cooperative during the preparation for this exercise.

C. Neutron and Gamma-Ray Dose Measurements for High Enrichment Spent Fuel Assemblies (G. W. Eccleston, H. O. Menlove, and E. Medina)

Information on the neutron and gamma-ray dose rates from spent fuel elements is needed for the design of the Shuffler neutron interrogation system for the Idaho Chemical Processing Plant (ICPP). This assay system will measure the ^{235}U content in

TABLE II

**RELATIVE FLUX CONTRIBUTION FROM SOURCE NEUTRONS
STARTING IN VARIOUS FUEL PIN ROWS**

Thermal Flux Along Midplane Normal

<u>Position:*</u>	<u>5</u>	<u>7.5</u>	<u>10</u>	<u>12.5</u>	<u>15</u>	<u>17.5</u>	<u>20</u>
<u>Pin Row</u>							
7	0.95	1.20	0.95	1.07	0.86	0.79	0.83
5,6	1.00	0.92	1.16	1.05	1.17	1.01	1.04
3,4	0.72	1.08	0.84	0.87	0.79	0.75	1.01
1,2	1.23	0.95	1.06	1.08	1.11	1.22	0.99
<u>1-7</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>
S.D.	0.21	0.13	0.14	0.099	0.19	0.22	0.094

Fast Flux (1×10^{-4} to 1.3 MeV) Along Midplane Normal

<u>Position:*</u>	<u>5</u>	<u>7.5</u>	<u>10</u>	<u>12.5</u>	<u>15</u>	<u>17.5</u>	<u>20</u>
<u>Pin Row</u>							
7	1.05	1.20	0.96	1.16	0.88	0.97	0.87
5,6	1.14	0.92	1.03	1.00	0.86	0.92	1.00
3,4	0.86	0.98	1.09	1.02	0.99	1.18	0.95
1,2	1.00	0.94	0.91	0.97	1.09	0.89	1.05
<u>1-7</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>
S.D.	0.12	0.13	0.079	0.084	0.11	0.13	0.068

Thermal Flux Along Midplane Diagonal

<u>Position:*</u>	<u>0</u>	<u>2.5</u>	<u>5</u>	<u>7.5</u>	<u>10</u>	<u>12.5</u>	<u>15</u>
<u>Pin Row</u>							
7	0.92	0.86	0.84	0.85	0.88	0.94	0.98
5,6	0.85	0.88	1.00	0.95	0.80	0.80	0.84
3,4	0.69	1.03	0.90	1.09	1.06	1.14	0.97
1,2	1.33	1.04	1.09	0.96	1.06	0.99	1.09
<u>1-7</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>	<u>1.00</u>
S.D.	0.29	0.097	0.11	0.098	0.13	0.14	0.10

TABLE II (cont)

Fast Flux (1×10^{-4} to 1.3 MeV) Along Midplane Diagonal							
Position:*	0	2.5	5	7.5	10	12.5	15
Pin Row							
7	0.81	0.84	0.90	0.96	0.99	0.91	0.96
5,6	0.80	0.69	0.88	0.79	0.89	0.79	0.75
3,4	0.96	1.53	0.95	0.94	1.10	1.02	1.04
1,2	1.15	0.72	1.10	1.14	0.97	1.10	1.08
1-7	1.00	1.00	1.00	1.00	1.00	1.00	1.00
S.D.	0.16	0.39	0.099	0.14	0.087	0.14	0.15

*Distance in centimeters from the surface or corner of the assembly.

waste canisters from the fluorine dissolver and also in spent fuel assemblies before dissolution. High dose rates ($\sim 10^4$ R/h) are expected from the spent fuel, and appropriate shielding is being designed into the interrogator system.

To obtain more accurate dose information, a series of measurements were performed at the spent fuel storage facility at Idaho Falls. Personnel from LASL, Allied Chemical Corp. (ACC), and Westinghouse participated in the measurements. Two types of fuel assemblies, typical of the fuel to be assayed in the neutron interrogator, were measured in the hot cell facilities at Idaho Falls.

Neutron yields and burnup profiles were measured with a modified SNAP-II detector² and

gamma-ray dosimeters. The SNAP-II detector modification involved replacing the ^3He neutron detector tubes with ^{235}U fission chambers. The fission chambers can operate in the high gamma-ray fields ($\sim 10^4$ R/h) in the hot cell near the fuel assembly. The efficiency of the SNAP-II detector with the fission chamber modification decreases by about two orders of magnitude but is not a serious problem because of the high neutron yield from the assemblies. Calibration and reduction of the data to obtain absolute neutron yields and gamma-ray doses are not completed but relative data are available.

II. NEUTRON ASSAY TECHNIQUE DEVELOPMENT

A. In-Process Measurements of Wet and Dry Plutonium Samples (N. Ensslin, C. Spirio, P. Collinsworth,* D. Bowersox,** and D. Mignogno**)

Nondestructive assays of wet plutonium oxalate cake and dry PuO_2 were carried out at the CMB-11 plutonium recovery area. The measurements were made in support of a joint CMB-11/Q-4/Q-1 study of plutonium-uranium coprocessing. The process studied here was the precipitation of plutonium out

of solution in the form of wet oxalate. The oxalate was then calcined to PuO_2 . Both the wet oxalate and the PuO_2 were assayed with a SNAP-II total neutron counter (Ref. 2, pp. 5-7) and a thermal-neutron coincidence counter. Known PuO_2 standards were used to calibrate the counters. The results were compared with the measured weight of the samples.

The SNAP-II total neutron counter is a small, portable, undermoderated detector that is relatively insensitive to self-multiplication in the sample but very sensitive to the presence of α, n reactions or moderating material. The strong neutron background in the plant also affects assay precision. When assayed, dry samples placed ~ 60 cm from the

*Group E-2.

**Group CMB-11.

SNAP-II averaged 91% of the measured weight. However, the assay values fluctuated $\pm 13\%$ compared to an expected statistical error of 2-3%; wet samples averaged 134% of measured weight and their assay values fluctuated $\pm 12\%$. The high values for the wet samples are presumed to be caused by α, n reactions and strong neutron moderation by water, which was $\sim 70\%$ of the sample weight. The assay of these materials with a portable neutron counter is not recommended.

The neutron coincidence counter used for these measurements is a well-shielded counter of $\sim 25\%$ efficiency and 70- μ s die-away time. It is mounted in one of the process lines (for these measurements it was open to the room) and the samples were assayed in plastic bags. For this experiment the ^3He tube junction box was rebuilt to permit the use of all 15 tubes. New shift-register electronics minimized the count rate corrections associated with large samples and high count rates. Also, the coincidence-counter lifting mechanism was modified to accommodate the large wet oxalate samples. With these changes, the detector response was stable within $\pm 0.2\%$.

Both the total count rate and the coincidence count rate obtained with this detector were recorded. Dry samples assayed at $110 \pm 3\%$ of the measured weight, and wet samples at $136 \pm 2\%$, using total neutron counting—results that are more precise than those obtained with the portable counter. They would yield a reasonable assay if either (1) suitable standards of wet and dry material were available, or (2) calculated or measured correction factors (such as 100/136 for the wet samples above) were applied to the data.

The assays obtained with neutron coincidence counting are listed in Table III. The dry assay averaged $94.3 \pm 1.1\%$ of the weight; the wet assay, $112 \pm 4\%$. The discrepancy for the dry weight is believed to be caused by the fact that the process material differs from the calibration standards in density, can diameter, and ^{240}Pu enrichment. Recent Monte Carlo calculations of the dependence of self-multiplication on these parameters (see Pt. 1, Sec. II-D) yield a 4% correction for these factors. The dry assay is then $98.1 \pm 1.2\%$ of the weight, essentially in agreement with the calculations. Similar calculations will be done for the wet oxalate. They should also provide good correction factors, but the $\pm 4\%$ variation on the wet assays suggests that the wet material may be less homogeneous than the dry. Under such conditions, however, coincidence counting should provide a more accurate assay than total neutron counting.

B. Neutron Assay Methods for Verification of Plutonium and Uranium in Fast Critical Facilities (H. O. Menlove, N. Ensslin, M. S. Krick, S. T. Hsue, and E. Medina)

During the past two years, we have been developing instrumentation for the NDA of fast critical assembly- (FCA-) type fuel drawers for the IAEA. This work has resulted in development of a portable high-level neutron coincidence counter (HLNCC) (Ref. 3, p. 10) being used by IAEA inspectors for the verification of FCA-type drawers. This passive neutron assay technique measures the effective

TABLE III
NEUTRON COINCIDENCE COUNTING OF WET AND DRY PLUTONIUM SAMPLES

Sample	Plutonium from PuO_2 (g)	Wet Oxalate (g)	Dry Assay (% of wt)	Wet Assay (% of wt)
1	533	1460	94.0 ± 1.2	111 ± 1
2	471	1374	94.9 ± 1.2	111 ± 1
3	480	1138	92.5 ± 1.2	106 ± 1
4	502	1644	95.0 ± 1.2	117 ± 1
5	457	1430	95.2 ± 1.2	115 ± 1
		Average	94.3 ± 1.1	112 ± 4

^{240}Pu content in the sample, and additional gamma-ray spectroscopy measurements are performed by the IAEA¹ to verify the isotopic composition. Methods are now being developed to correct for self-multiplication effects⁴ in the high-mass samples.

In addition to the passive neutron techniques, systems such as the ^{252}Cf Shuffler⁵ and the active well coincidence counter (AWCC)⁶ are being evaluated for use on FCA fuel. The ^{235}U plates and drawers containing a mixture of plutonium and uranium plates are of special interest for active neutron interrogation because the passive neutron methods are not directly applicable to ^{235}U metal.

Additional passive gamma-ray and neutron measurements have been performed to study techniques for the quantitative verification of the plutonium and uranium content in FCA-type coupons, drawers, and storage canisters. The plutonium and uranium plates used in an FCA facility generally present a favorable situation for quantitative NDA methods of verification because of their well-characterized shape, isotopic composition, and packaging. In evaluating the measurement approaches, we normally can assume that the composition and configuration of the material is given either by the facility operator or by previous measurements on the same sample. The role of the NDA instrumentation is to give a quantitative verification that the sample contains the expected loading of plutonium or ^{235}U .

1. Passive Neutron Verification

Measurements were made in two thermal-neutron coincidence counters; the dual range counter⁷ in its high-efficiency mode and the AWCC in its passive mode with shift-register coincidence counting electronics (see Ref. 7, pp. 4-6).

The dual range counter was placed on its side for the measurement of fuel drawers. The fuel canister does not fit inside the sample cavity of the dual-range counter and therefore was measured in the larger AWCC operated in the passive mode. A coincidence counter designed for permanent installation for the assay of plutonium fuel drawers would differ in three ways from the dual range counter used for the present measurements: (1) The sample cavity would be smaller in diameter and longer to provide a more uniform response along the length of the

drawer; (2) the detector would be designed for higher efficiency (~40% compared to the present 27%); and (3) the outer shielding would be increased to reduce room background.

The measurements were done with 15 FCA fuel plates varying from 2.54 to 17.18 cm in length (26 to 200 g of plutonium). The isotopic composition was either 11.6 or 8.7% ^{240}Pu . All fuel plates were plutonium-uranium-molybdenum alloy; all were 5 cm deep and most were 0.64 cm thick.

Individual coupons with 11.6% ^{240}Pu were counted in the dual range counter. The totals counting rate with a 64- μs coincidence gate was 290 cps/g ^{240}Pu ; the coincidence counting rate was 39 cps/g ^{240}Pu . Therefore, a fuel drawer containing 1 kg of plutonium at 11.6% ^{240}Pu has a total counting rate of 3.4×10^4 cps and coincidence rate of 4.5×10^3 cps. For a 100-s measurement the standard deviation on the totals count attributable to counting statistics only will be 0.05% and the standard deviation on the coincidence count attributable to counting statistics will be 0.9%. For the coincidence counter with 40% efficiency, the standard deviation for the coincidence counts will decrease to 0.5% for 100-s measurement. The removal of a 2.54-cm-long plate from the drawer will result in a 3% reduction in the counting rate and would be easily detected.

The plutonium coupons in a fuel drawer can be arranged to increase the totals and coincidence counting rates by placing two or more plates side by side to increase the neutron multiplication. Consequently, some fuel coupons can be removed from a drawer and the remaining fuel rearranged to produce the original totals or coincidence counting rates. However, the original totals and the original coincidence rates cannot be produced simultaneously.

Multiplication in FCA drawers was studied by stacking from 1 to 11 fuel plates in the center of the dual range counter. Plots of the totals and coincidence rates per gram of ^{240}Pu (effective) are shown in Figs. 3 and 4, respectively. These rates corrected for multiplication are also shown. The corrected coincidence rates vary <10% between a single coupon and 11 coupons, whereas the uncorrected coincidence rate varies by 60%.

Passive neutron assay of FCA drawers can be based on totals counting or coincidence counting.

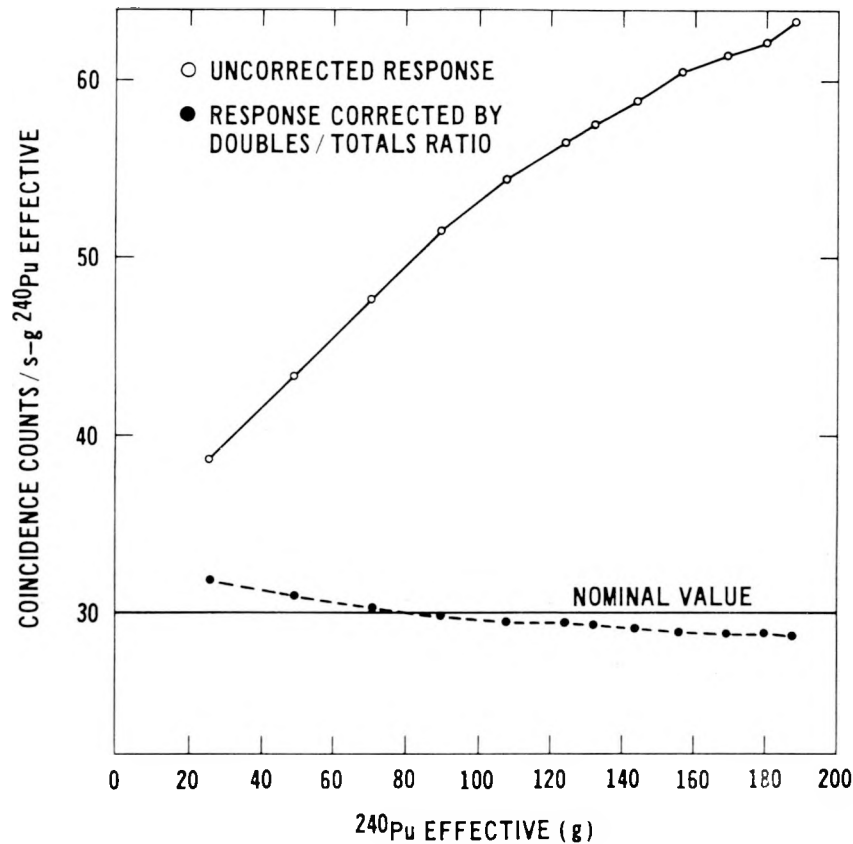


Fig. 3.

Neutron coincidence counts per gram as a function of plutonium loading for stacked plates. The curve would be a horizontal line if there were no multiplication or absorption effects.

One advantage of totals counting is the simpler electronic circuitry required for the assay. Another advantage is that self-multiplication affects the totals count rate much less than it affects the coincidence count rate. The difference for plutonium is about a factor of 3.

One disadvantage of assays based on total neutron counts is that separate measurements are required for a thorough knowledge of the background. Often the room background varies during the measurements, depending on the location of other material or of people in the room and on the configuration of the detector itself.

Any α, n reactions present will contribute a strong background originating from the sample. This background affects the total count rate much more than it affects the coincidence count rate. An assay based on totals would not be useful unless the ratio of α, n reactions to spontaneous fission neutrons was

precisely fixed and the room background remained constant during the measurement period.

The discussion of the coincidence and totals counting of fuel drawers applies also to plate storage canisters, except that (1) a different-size sample chamber is required for the canisters and (2) multiplication effects are smaller in a normally configured fuel drawer than in a filled canister. In general, there is more plutonium in the canisters than in the drawers, and a higher counting rate and better statistical precision result. However, multiplication effects will be larger in the canisters. To the first order, multiplication increases the sensitivity to plate removals; that is, a 10% reduction in plutonium reduces the counting rate by >10%. Geometric variations might overshadow this statistical advantage.

For a partially filled canister, one cannot rearrange the loading of the coupons to adjust both

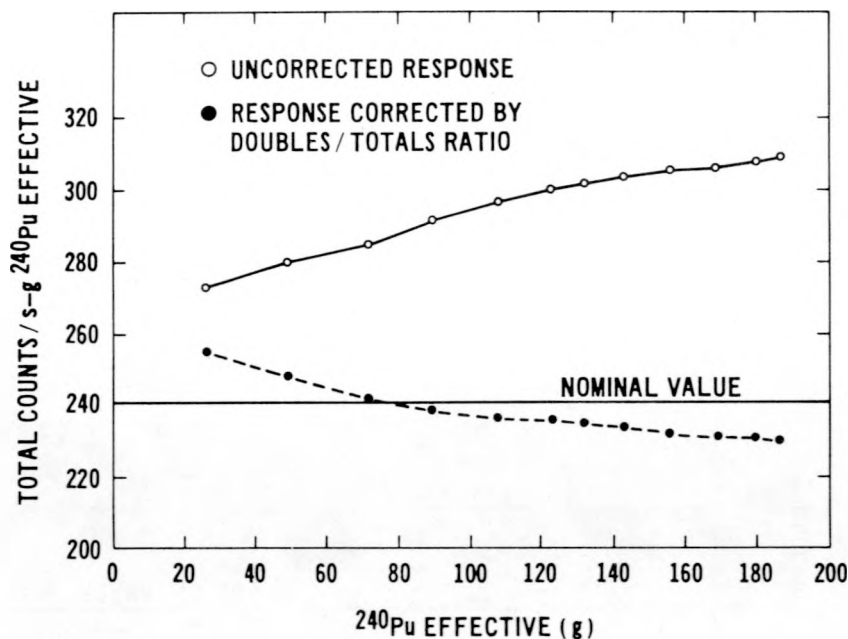


Fig. 4.

Total neutron counting rate per gram as a function of plutonium loading for stacked plates.

the coincident and total counting rates so as to disguise missing coupons.

To investigate absorption and multiplication effects in a storage canister, we made a series of measurements in the AWCC in the passive mode. The coupons were placed in the canister one at a time and the individual results were summed to correspond to the higher loadings. In addition, the higher loadings were obtained by putting groups of coupons in the counter side by side in alternating rows (every other row). The results of these measurements are given in Fig. 5. A small multiplication effect in the coincidence results amounts to ~10% for the 750-g plutonium loading and would be considerably larger for the fully loaded canister.

We recommend that the passive neutron assay of fuel drawers and canisters be based on coincidence counting to eliminate background problems and that the total neutron count rate also be recorded. The ratio of coincidences/totals can then be used to correct for self-multiplication. By measuring both the coincidence and total count rates and by using the information that can be extracted from a comparison of the two rates, one can be almost certain of detecting any combination of coupon removal, rearrangement, or substitution.

Passive gamma-ray and neutron coincidence methods are adequate for the rapid quantitative verification of plutonium in FCA fuel. For alloyed FCA fuels with a significant α, n yield, such as those containing aluminum, the combination of the neutron total and coincidence counting gives a check of the expected plutonium isotopic mixture. When ^{235}U coupons are present, the ^{252}Cf Shuffler system would be the most promising active neutron method. In the combination active-passive mode, the Shuffler can be used for the verification of both uranium and plutonium coupons.

The NDA measurement should be sensitive to the following cases:

- Removal of one or more coupons from the container or the substitution of coupons containing only a portion of the original loading (such as the top edge).
- Substitution of high ^{240}Pu content coupons for low ^{240}Pu coupons.
- Substitution of radioactive gamma-ray and neutron sources for the plutonium in the container.
- Change of geometric configuration within the container to give a higher counting efficiency

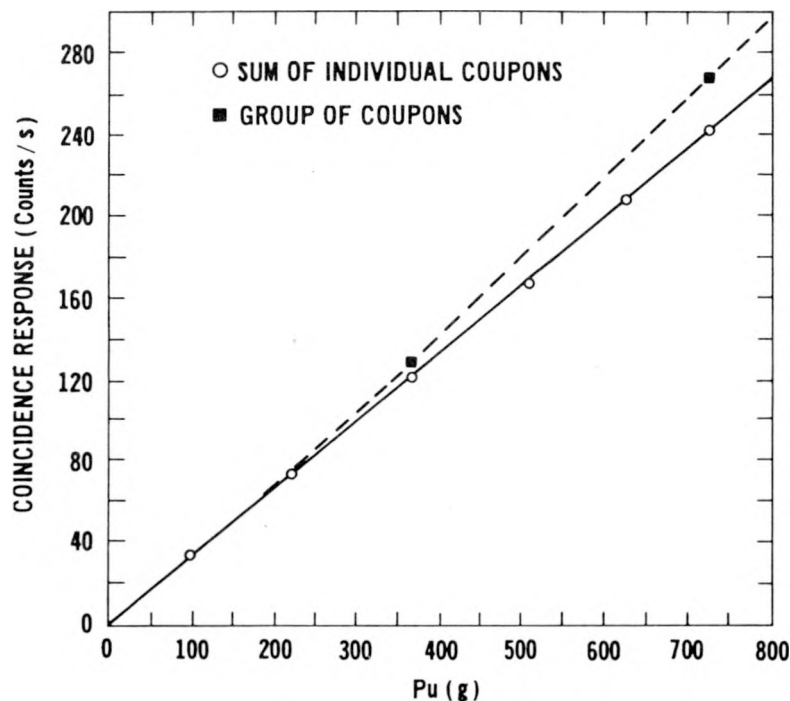


Fig. 5.
Coincidence response vs plutonium (8.7% ^{240}Pu) loading for coupons in a storage canister.

or multiplication effect, thereby masking the removal of part of the plutonium.

The N verification should give a qualitative measure of the special nuclear material (SNM) for the expected configuration of the material. However, if SNM removal has been masked by any of the above cases, the instrumentation should signal this irregularity. The passive neutron counting takes care of items (a), (c), and (d) and the gamma-ray verification deals with items (b), (c), and (d). Thus, the combination of passive neutron and gamma-ray techniques (possible in the same instrument) will give a high level of confidence in the verification. For plutonium fuels alloyed with aluminum, the α, n passive neutron yield makes the neutron counting sensitive to item (b) also.

C. Self-Multiplication Correction Factors for Neutron Coincidence Counting (N. Ensslin)

Neutron coincidence counting of large plutonium oxide or metal samples is strongly affected by self-multiplication within the sample. Experimental

data and Monte Carlo calculations on 800 g of PuO_2 show a 35% increase in the coincidence response.¹ For 1- to 2-kg plutonium metal buttons, the response may be increased by a factor of 2 or more. Self-multiplication is manifested in the following ways:

- Fissions induced by neutrons from spontaneous fission will increase the observed spontaneous fission multiplicity ν_s to $M\nu_s$, where M is the leakage multiplication. That statement is valid if these induced fissions occur on a time scale short compared to the coincidence resolving time. This should apply to thermal-neutron detectors but perhaps not to fast coincidence detectors such as the random driver.
- Fission induced by neutrons produced in α, n reactions will increase the coincidence response. These fissions are not correlated in time to spontaneous fission events and will show a separate induced fission multiplicity $M\nu_i$.

For in-plant assay of large plutonium samples, self-multiplication effects must be corrected for. Here we describe simple correction factors for neutron coincidence counting that are based on the

principle that the ratios between single, double, or triple events recorded by the coincidence circuitry contain information on the effective fission multiplicity $M\nu_s$. For the specific cases described below, multiplicity $M\nu_s$ can be estimated from the data without additional circuitry.

1. Application of Double/Singles Ratio to Pure Metal

For pure metal, all neutrons must come from spontaneous fission events or background. For a very small sample (no multiplication):

$$\text{Singles rate } S_o = mG\epsilon\nu_s + B,$$

$$\text{Doubles rate } D_o = mG\epsilon^2 F\nu_s(\nu_s - 1)/2.$$

For a larger sample (with multiplication):

$$\text{Singles rate } S_M = mG\epsilon(M\nu_s) + B,$$

$$\text{Doubles rate } D_M = mG\epsilon^2 F(M\nu_s)(M\nu_s - 1)/2.$$

Here m = mass of sample in grams ^{240}Pu effective,

G = fission/second-gram in ^{240}Pu

ϵ = detector efficiency

B = room background

F = fraction of coincidences seen by coincidence circuit.

Solving for M yields

$$M = \frac{1}{\nu_s} + \frac{\nu_s - 1}{\nu_s} R \quad (1)$$

with

$$R = \frac{D_M/(S_M - B)}{D_o/(S_o - B)}.$$

To correct the measured doubles for self-multiplication, use the relation

$$\begin{aligned} D_{\text{corrected}} &= D_{\text{measured}} / \{ [M\nu_s(M\nu_s - 1)] / [\nu_s(\nu_s - 1)] \}. \end{aligned}$$

Substituting for M yields the correction factor

$$D_{\text{corrected}} = \frac{D_{\text{measured}}}{M \cdot R}. \quad (2)$$

Equation (2) involves no arbitrary constants. It requires that the background be known and fixed (no α, n reactions in the sample). The ratio of doubles/singles for a small, nonmultiplying sample must be measured. Then the doubles/singles ratio observed during the assay of the unknown sample is used to calculate R and M .

An application of Eq. (2) to plutonium fuel coupons is shown in Figs. 3 and 4. The detector was a thermal-neutron coincidence counter. The attached shift-register coincidence circuit actually measures coincidences of all orders, but most are doubles. Deviation of the corrected response from the nominal value may be due to the approximations in the equations or to efficiency variations in the well counter.

A further application of Eq. (2) with the same detector is illustrated in Fig. 6. There two batches of plutonium metal plates were moved progressively closer together. The corrected response is flat within statistical errors, implying that the assay of plutonium metal in *any* geometrical configuration can be corrected for the self-multiplication produced by that configuration. No calibration curve, parameterization of the coincidence response, or Monte Carlo calculations can allow for an arbitrary geometry. However, the observed multiplicity $M\nu_s$ can provide the information for such a correction.

2. Application of Double/Singles Ratio for Fixed α, n

For material such as PuO_2 where the number of α, n emitters is fixed relative to ^{240}Pu , one can derive new versions of the above equations. For simplicity, define $\alpha = N_{\alpha n}/N_{\text{SF}}$, the ratio of (α, n) -produced neutrons to spontaneous fission neutrons, and let the ratio of (α, n) -induced coincidence counts to spontaneous fission-induced coincidence counts be

$$\beta = \frac{D_{\alpha n}}{D_{\text{SF}}} = \frac{G_I M \nu_I (M \nu_I - 1)}{G M \nu_s (M \nu_s - 1)},$$

where G_I is the number of (α, n) -induced fission/s-g.

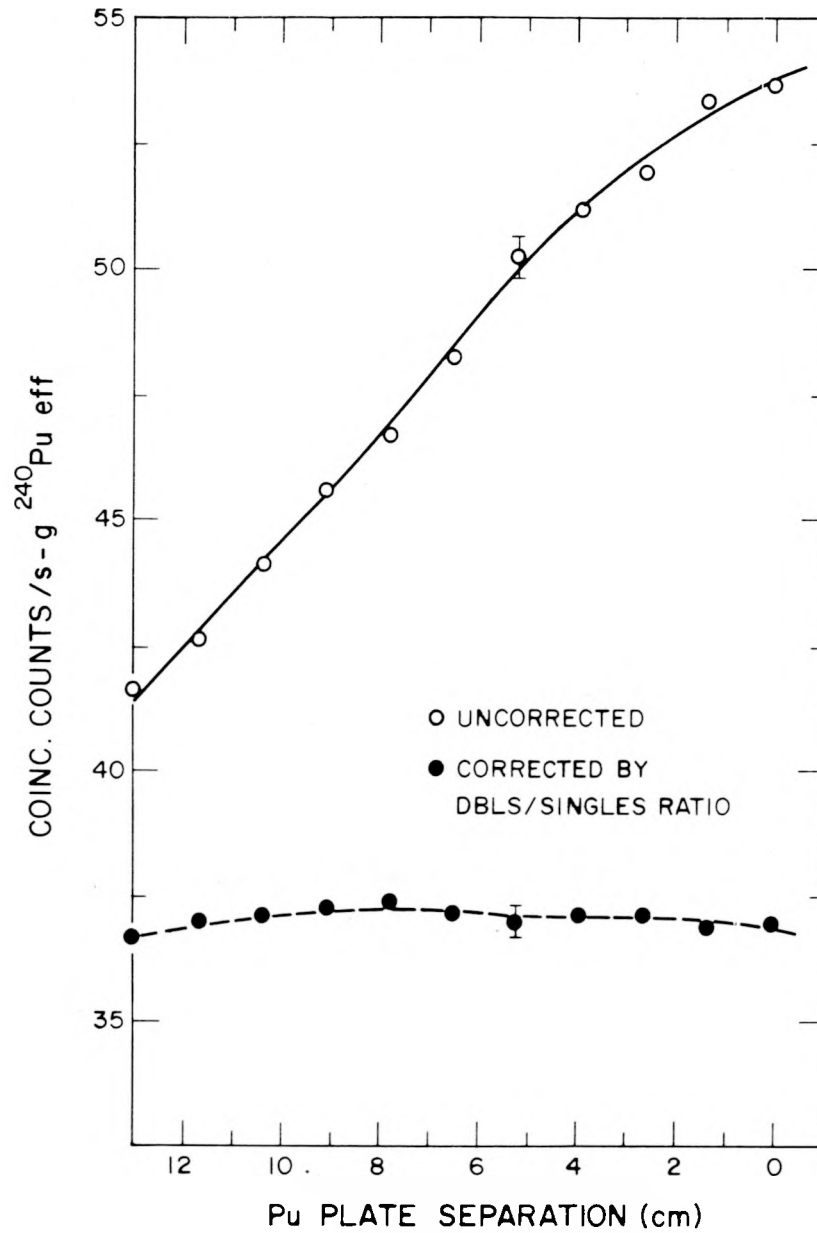


Fig. 6.
Application of self-multiplication correction to 2 kg of plutonium metal plates. As the plates are moved closer together, the uncorrected response per gram increases markedly.

Let p_f be the probability that one neutron from a spontaneous fission or from an α, n reaction will induce a fission. In general, p_f will be different for these two classes of neutrons; for PuO_2 , however, p_f is nearly the same for spontaneous fission and α, n neutrons. Then it can be shown that

$$M = \frac{1}{1 - p_f(\nu_I - 1)}$$

and

$$\beta = \alpha \left(\frac{M - 1}{M} \right) \frac{\nu_I}{\nu_I - 1} \frac{M\nu_I - 1}{M\nu_S - 1} .$$

Equation (3) shows that for PuO_2 only α is fixed; β is a function of M . In other words, the geometric factors that determine self-multiplication of spontaneous fission neutrons also determine (α, n) -induced multiplication.

The singles and doubles count rates are given by

$$S_o = N_{SF} + N_{an} = mG\epsilon\nu_S (1 + \alpha),$$

$$D_o = mG\epsilon^2 F\nu_S(\nu_S - 1)/2,$$

$$S_M = mG\epsilon\Lambda_S M(1 + \alpha),$$

$$D_M = mG\epsilon^2 F(M\nu_S) (M\nu_S - 1) (1 + \beta)/2.$$

Now

$$M = \frac{1}{\nu_S} + \frac{\nu_S - 1}{\nu_S} \frac{R}{1 + \beta} \quad (4)$$

Eliminating β between Eqs. (3) and (4) leads to a quadratic equation for M . With this value for M the correction factor for self-multiplication is again given by Eq. (2). To use this correction, measured values of unmultiplied doubles/singles, background, and α are needed.

The application of this correction factor to PuO_2 samples in a thermal-neutron counter is shown in Fig. 7. Alpha was determined by measurement and by calculation, with both methods giving values between 0.60 and 0.66. The corrected response is about four times better than the uncorrected, but the results are less clear-cut than in the case of pure metal. Poor values for the background and for the value of unmultiplied doubles/singles were used in the interpretation of these data. More data are needed to determine how useful this correction factor will be for PuO_2 .

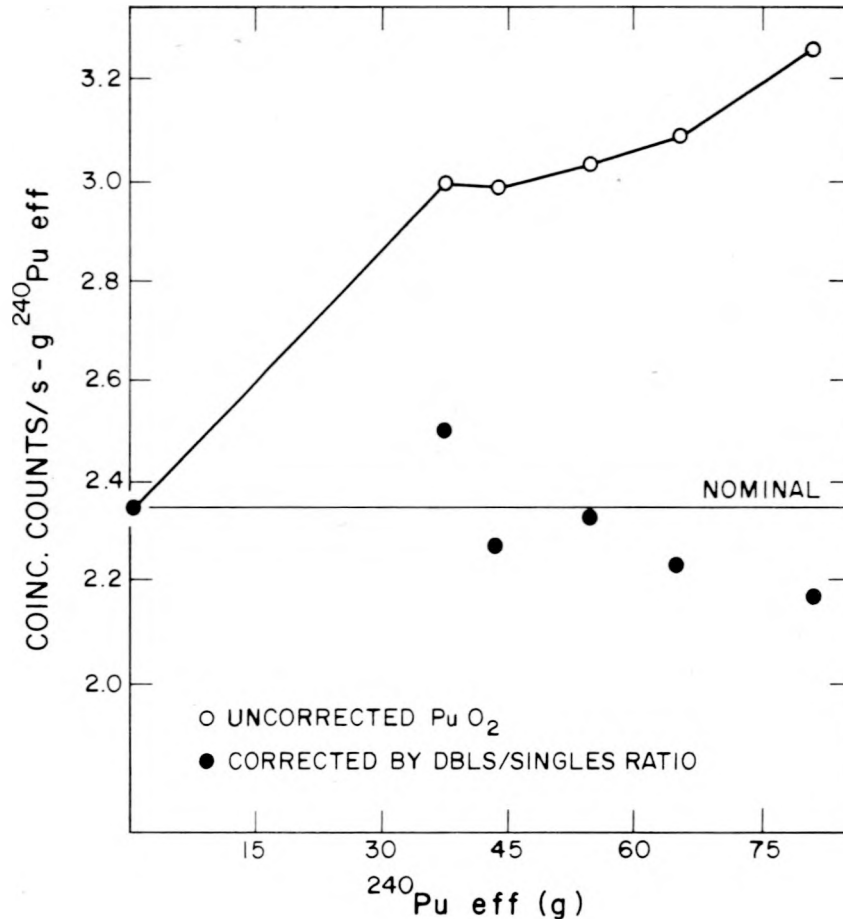


Fig. 7.
Application of self-multiplication correction to PuO_2 .

3. Application of Triples/Doubles Ratio to Pure Metal

The random driver built for the DYMAC program⁸ is a fast-neutron coincidence counter with three scintillators. Single, double, and triple events are recorded. Thus a multiplication correction for pure metal can be based on the ratio of triples/doubles, which are virtually background-free because of the short coincidence resolving time. The count rates (T =triples) are given by

$$D_o = mG\epsilon^2 F\nu_s(\nu_s - 1)/2,$$

$$T_o = mG\epsilon^2 F'\nu_s(\nu_s - 1)(\nu_s - 2)/6,$$

$$D_M = mG\epsilon^2 FM\nu_s(M\nu_s - 1)/2,$$

$$T_M = mG\epsilon^3 F'M\nu_s(M\nu_s - 1)(M\nu_s - 2)/6.$$

Then

$$M = \frac{2}{\nu_s} + \frac{\nu_s - 2}{\nu_s} R,$$

with

$$R = \frac{T_M/D_M}{T_o/D_o}.$$

The correction factor is

$$D_{\text{corrected}} = \frac{D_{\text{measured}}}{M \left[\frac{1}{\nu_s - 1} + \frac{\nu_s - 2}{\nu_s - 1} R \right]}.$$

This correction factor was applied to metal buttons assayed in the random driver, as illustrated in Figs. 16 and 17 of Ref. 8. The correction was 1.40 and 2.10 for 500- and 1000-g buttons, respectively. With this correction the assay was within 5% of the

nominal passive response for nonmultiplying material.

4. Application of Triples/Doubles Ratio to Fixed α, n

These equations have not yet been derived; however, the correction for pure metal described in the preceding section was applied to PuO_2 in Ref. 8, Figs. 16 and 17. With the correction, 20-35% self-multiplication effects were reduced to <5%. This implies that the fixed α, n equations would be useful and should be derived.

5. Conclusion

The correction factors derived should be useful in removing the effects of self-multiplication on neutron coincidence counting. Further work is needed on the application of the doubles/singles ratio to fast-neutron coincidence counters. Also, small nonmultiplying standards of PuO_2 are needed so that better measurements can be made.

Several important assumptions are made in the derivation of these simple corrections. One is that the detector efficiency is constant over the volume of the sample, and this usually can be realized in practice. Another assumption is that all events in the fission chain occur on a time scale small compared to the detector response. The validity of this assumption for fast coincidence systems (≤ 40 -ns resolving time) needs to be examined. The most serious simplification is the use of an effective average multiplicity $M\nu$ rather than the actual multiplicity and discrete leakage multiplication distributions. Large errors may be introduced, but they tend to cancel when ratios are used. Folding approximations and first and second moments of the distributions will be used to improve the correction factors. Future refinements in the multiplication correction factors will significantly reduce the errors encountered in the assay of large mass plutonium samples.

D. Monte Carlo Calculations of Self-Multiplication Effects in PuO₂ (N. Ensslin and J. Stewart*)

Calculations of self-multiplication effects in PuO₂ are being carried out by Group TD-6 in support of a variety of measurement problems involving the use of thermal-neutron coincidence counters to assay large PuO₂ samples. The calculations employ a recent version of the Monte Carlo code MCNP and represent an extension of the procedure outlined in Ref. 1, p. 14. The new code has been modified by Robert Schrandt, Group TD-6, to yield the discrete net leakage multiplication probabilities, and these are now calculated separately for spontaneous fission- and alpha-induced neutrons. On the basis of present results, a separate calculation probably is not necessary except for samples where the α, n contribution is larger than it is in pure PuO₂. Both multiplication calculations also allow for reflection of neutrons from the surrounding well counter, thereby yielding the net leakage from the sample. A separate computer code is then used to fold the spontaneous fission multiplicity distribution with the spontaneous fission discrete net leakage multiplication distribution. A correction factor F_{sf} results. This separate code also computes a correction factor $F_{\alpha n}$ using the alpha-induced net leakage multiplication distribution. The result is a correction factor of the form

$$F = 1.0 + F_{sf} + F_{\alpha n}$$

where F is the ratio of the coincidence response per gram for a multiplying sample to that for a sample whose multiplication is 1.

Figure 8 illustrates a series of calculations of the coincidence-counting correction factor for PuO₂ for varying mass, density, enrichment, and sample diameter. All calculations represent variations about a nominal case of 800 g PuO₂ at 1.3 g/cm³, containing 706 g Pu at 10% ²⁴⁰Pu (effective) and 1% water by weight, in a container of 8.35-cm i.d. The nominal calculation is shown as a solid point in Figs. 8a-d.

Figures 8 a, b, and d illustrate that self-multiplication corrections are substantial even in

small or low-density PuO₂ samples. Further work will be done on the effect of water on self-multiplication, in support of recent measurements of wet plutonium oxalate precipitate. J. Brandenberger, Group Q-1, is calculating the effect of 70-80% water on the α, n source strength used to determine $F_{\alpha n}$.

E. Calculation of (α, n) Neutron Production in Materials Containing Alpha Emitters (J. D. Brandenberger)

Neutron detection methods are frequently used to perform NDA of substances containing SNM. Because these substances are often light elements in the presence of alpha-particle emitters, there may be production of neutrons by α, n reactions. A quantitative knowledge of neutrons from α, n reactions for various materials is useful in the design of assay instruments, the analysis and interpretation of assay data, and the design of the standards to which the assays are normalized. An accurate and easy-to-use method of making the calculations is developed. These results, combined with the recent α, n data compilation of Liskien and Paulsen⁹ and α, n measurements being made at Oak Ridge, Harwell, Geel, and elsewhere should meet all of our requirements for the calculation of α, n neutron production.

Only homogeneous materials such as compounds, solutions, and fine-grind mixtures are considered here. A fine-grind mixture is defined as "well mixed" and containing particles sufficiently small that an alpha particle will lose only a small fraction of its energy in passing through any single particle. In a fine-grind, particles containing heavy elements must be $<1 \mu\text{m}$ in length.

Consider a homogeneous material that contains n elements (or isotopes for alpha emitters). One subset of n consists of ℓ alpha-emitting isotopes and another consists of m (α, n) neutron-emitting elements. For some of the constituents we may consider all of the isotopes of an element as a single species denoted by a single value of the index n . For this case in which there are $n = \text{isotopes (or ele-)}$

*Group TD-6

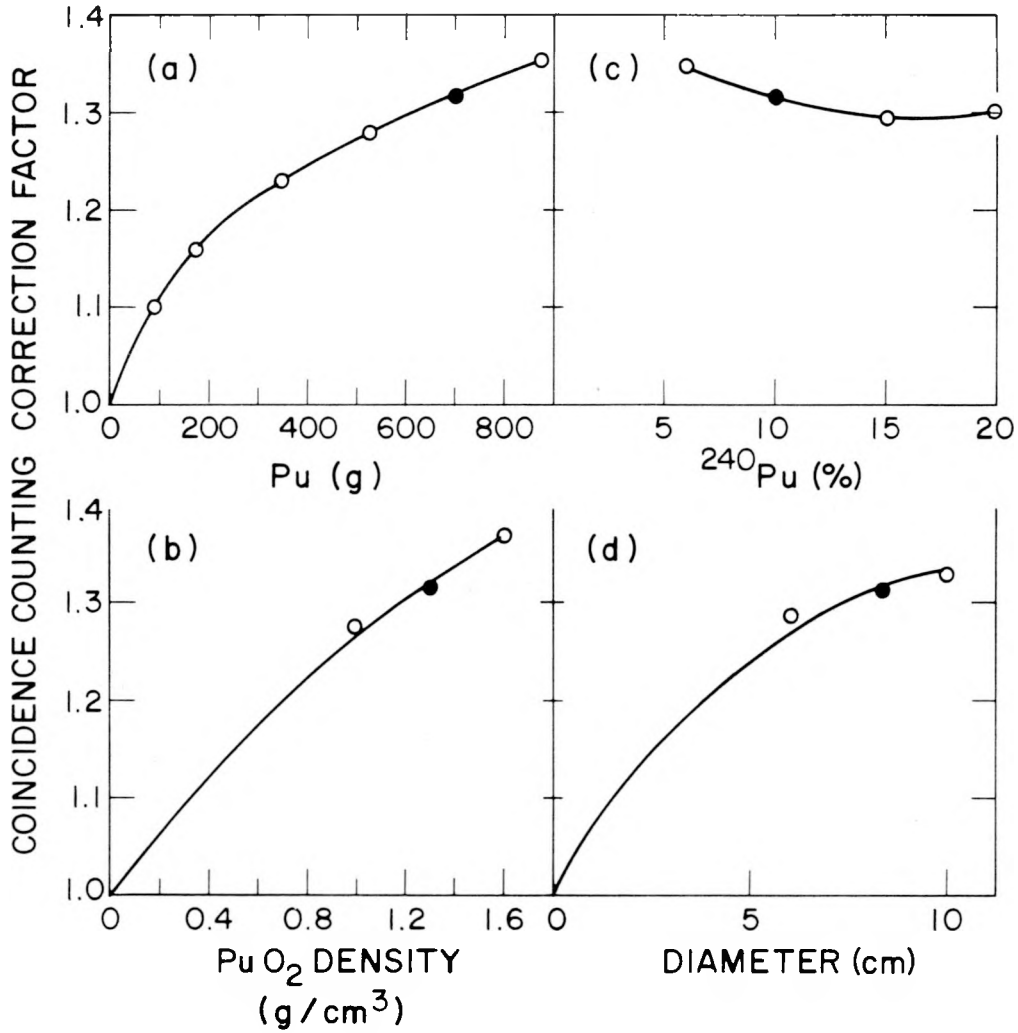


Fig. 8.
Monte Carlo calculations of self-multiplication effects of various parameters on coincidence counting of PuO₂. The solid data point denotes the nominal calculation for each case.

ments), ℓ = alpha-emitting isotopes, and $m = \alpha, n$ neutron emitters, the total number of α, n neutrons emitted per gram (or per cm³, per sample, etc.) in 1 s is

$$Y = \sum_{k=1}^{k=\ell} Y_K \int_{E_\alpha=0}^{E_\alpha^{K\max}} \frac{\sum_{j=1}^{j=m} \epsilon_j(E_\alpha) N_j \left(\frac{dY_j^{\alpha n}(E_\alpha)}{dE_\alpha} \right) dE_\alpha}{\sum_{i=1}^{i=n} \epsilon_i(E_\alpha) N_i} \quad (5)$$

Here,

Y_K is the number of alpha particles emitted per gram, per cm³, per sample, etc., by the Kth α emitter,

$Y_j^{\alpha, n}(E_\alpha)$ is the thick-target, α, n neutron yield of the jth element bombarded by an alpha particle of energy E_α ,

N_i, N_j are the number of ith or jth species per gram (or cm³, molecule, etc.), and

$\epsilon_j(E_\alpha), \epsilon_i(E_\alpha)$ are the stopping cross sections of the jth or ith elements (or isotopes).

The $Y_{j,n}^{\alpha}(E_{\alpha})$ are so small for $E_{\alpha} < 1$ MeV that it is sufficient to evaluate the integral from $E = 1$ MeV to $E = E_{\alpha}^{\text{Kmax}}$, where the latter is the energy of the alpha particles emitted by the Kth isotope. For safeguards SNM, E_{α}^{Kmax} lies between 4 and 6 MeV.

To evaluate Eq. (5), the stopping cross sections $\epsilon_1(E_{\alpha})$ must be determined. Attempts to fit the stopping cross-section data are conspicuous for their success for $E_{\alpha} \geq 4$ MeV, and conspicuous for their failure as E_{α} decreases in energy below 4 MeV (Ref. 10).

My approach was to construct a simple expression which fits the stopping cross-section data for the limited energy range $1.0 \leq E_{\alpha} \leq 6.0$ MeV. Although the fit is weighted to the higher energies (which produce most of the neutrons), even at $E_{\alpha} = 1.0$ MeV, the expression for $\epsilon_1(E_{\alpha})$ fits the data to within a few percent for all Z.

Interactive Focal* and RT-11 Basic programs to calculate α,n neutron production by Eq. (5) have been written. Data to determine the accuracy of the calculations are scarce. Two types of data are available: (1) neutron sources based on α,n reactions and (2) measurements of neutron production of materials such as UO_2 and PuO_2 which are bombarded with monoenergetic alpha particles.^{9,11} The first type consists of sources whose source strengths have been normalized to secondary standards measured at the National Bureau of Standards (NBS). Sources of this type, such as $^{241}\text{AmO}_2\text{-Li}$ sources, are well-suited for testing calculated α,n source strengths because they are truly independent of data input into Eq. (5). The second type gives a consistency test only, because the α,n neutron production from alpha-particle bombardment of UO_2 and PuO_2 is compared to calculated values based on the α,n measurements.⁹

Results of test calculations are given in Table IV. The second column gives data for a radioactive source (MRC-72) and the third and fourth columns show results for thick targets bombarded with alpha particles of 5.5 and 4.4 MeV energy, respectively, from a Van de Graaff.

The results for $^{241}\text{AmO}_2\text{-Li}$ indicate that the calculations are accurate within $\sim 10\%$ for SNM containing lithium. This accuracy is better than that of the α,n neutron production data which have an evaluated uncertainty of 25-30%,⁹ so at least part

TABLE IV
RESULTS OF TEST CALCULATIONS

Method of Calculation	$^{241}\text{AmO}_2\text{-Li}$ (MRC-72)	$^{239}\text{PuO}_2$ (g ⁻¹ -s ⁻¹)	UO_2 (g ⁻¹ -s ⁻¹)
Calculation by Eq. (5)	$6.7 \times 10^6 \text{ s}^{-1}$	8331	5.6×10^{-4}
"Experimental" value	$6.2 \times 10^6 \text{ s}^{-1}$	8676	5.2×10^{-4}
Relative difference (%)	-8.1	+3.9	-7.7

of the α,n data may be more accurate than previously believed. Recent unpublished data from Oak Ridge National Laboratory (ORNL), received by private communication from J. K. Bair, have a claimed accuracy of 5-10%. These data differ from the compilation of Liskien and Paulsen by 9% and 36%, respectively, for two elements.^{9,11} Results must thus be viewed cautiously until confidence is built in particular parts of the α,n data base. One conspicuous gap in data for safeguards NDA is $F(\alpha,n)$; however, measurements have been made on fluorine at ORNL and the data are being reduced.¹¹

Some of our SNM neutron sources have proved unsuitable for calculating values for α,n neutron production. For example, the NDA working standards made of PuO_2 from sample number CMB-11-FM-JOX-03907 (standards 3907-1 through 3907-6) are unsatisfactory in two respects. First, they contain light-element contaminants in such quantity that they may dominate over the α,n neutron production from the PuO_2 . Second the standards that are mixtures— PuO_2 mixed with aluminum, MgO , silicon, and boron, respectively—contain particles so coarse as to make calculations under the fine-grind assumption inapplicable. For meaningful comparison between measured and calculated strengths of neutron sources, care must be exercised in the design, fabrication, and documentation of the neutron sources.

F. Active Well Coincidence Counter (H. O. Menlove, N. Ensslin, and E. Medina)

The passive neutron well coincidence counter has proven to be one of the most useful NDA instruments for plutonium assay. IAEA inspectors have

*Trademark of the Digital Equipment Corp.

found the portable high-level neutron coincidence counter (HLNCC) unit particularly useful for field applications. However, the instrument has not been applicable to the assay of ^{235}U or ^{233}U because of their extremely low spontaneous fission yields. To make the instrument applicable to the two uranium isotopes, we are working on a combination of a small AmLi neutron interrogation source and a ^3He thermal-neutron well coincidence counter.

Also under development is an active well coincidence counter⁶ (AWCC) that will be more portable, lightweight, stable, and less subject to effects of matrix materials and gamma-ray backgrounds than are conventional fast random-driver assay systems. The last feature makes the AWCC applicable to ^{233}U -Th cycle materials that generally have high gamma-ray backgrounds from the decay of ^{232}U . The counter uses an AmLi source of low intensity ($\sim 5 \times 10^4$ n/s) to interrogate the assay sample in a ^3He well counter that detects coincidence neutrons from the induced-fission reactions.

Figures 9 and 10 show the recently fabricated prototype AWCC. Its design is optimized for counting the induced-fission reactions and discriminating against the lower energy AmLi interrogation background neutrons. The sample cavity is lined with a nickel reflector to give a more penetrating neutron interrogation. The CH_2 moderator and cadmium sleeves are designed to give maximum efficiency in counting the induced-fission spectrum neutrons while giving a low efficiency in counting the α, n neutron from the AmLi interrogation source.

A series of measurements were performed to determine the optimum source intensity and to evaluate the effectiveness of the different internal sleeves of cadmium, nickel, and CH_2 in reducing the assay error. The sample used for the evaluation was a polyethylene bottle containing 200 g of uranium (93% ^{235}U -enriched) mixed with graphite powder. Three different AmLi neutron interrogation sources with intensities of 2.4×10^4 , 1.2×10^5 , and 5×10^5 n/s were intercompared.

The detector's efficiency in counting fission source neutrons was measured with a calibrated ^{252}Cf source. The absolute efficiency varied from 20-30%, depending on the detector configuration. In general, the higher the efficiency, the lower the

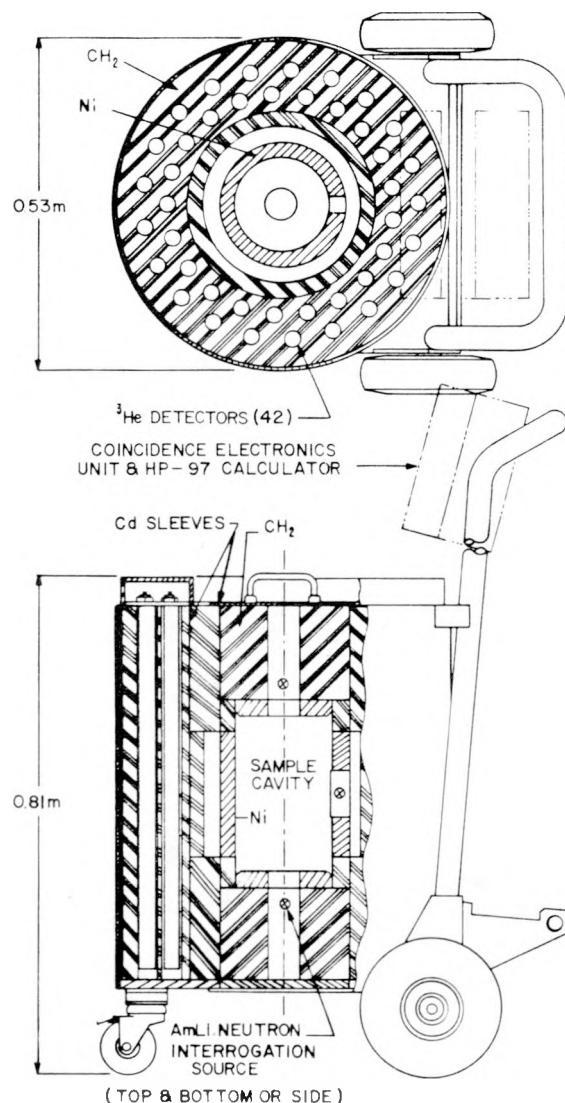


Fig. 9.
Laboratory prototype of AWCC used for the assay of uranium and plutonium.

assay error,⁶ as might be expected because the error E for a given counting time is

$$E \sim \frac{\epsilon_\alpha}{(\epsilon_f)^2},$$

where ϵ_f is the efficiency in counting fission spectrum neutrons and ϵ_α is the efficiency in counting AmLi interrogation neutrons. E_α varied from 12-24% when the source was positioned in the center of



Fig. 10.
Detector body and transport cart for the
laboratory prototype AWCC for the assay of
 ^{235}U and ^{233}U .

the counting chamber and decreased by a factor of 5 when the source was returned to its normal shielded position in the end plugs (see Fig. 9). This factor-of-5 reduction in ϵ_a , and consequently in the assay error E , is the key parameter in the application of the ^3He well counter to the active interrogation mode.

The interrogation source strength intercomparison shows that the error does not change going from 5×10^6 n/s to 1.2×10^6 n/s but that there is a factor-of-1.4-error increase in going from 1.2×10^5 to 2.4×10^4 n/s. For the high-efficiency counter shown in Fig. 9, the optimum source

strength is $\sim 1 \times 10^6$ n/s. That value is somewhat lower for less efficient counters such as the HLNCC.

The cadmium sleeve placed between the ^3He detector rings and the interior of the counter improved the assay error only slightly. The improvement in relative error when going from no cadmium, to cadmium near the ends, to cadmium covering the entire sleeve was 1:1.06:1.10, respectively. The role of the cadmium is to improve the shielding for AmLi and thus decrease ϵ_a ; however, it also decreases ϵ_r by a small amount and thus the error proportional to ϵ_a/ϵ_r^2 changes only slightly. For each of the above cases, the sample chamber was lined with cadmium to avoid thermal-neutron interrogation and the resulting loss of penetrability. When thermal neutrons are used in the assay, the signal rate increases by a factor of 25 because of the high fission cross section of ^{235}U for thermal neutrons. This thermal-neutron mode might be useful in the assay of low-enrichment uranium or some categories of scrap and waste.

The measured neutron die-away time in the detector was $\sim 50 \mu\text{s}$; consequently, a coincidence gate setting of $64 \mu\text{s}$ gives the smallest error. A gate setting of $32 \mu\text{s}$ increases the error by $\sim 10\%$.

The 2.54-cm-thick nickel liner affects the counting statistical error as well as the response linearity as a function of sample mass. The nickel liner, which acts as a fast-neutron reflector for the AmLi interrogation neutrons reduces the statistical error by $\sim 10\%$. To evaluate the usefulness of the nickel in improving response linearity, we counted uranium (93% enriched) metal disks in the center of the AWCC. The disks were stacked on top of each other to obtain the higher mass values; thus the self-shielding of the neutrons increased as the stack thickness increased. The neutron source was placed in both the top and bottom plugs to make negligible the geometric effect of the stack height. Figure 11 shows the normalized response for the cases with and without the nickel liner. The response for the nickel case is more linear but the problem is not severe for either case. The nickel liner results in better performance, but it is not clear that the improvement merits the extra cost and weight of nickel for a portable detector unit.

Figure 9 shows AmLi sources located in both the top and bottom end plugs to obtain more uniform

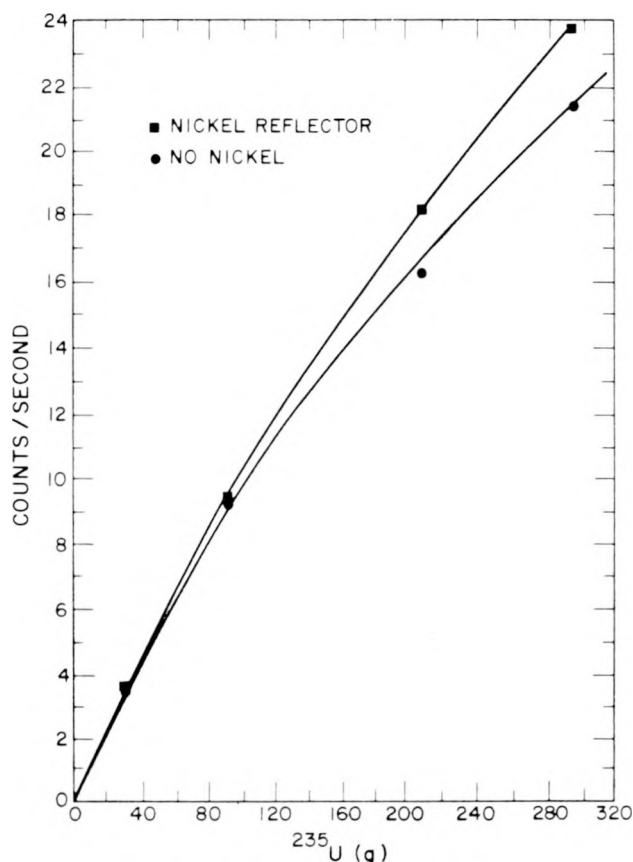


Fig. 11.
Comparison of response linearity for 2.54-cm-thick nickel liner vs CH_2 liner.

response for all locations within the sample chamber. This uniformity is helped by the fact that the efficiency for counting the induced-fission neutrons is greatest in the central section of the well counter. To measure the spatial response uniformity, a flat disk sample containing 300 g of ^{235}U was measured at increments from the bottom to the top of the chamber, with the AmLi source positioned in both the bottom and top plugs. The induced response vs height given in Fig. 12 shows the response to be uniform ($\pm 2\%$) for sample heights between 2 and 24 cm. For a single source located in the bottom, the response changes by more than a factor of 3 over the same height range.

The performance information obtained from this prototype AWCC is being used to design a simplified version of the AWCC for use by the IAEA.

For portable applications, the unit should be lightweight, rugged, and stable over wide temperature variations. The first portable version will be a prototype unit using the same electronics package used by the HLNCC (Ref. 8, pp. 18-20).

G. Enrichment Assay Applications for the Shuffler (T. W. Crane)

The ^{252}Cf Shuffler is an NDA instrument designed to measure the fissile content of a sample by neutron interrogation and delayed neutron counting. A small portion of the delayed neutron signal is due to fissions in the nonfissile isotopes.⁵ By analyzing the delayed neutron kinetics, a measure of the enrichment is possible.¹²

Calculations were performed to investigate the feasibility of using the Shuffler designed for the Savannah River Plant (SRP) test and evaluation program (Ref. 8, pp. 3-5) separating the delayed neutron contributions of ^{235}U and ^{238}U isotopes. The calculations indicate that the error in the individual isotopes is ~ 8.4 times the error in the ^{235}U mass claimed by assuming that the isotopic mixture is fixed. Because the isotopes have similar delayed neutron kinetics for the Shuffler's duty cycle, the individual isotope mass measurement errors have this large increase in their uncertainty.

The uncertainty in the mass measurement affects the accuracy with which the enrichment can be determined. The relative error of the enrichment (in percentage units) divided by the relative error in the response is shown in Fig. 13. The data in Fig. 13 show that the enrichment can be measured more accurately for low-enrichment samples (3-6%) than for highly enriched samples. For example, assuming a 0.2% relative error in the delayed neutron response, the enrichment can be measured with a relative error of about $\pm 10\%$ at 20% enrichment and with a relative error of about $\pm 29\%$ at 80% enrichment. Neither of these measurements would justify the use of the Shuffler as an enrichment meter, but the enrichment measurements made by the Shuffler could be useful either for correcting for small biases associated with the enrichment or for spotting material-type substitutions.

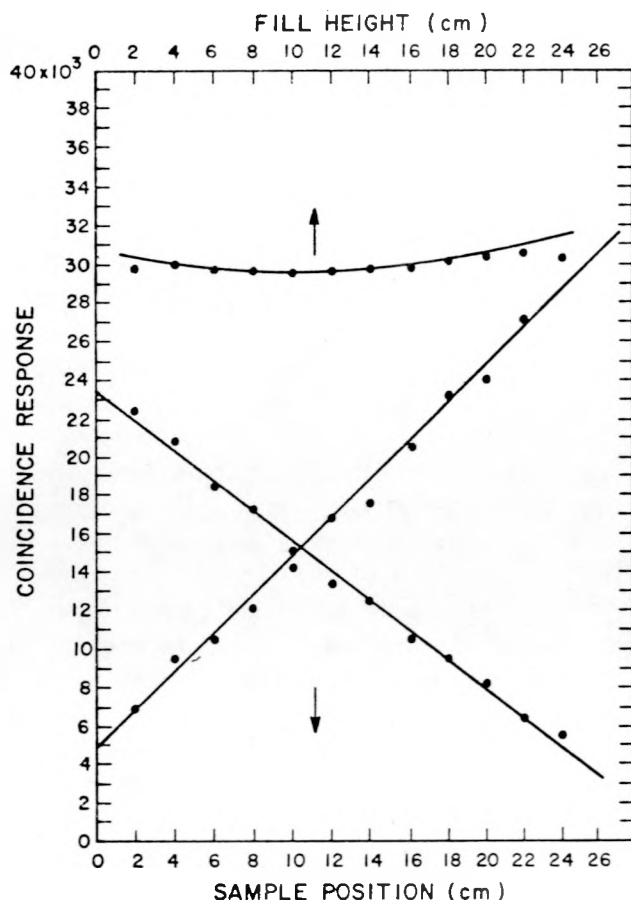


Fig. 12.

Coincidence response vs sample fill height for the AWCC using single source (bottom curves) and double source (top curve).

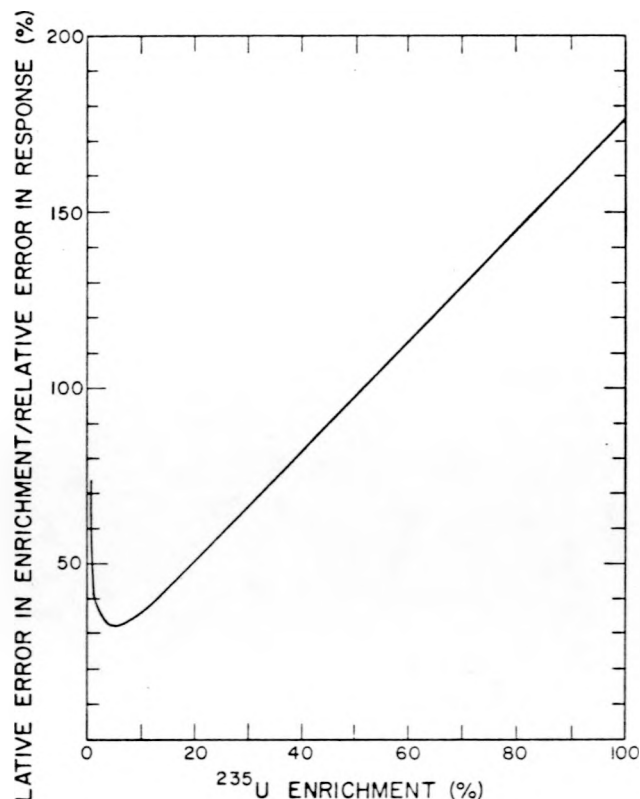


Fig. 13.

Relative error in enrichment divided by relative response error determined by delayed neutron kinetics using the SRP Shuffler system.

III. GAMMA-RAY ASSAY TECHNIQUE DEVELOPMENT

A. Construction of a Wavelength-Dispersive Filter for Application to Densitometry and XRF Studies of Hot Solutions (J. W. Tape and L. R. Cowder)

A preliminary study on the use of a Bragg-reflection wavelength dispersive spectrometer as a narrow band-pass filter for low-energy photons has been reported previously.¹ The device used in those studies was salvaged from an electron microprobe and employed a small 3.8 cm x 1.3 cm LiF crystal and a precision mechanism for positioning the

crystal and detector with respect to the source point. A Bragg crystal spectrometer used in the filtering mode does not require a precision positioning mechanism because the angles used for a particular application are fixed and the slits are relatively wide. It should, however, employ the largest practical crystal to improve the efficiency of the device.

A simple prototype Bragg filter (Fig. 14) has been constructed using a 5.7 x 2.5 cm LiF crystal with a 28-cm radius of curvature. It is designed for testing purposes only and does not use the thick shielding required for actual measurement of hot solutions.

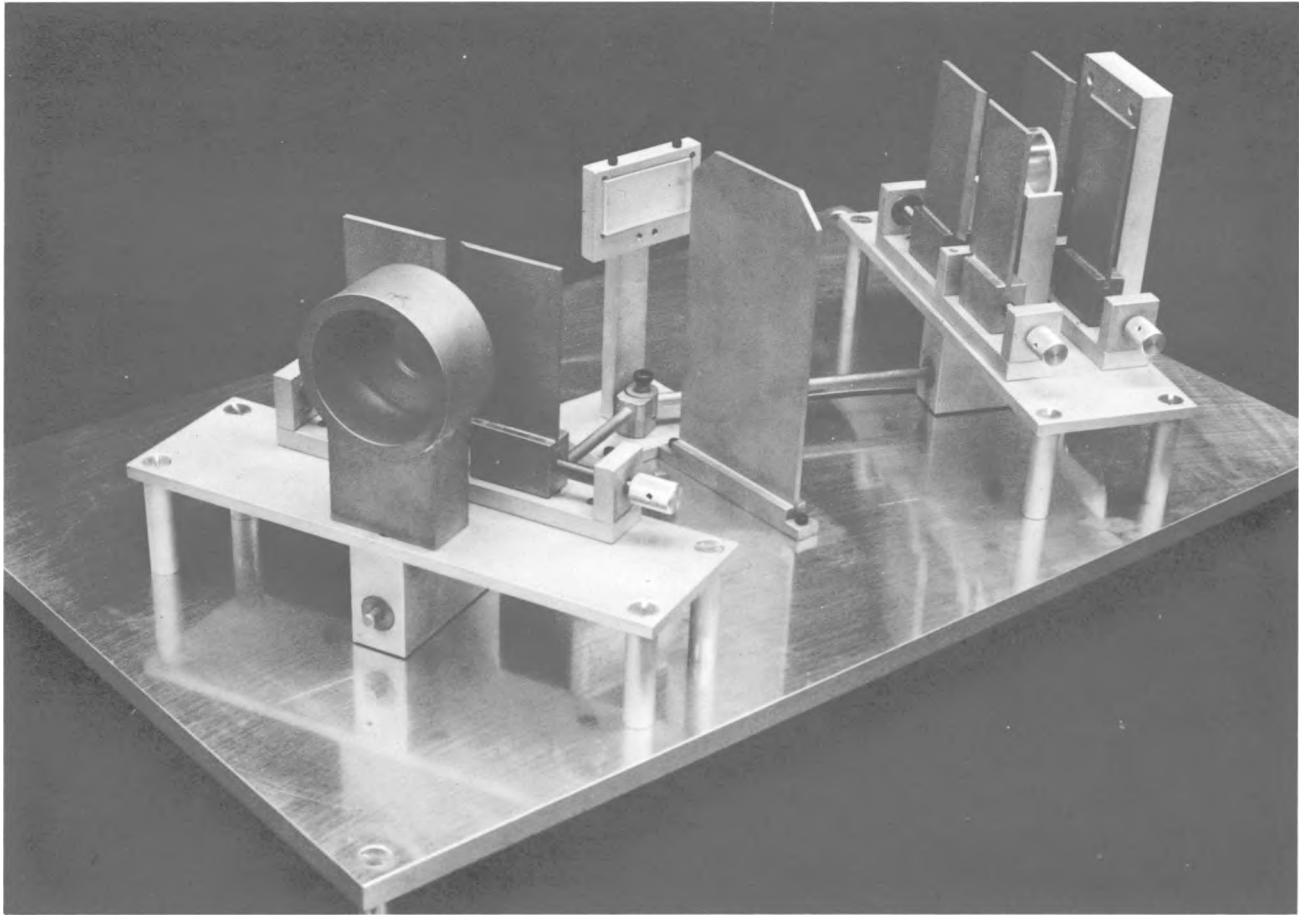


Fig. 14.
Prototype Bragg filter.

At a particular Bragg angle, the conditions for focus are that the source slits, the crystal, and the detector slits lie on the 28-cm-radius focal circle (Rowland circle) and that the source-crystal and the detector-crystal distance be equal. In this design the crystal position is fixed with crystal curvature tangent to a pair of arc-shaped grooves of 28-cm radius of curvature. The filter is set up to pass a particular energy (wavelength) band near 17 keV (the L x-ray region of SNM) by fixing the detector and source slits at the appropriate angle and distances and adjusting the width of the slits for the desired wavelength band width.

The source slit arrangement seen in Fig. 14 is designed for densitometry applications with a small x-ray generator. By suitable rearrangement of the source geometry, the filter can also be used in conjunction with x-ray fluorescence (XRF) methods.

Preliminary tests with this system indicate that the larger LiF crystal performs adequately for the proposed application.

B. Gamma-Ray Assay in Fast Critical Assembly Fuel Drawers (S. T. Hsue)

Passive gamma-ray techniques are being evaluated for the quantitative verification of plutonium in FCA fuel drawers. The plutonium content of fuel drawers can be assayed by using an enrichment type of measurement. The ^{239}Pu and ^{241}Pu content of the entire drawer and the isotopic ratio of the two can be measured simultaneously. The counting rate is proportional to the product of the fraction of plutonium isotope to the heavy metal (enrichment) and the surface area of the fuel plates

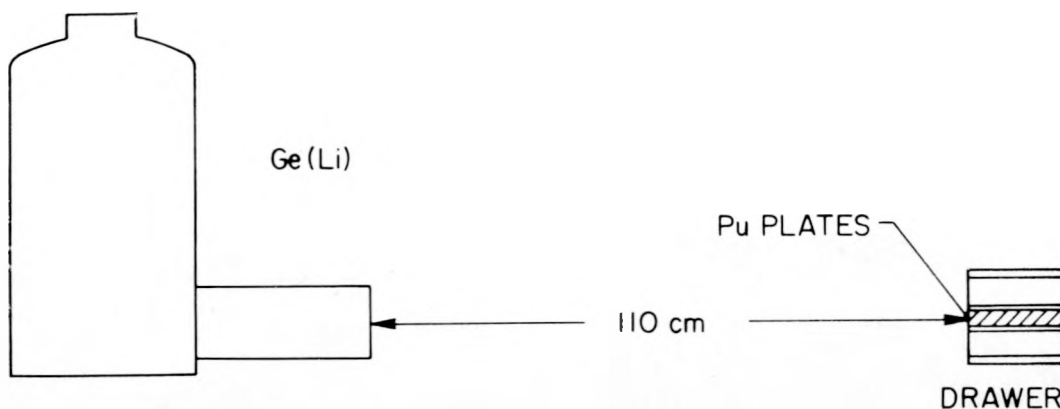


Fig. 15.
Experimental configuration for gamma-ray measurements on an FCA fuel drawer.

viewed by the detector. Moreover, if the plutonium fraction, the type of alloy, and the widths of the fuel plates are the same, the counting rate is proportional to the total length of fuel plates in the drawer.

The plutonium was assayed in the experimental setup shown in Fig. 15. The drawer is placed sideways so that the edges of the plates face the detector. The detector-to-drawer distance should be sufficiently large so that the $1/r^2$ variation across the drawer is negligible.

The following data were taken with a Ge(Li) detector (efficiency, 16%; surface area, 18 cm²) placed 110 cm from a drawer containing plutonium fuel plates. Figure 16 shows the relative counting rate of the ²³⁹Pu 414-keV gamma line vs the total ²³⁹Pu content of the drawer. The ²³⁹Pu content in the drawer is proportional to the counting rate. The 414-keV gamma peak yields the most precise determination of ²³⁹Pu content; the 208-keV gamma peak yields the most precise determination of ²⁴¹Pu content. For a drawer containing two rows of plutonium plates, the precision at the 1- σ level for a 200-s assay is 1.5% for ²³⁹Pu and 1.0% for ²⁴¹Pu. With some optimization of the detector size and detector-to-drawer distance, the assay time should be reduced by a factor of 2 without changing the precisions.

In this type of measurement, the counting rate is proportional to the product of the enrichment and the total exposed area of the plate edges. If the drawer contains fuel plates of different enrichments,

the gamma-emission rate alone cannot uniquely determine the total length of the plutonium fuel plates. However, if the plutonium enrichment is the same among the various fuel plates in the drawer, the counting rate is proportional to the total length of fuel plate. Also, the 208-keV gamma ray arises not only from ²⁴¹Pu decay but also from ²⁴¹Am decay. For an equal amount for ²⁴¹Pu and ²⁴¹Am (14.35 yr after the separation of americium), the americium decay contributes ~3.8% of the 208-keV gamma peak. In addition, the plutonium mass fractions of the different fuel plates should be similar. If these assumptions are satisfied and the ²⁴¹Am buildup is known, the plutonium content of the drawer can be measured by gamma rays.

The plutonium fuel drawers and certain types of storage canisters can be assayed rapidly by gamma measurements. The main advantage of the gamma assay is that it uniquely identifies the fissile isotopes and can be used to determine the isotopic ratios and, with proper calibration, the isotopic content of the fuel containers. The unique gamma signature of various plutonium isotopes makes plate substitution difficult. However, the fuel plates are dense and the gamma rays give only a verification of the enrichment of the outer surface on the top edge of the plates. Investigations are needed to determine the optimum assay geometry and detector size for gamma measurements of critical facility fuels.

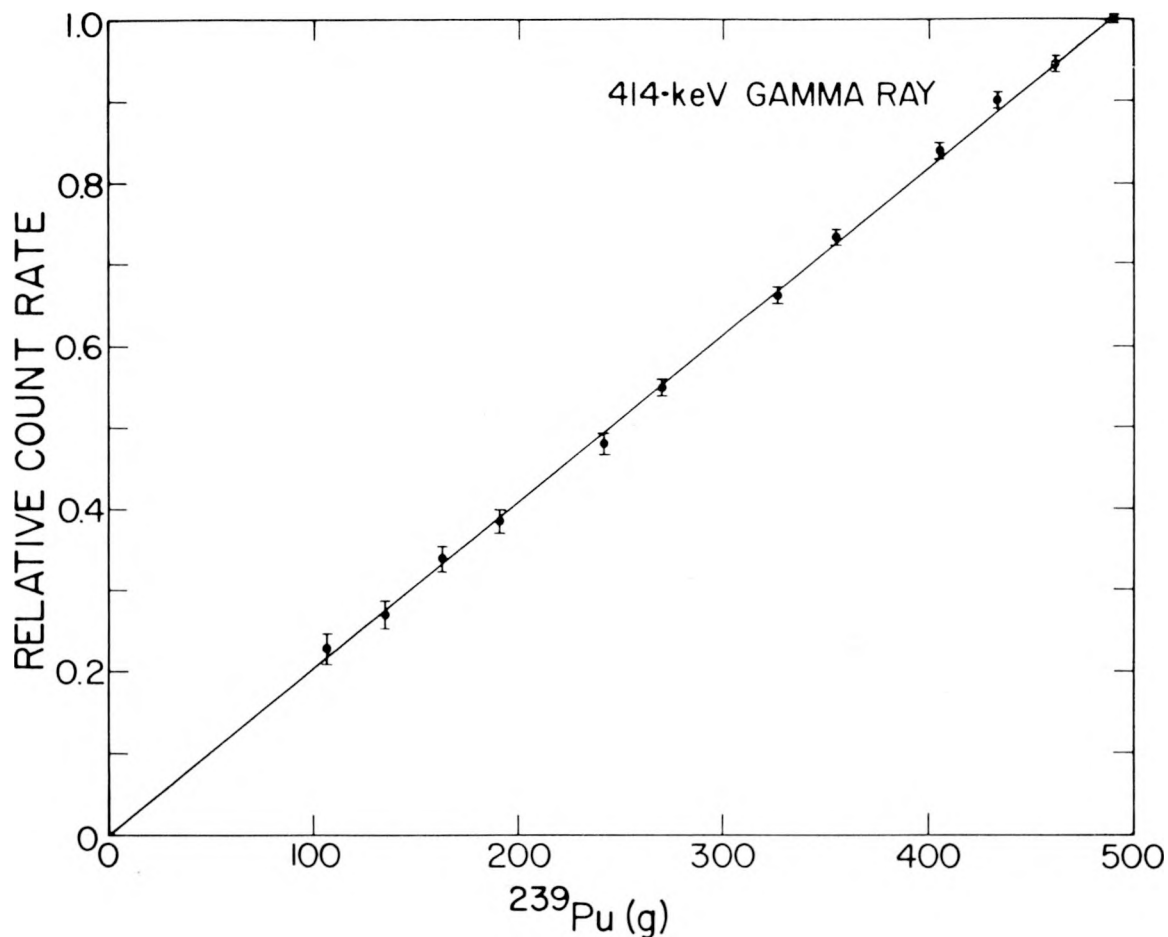


Fig. 16.

Relative counting rate for the 414-keV gamma line as a function of plutonium loading in the drawer.

IV. FIELD TESTS AND EVALUATIONS

A. Random Driver for DYMAC (D. M. Lee, N. Ensslin, T. Van Lyssel, C. Shonrock, and R. Marshall)

A random driver intended for the assay of plutonium has been installed in the TA-55 plutonium facility at LASL. The device has been described in Ref. 8. Response curves for height variation were measured before the device was installed. Because the random driver is run in both the passive and active modes, two separate curves were obtained. The height response for the passive assay is shown in Fig. 17. A 485-g PuO_2 standard, 10

cm high, was moved vertically through the sample well in 2.54-cm steps. The passive response was, as expected, a geometrical effect, due to the variation in the solid angle of the standard with respect to the three scintillators. The height response for the active-passive assay is shown in Fig. 18. The procedure used here is the same as in the passive measurement except that the sources were in place. The response was made almost independent of height by adjusting the separation of the two AmLi sources. Both height responses, when integrated over typical sample heights, are so nearly constant that no fill-height correction is required.

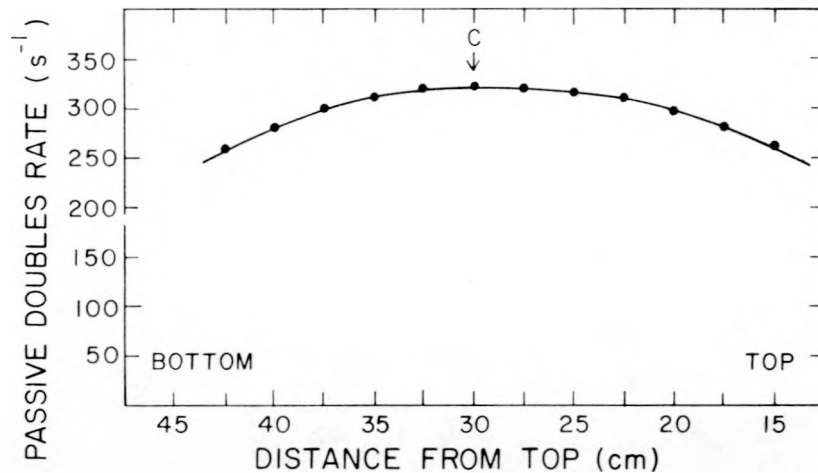


Fig. 17.

Vertical efficiency profile for the passive doubles counting of plutonium in the DYMAC random driver.

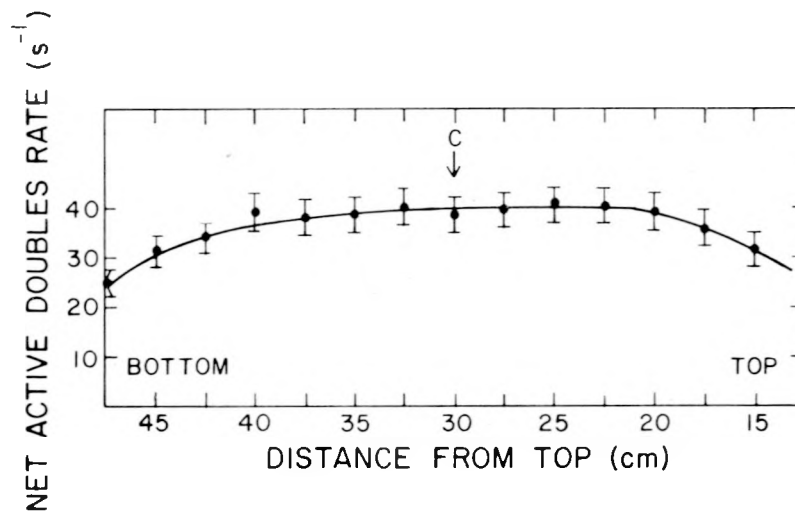


Fig. 18.

Vertical efficiency profile (doubles rate) for the active-passive counting of plutonium in the DYMAC random driver.

The computer program to control the random driver is now complete and debugged. It incorporates corrections for temperature response, neutron-flux thermalization, and leakage multiplication. The program gives the assay as the total number of grams of ^{240}Pu from the passive assay (or ^{239}Pu from the active-passive assay) and the total

number of grams of plutonium (if the enrichment is given). All raw data plus corrections are recorded so that a complete record can be maintained.

The installation of the random driver took ~ 1 wk, and the device is now being calibrated. The formal test and evaluation phase will begin in June 1978.

B. Fabrication and Testing of an L-Edge Densitometer for Assay of Mixed SNM Solutions (T. Canada, L. Cowder, S. T. Hsue, D. Langner, P. Russo, C. Spirio, J. Tape, and T. Van Lyssel)

1. Introduction

The L-edge densitometer described here provides a simple, rapid NDA alternative to costly and time-consuming routine chemical analysis for total elemental content in solution. The instrument was designed to measure multiple concentrations of SNM in solution and was built to determine the range of applicability of L-edge densitometry to the simultaneous assay of uranium and plutonium. Its particular application is in the assay of mixed SNM streams generated in coprocessing demonstrations at Savannah River Laboratory (SRL).

The densitometer, recently calibrated at LASL, was shipped on May 15, 1978, for installation, test, and evaluation at SRL. It will be used in the 1BP, 2BP, and 1EU lines at SRL where the uranium-to-plutonium concentration ratios fall between 4:1 and 10:1 with $2 \text{ g/l} \leq \rho_{\text{Pu}} \leq 10 \text{ g/l}$. Initial on-site testing of the instrument will involve the 1EU (uranium only) line with $70 \text{ g/l} \leq \rho_{\text{U}} \leq 90 \text{ g/l}$.

Here we summarize the measurement concepts of the L-edge densitometer, the hardware and software features of the instrument design, the test and evaluation procedures, and the results of the calibrations performed at LASL.

2. Concepts of L-Edge Densitometry Measurements

An assay that uses L_{III} densitometry and a continuous x-ray source is performed by comparing the sample-attenuated x-ray spectrum with the unattenuated spectrum (reference spectrum obtained with solvent only). The resulting transmissions just below and above the L_{III} absorption edge are, respectively,

$$T_1 = e^{-\mu_1 \rho x} \times T_m,$$

$$T_2 = e^{-\mu_2 \rho x} \times T_m,$$

where ρ is the sample elemental concentration of interest and the μ 's are mass attenuation coefficients. T_m includes any matrix contribution to the transmission. The ratio of these transmissions removes the matrix effects from the concentration measurement:

$$R = T_1/T_2 = e^{-\Delta\mu\rho x} \quad (6)$$

and

$$= \ln R/k \quad (\text{where } k = -\Delta\mu x) \quad (7)$$

The calibration factor k can be evaluated empirically using standard solutions of known ρ .

In coprocessing, the assay difficulties associated with the finite detector resolution and the proximity of the uranium and plutonium L_{III} edges are compounded by the large uranium-to-plutonium ratios. Poor statistics on the data between the two edges make this region unusable for background determination. (The uranium and plutonium L_{III} edges lie at 17.168 and 18.057 keV, respectively, where the energy resolution is ~ 0.375 keV, FWHM.) Furthermore, the incident flux is so attenuated by the relatively high uranium concentration that the fractional contribution of events above the uranium edge is small and the statistics on the plutonium assay are considerably worse than those on the uranium assay. This feature is treated quantitatively below.

3. Hardware Design Features

The L-edge densitometer has four basic parts: the x-ray generator and its associated electronics, the sample and sample-holder assembly, a Si(Li) detector and associated electronics, and a computer-based, multichannel analyzer (MCA) for data acquisition, data reduction and analysis, and measurement control. The source, sample, and detector assembly are illustrated in Fig. 19.

The x-ray source consists of an x-ray tube, enclosed high-voltage power supply, and dual (independent) voltage and current controls. The controls are located remotely (23 m) from the x-ray generator to satisfy physical constraints at SRL.

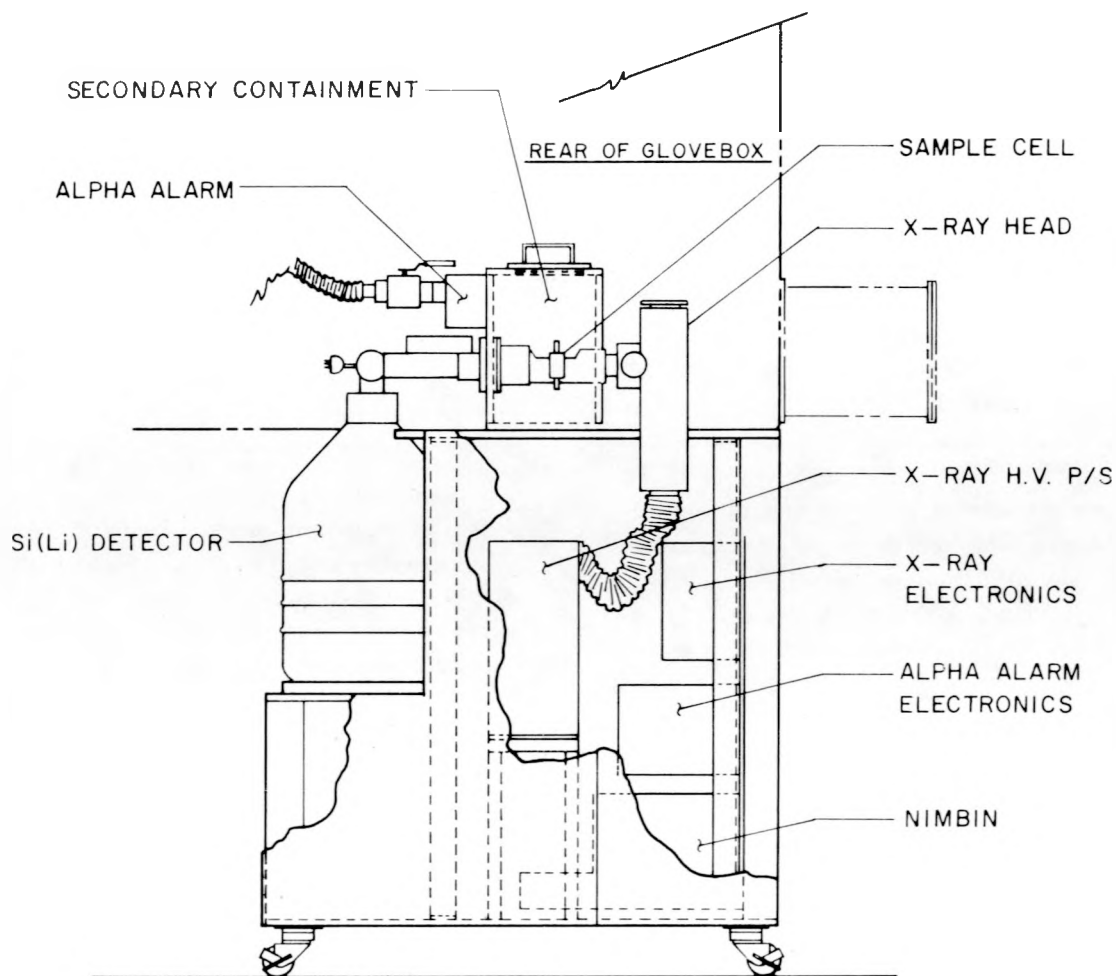


Fig. 19.
Source, sample, and detector assembly for the SRL densitometer.

The x-ray tube operates in the 3-kV to 50-kV range. The present system is run typically between 20 and 21 kV. The x-ray beam shutter is controlled automatically by safety interlock switches on the mechanical system and on the x-ray power supply. The x-ray current control is equipped with an automatic (optional) regulation system based on the detector count rate.

The x-ray spectrum generated by the source exhibits a relatively constant intensity in the vicinity of the L_{III} edges. Thus, attenuation by uranium (typically 4 to 10 times more concentrated than plutonium) reduces the flux of events above the uranium edge to approximately half that below the uranium edge. This effect is somewhat reduced by the insertion of a molybdenum foil between the

source and the sample. The molybdenum foil, which decreases the relative flux below the uranium edge, tailors the spectrum of incident x rays to cut off sharply above the plutonium edge. Figure 20 shows examples of the x-ray spectrum [with (20b) and without (20a) the molybdenum foil] transmitted through a sample cell containing only solvent.

The x-ray beam is collimated to 5 mm both upstream and downstream of the sample.

Sample cells for the calibration phase at LASL were fabricated of stainless steel with 10-mm-diam, 0.75-mm-thick Kel-F windows. These cells were identical to the flow-through type of container assigned for use at SRL except that each had one rather than two (inlet and outlet) solution ports. The thickness of the sample cell was determined

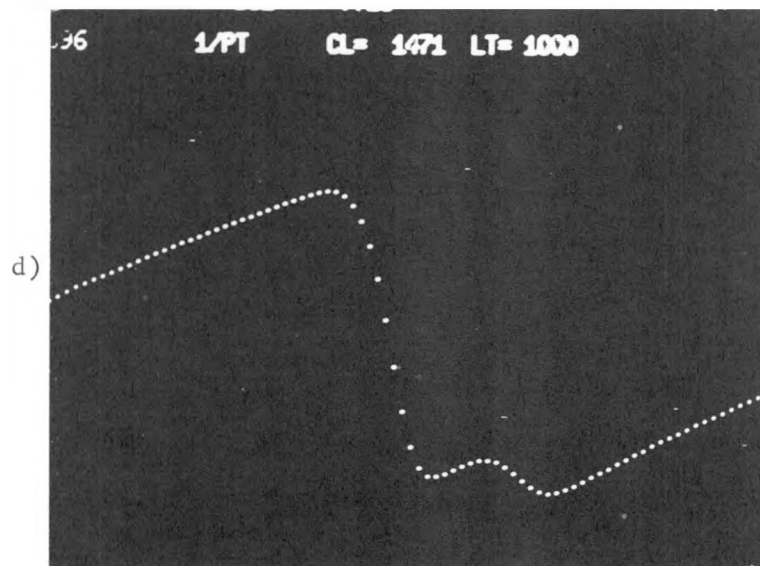
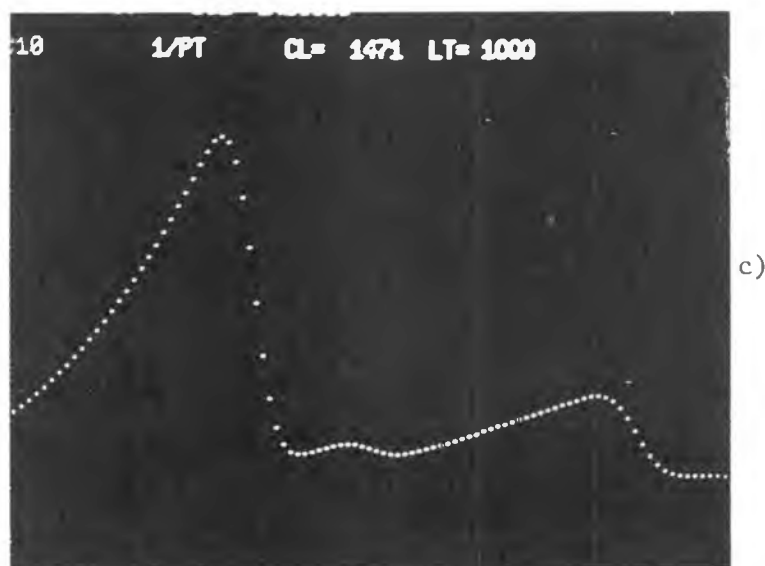
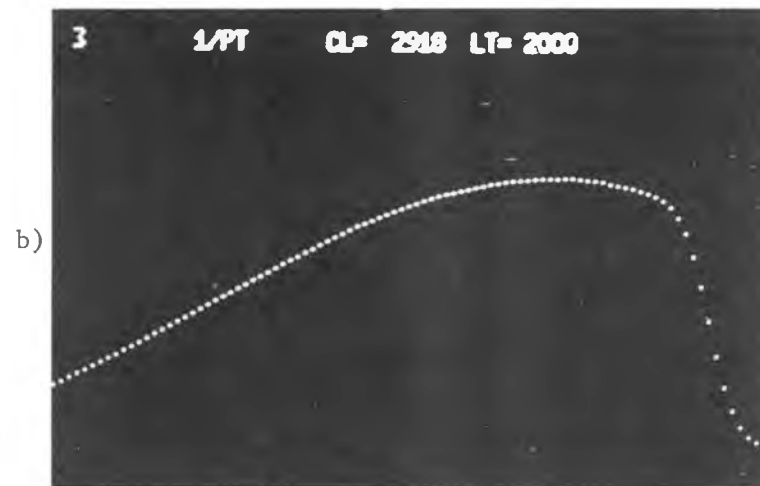
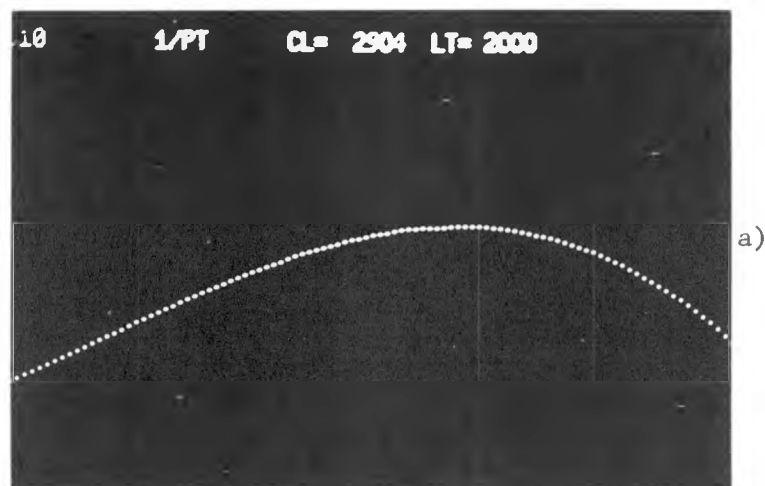


Fig. 20.

Transmitted x-ray spectra taken at a rate of 20×10^3 per second through sample cell with (a) solvent alone—2000-s count time; (b) solvent and molybdenum foil (10 mg/cm^2)—2000 s; (c) molybdenum foil and uranium-plutonium sample ($\rho_U = 40.38 \text{ g/l}$, $\rho_{Pu} = 10.08 \text{ g/l}$)—1000-s count time. The spectra in (a), (b), and (c) are displayed on the same scale. (d) Spectrum (c) normalized to the reference spectrum obtained with the molybdenum foil (b).

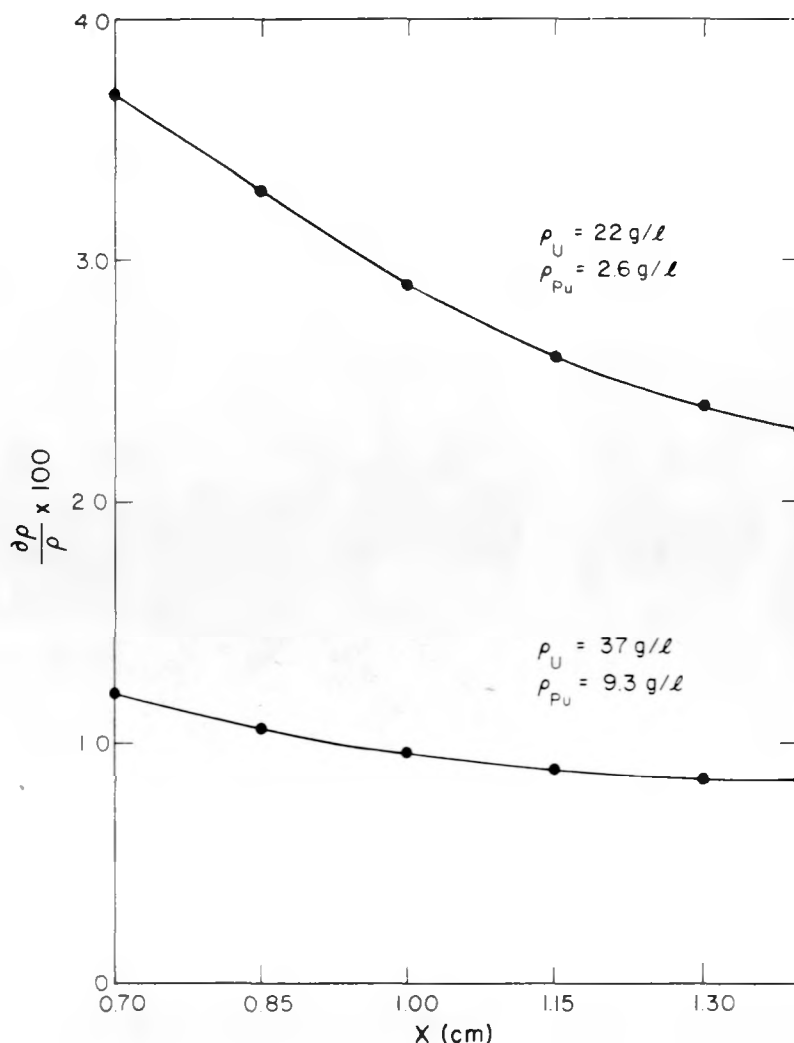


Fig. 21.

Calculated precision in plutonium assay vs thickness of mixed uranium-plutonium sample. Results are shown for two different solution concentrations.

from a calculation of the precision in the plutonium assay for mixed solutions of uranium and plutonium. The results of these calculations are shown in Fig. 21 for two solution concentrations. The precision improves with increasing sample thickness up to 1.4 cm owing to the improvement in the transmission ratio R across the edge. A compromise of 1.15 cm was used for the sample thickness to avoid excessive attenuation by the sample.

The cells were filled with standard uranium, plutonium, and mixed uranium-plutonium solu-

tions. The solvent was 1N HNO_3 . The concentrations of these standards were:

- | | |
|------------------------|-------------|
| 1) 10.08 g Pu/l | |
| 2) 2.015 g Pu/l | |
| 3) | 25.24 g U/l |
| 4) 10.08 g Pu/l | 40.38 g U/l |
| 5) 5.038 g Pu/l | 35.34 g U/l |
| 6) 2.509 g Pu/l | 22.21 g U/l |
| 7) 1.0N HNO_3 | |

The Si(Li) detector is 30 mm² in area, with a resolution of 155 eV FWHM at 5.7 keV. The preamplifier uses a cooled field effect transistor and operates with pulsed optical feedback for optimum resolution. Pulse pile-up rejection and relatively short pulse shaping times (2 μ s) were employed to allow maximum count rates. A detector count rate of $\sim 20 \times 10^3$ per second, which optimizes the data rate for the system, was maintained for all measurements.

The linear signals from the amplifier are fed, along with a gate from the pile-up rejector, to the MCA. The 22-keV x ray produced by a ¹⁰⁹Cd source positioned near the detector is used to monitor (via software) the gain stability of the system. A TN-1700 MCA based in a Data General Supernova computer is used for data acquisition. The procedures for data collection and analysis and measurement control are programmed in a BASIC environment using the software capabilities of the system for computer control of the MCA. The raw data are stored in 4096 channels of analyzer memory. The

remaining 4096 channels of analyzer memory are used for storage of messages and parameters and for data reduction work space.

The user terminal (G.E. model) is equipped with a cassette tape used for transfer of programs to computer memory and for automatic storage of data during an assay.

Figure 22 is a photograph of the system as assembled during the calibration period at LASL. The terminal, MCA, and computer, along with the x-ray power controls, are adapted for remote location from the source-sample-detector assembly.

4. The Automatic Assay Program

Figure 23 is a block description of the computer program that governs the acquisition and analysis of the data in a mode designed for measurement control.

An initialization routine stores calibration and control parameters and messages in unused portions



Fig. 22.
Entire system during test and evaluation period at LASL.

SRL DENSITOMETER ASSAY PROGRAM

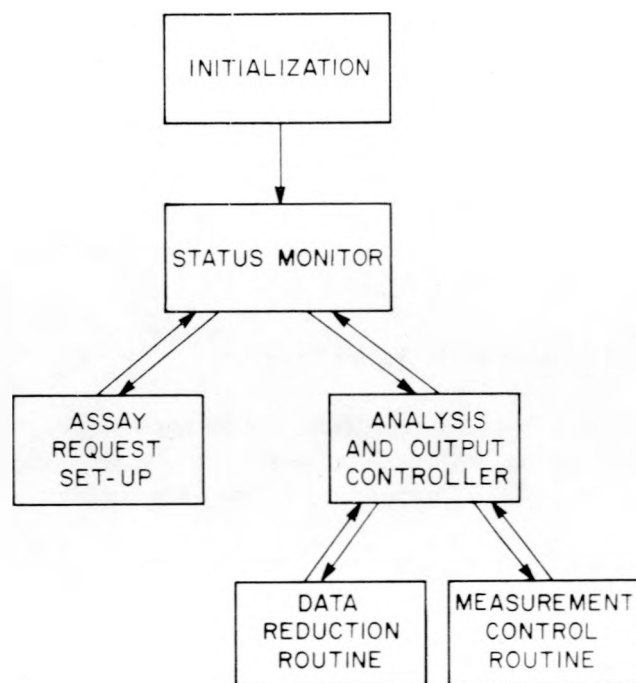


Fig. 23.

Block flow diagram for the SRL densitometer assay program.

of the analyzer memory. The main (data acquisition/analyzer) code is initiated with the input of the date and time and a decision on the data acquisition mode. (An option permits successive assays on a given sample.)

The status monitor transfers control either to the user for inputting details about the assay (initiated by a hardware switch requesting an assay) or to the analysis portion of the program as data acquisition is completed. Data reduction is performed either for measurement control or for assay of a sample of unknown concentration.

With each date change, the measurement control cycle is initiated. This requires obtaining first a reference spectrum and then a foil spectrum as described in the following paragraphs. These specific demands for measurement control are imposed during the assay request set-up.

The reference spectrum is the transmitted x-ray spectrum obtained when only solvent is in the sam-

ple cell. All successive data are normalized to this spectrum to remove, to first order, the matrix effects. The reference spectrum is not analyzed but is stored permanently in analyzer memory.

A foil spectrum is a transmitted x-ray spectrum obtained with the reference sample still in place and with a standard foil (yttrium, zirconium, or uranium) inserted in the beam path in a holder designed for the purpose. The yttrium and zirconium foils simulate uranium and plutonium, respectively, in that their K edges are of nearly the same energy as the uranium and plutonium L_{III} edges.

The concentration equivalents of the foil thicknesses lie in the range of the uranium sample concentrations. Calibration procedures which have determined the precision in the foil measurements are used to establish alarm limits for the foil runs. If an alarm limit is exceeded, the normal assay sequence is interrupted until the problem is corrected. Although the densitometer must be calibrated with primary solution standards, the foil standards insure the validity of the calibration over a period of time that is long compared with the finite lifetimes of the primary solution standards. The technique has been used previously with success.¹

The reduction of data obtained with foils and with samples of unknown uranium and/or plutonium concentrations proceeds first with a normalization, followed by a difference treatment and the analysis routine to determine the experimental value of R . Normalization of the raw data to the data in the reference spectrum already has been discussed. Figures 20c and d show the unnormalized and normalized data for a mixed uranium-plutonium solution where $\rho_U = 40.38$ g/l, $\rho_{Pu} = 10.08$ g/l.

A first difference spectrum⁶ is generated from the normalized data in semilog space. The difference spectrum displays peaks, the centroids of which coincide with the absorption edges in the normalized data. The background-subtracted integral under the peak is the ratio R , defined in Eq. (6). An example of this spectrum is shown for the same solution ($\rho_U = 40.38$ g/l, $\rho_{Pu} = 10.08$ g/l) in Fig. 24.

The first difference spectrum is analyzed by determining the background-subtracted integrals under the uranium and plutonium peaks. The data within specified regions of interest (either hardware- or software-defined), containing either peak or

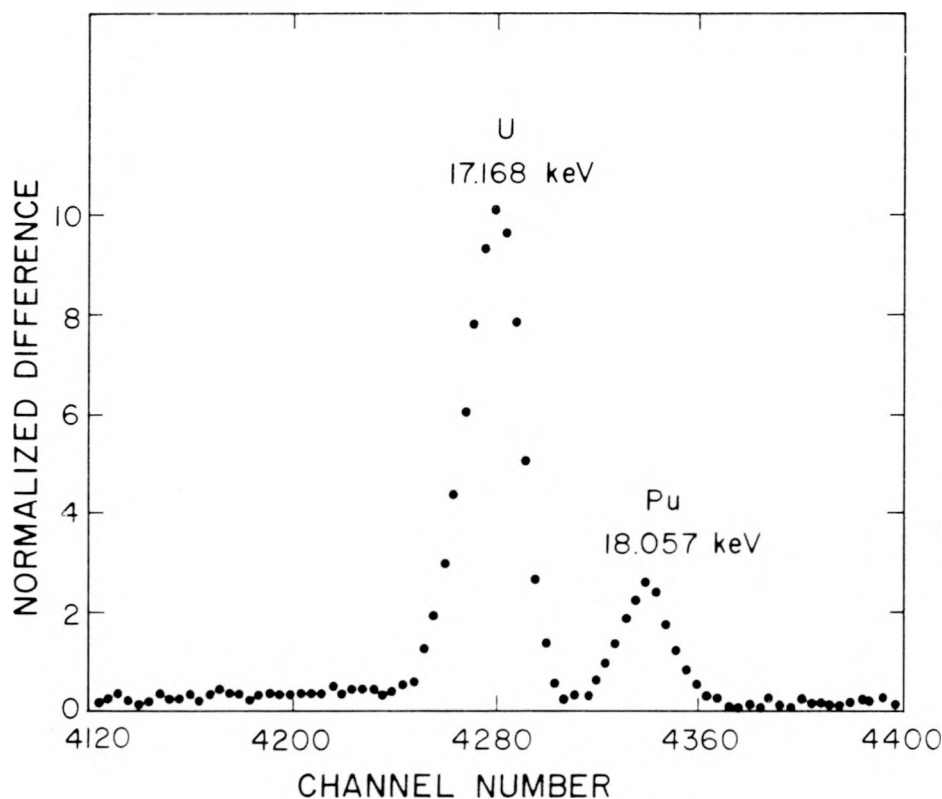


Fig. 24.

Normalized difference spectrum obtained in the assay of a mixed uranium-plutonium solution (40.38 g U/l, 10.08 g Pu/l). Count period 2000 s.

background information, are used for this procedure. The analysis differs if the samples are mixed (uranium plus plutonium) or single elements as described below.

For the mixed uranium-plutonium samples, only one background region is set for each peak because the data between the peaks cannot be used effectively. The uranium background region is just below the uranium peak region, and the plutonium background region is above the plutonium peak region. Data in the background regions are fitted with a straight line which is extrapolated to the edge of the peak region. A horizontal background determined at this intersection is subtracted from the peak data.

Single elements are assayed by using two background regions—one above and one below the peak—set symmetrically about the peak. The flat background is determined by the average number of counts and the average channel in each of the two background regions.

The background-subtracted integrals, which are the experimental values of R , are used to determine the assay by using stored calibration constants according to Eq. (7).

5. Test, Evaluation, and Calibration at LASL

The test and evaluation period at LASL involved the assay of the primary solution standards described in Part 1, Sec. III-B-3, and the secondary foil standards described in Sec. III-B-4. The physical configuration of the hardware during the testing phase was identical with that to be installed at SRL. The calibration of the instrument and the assay precision were established on the basis of the standards. The alarm limits for measurement control were also established from the experimental precision in the foil measurements. The results discussed below are based on data obtained in a 2-wk testing period.

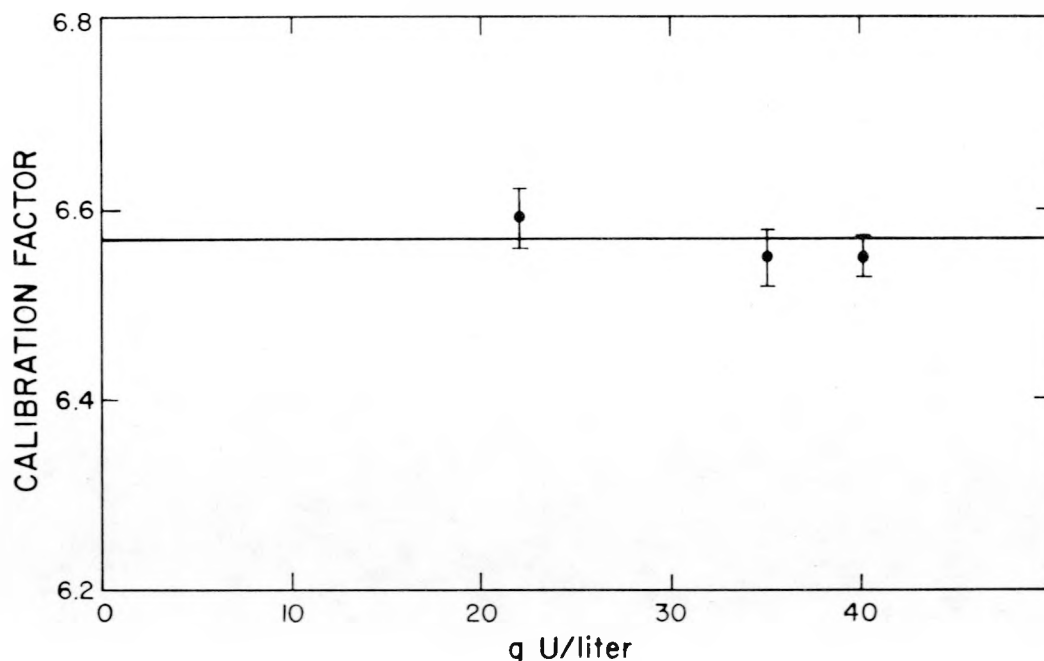


Fig. 25.

Calibration results for uranium based on the three uranium-plutonium mixed solution standards. Note that zero is suppressed to magnify the display.

The measurement precision for the foil runs was established for yttrium and zirconium foils with an equivalent density of ~ 25 g/l. This precision is approximately (less than) $\pm 0.4\%$ at the $1-\sigma$ level. The alarm limits were set for the $2-\sigma$ level.

Because the analysis techniques for the mixed- and single-element solutions differ, the calibration factors for the two solution types will also differ. Figure 25 shows the calibration results obtained for uranium in the mixed solution samples with counting times at 1 to 2×10^3 s. The calibration factors derived for each solution are consistent within the $\pm 0.3\%$ to $\pm 0.4\%$ ($1-\sigma$) experimental precision. Only a single solution standard was available for the uranium single-element case, so the results cannot be displayed graphically. However the experimental precision at the $1-\sigma$ level is $\pm 0.5\%$ for this measurement, performed in 1000-s counting periods. The solid points in Fig. 26 are the plutonium calibration results for the mixed solution standards. For comparison, the results for the single-element plutonium standards are also plotted (open circles)

after having been normalized to the mixed solution results at the 10-g/l point. These data are also consistent (within the $1-\sigma$ experimental precisions) with a constant calibration factor that is $\sim 5\%$ less than that derived from the mixed solution data for uranium. The 5% difference is consistent with the difference between the $\Delta\mu$ values for uranium and plutonium at their respective L_{III} edges. The experimental precision for the mixed solution measurements for plutonium varies from $\pm 2\%$ to $\pm 4\%$ ($1-\sigma$) in counting times varying from 1000 to 2000 s. For the single-element plutonium results, these vary from $\pm 0.8\%$ to $\pm 3\%$ for the same count times.

The test and evaluation results for the SRL densitometer indicate that the instrument can be applied to the assay of coprocessed solutions. The accuracy of the measurements performed on mixed solutions of uranium and plutonium where the uranium/plutonium concentration ratio is 10:1 is $>5\%$ for plutonium and $>1\%$ for uranium.

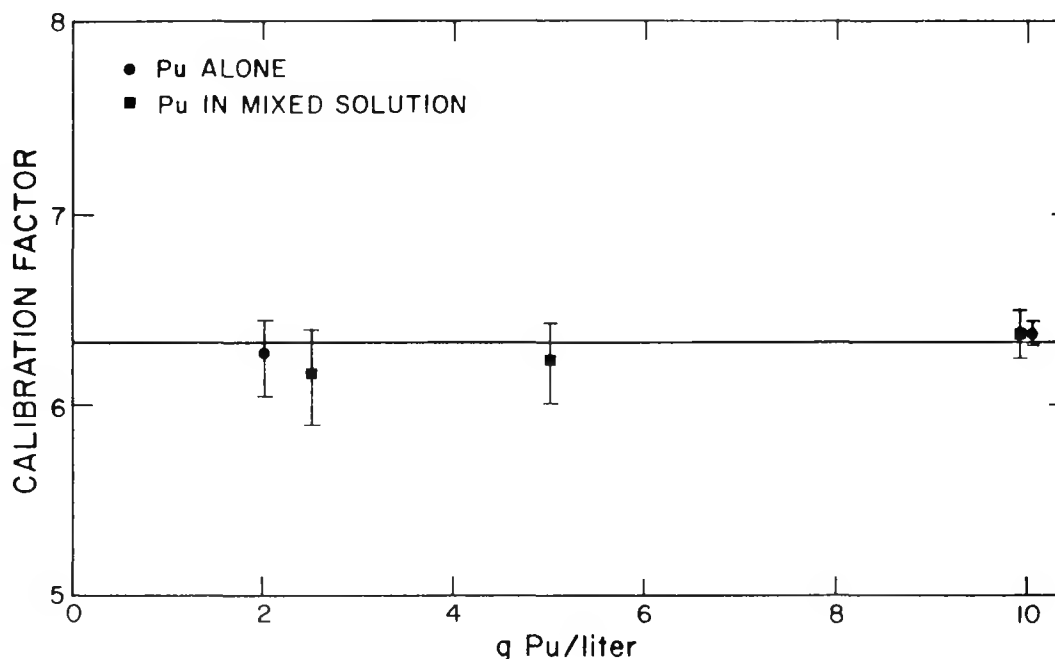


Fig. 26.

Calibration results for plutonium based on the three uranium-plutonium mixed solution standards (solid points). Also shown are the calibration results for the two single element plutonium solutions which have been normalized, at the 10 g/l point, to the mixed solution results.

C. ^{252}Cf Shuffler for Test and Evaluation at SRP (T. W. Crane, G. W. Eccleston, and L. G. Speir)

The goal of this project is to test and evaluate a ^{252}Cf Shuffler neutron interrogation system for the assay of highly enriched ^{235}U materials at the reactor fuel (U-A1) fabrication facility at SRP. The NDA system consists of a sample assay chamber surrounded by neutron detectors, a ^{252}Cf neutron source with a shielded storage tank and source-transfer hardware, and a computer-based control unit that has data reduction capabilities.

The materials to be assayed at SRP include pure uranium-aluminum ingots with up to 4 kg ^{235}U content and scrap and waste items with a lower ^{235}U content. The NDA measurement begins with a sample weight measurement in the Shuffler, followed by a background neutron count. The sample assay continues with a cyclic measurement sequence consisting of sample irradiation using the ^{252}Cf source, followed by delayed neutron counting after the source has been transferred to its storage position.

1. Neutronics Calculations

Fabrication of the ^{252}Cf Shuffler for the test and evaluation program at SRP was initiated during this reporting period. The basic design of the ^{252}Cf Shuffler followed the criteria established by the neutron Monte Carlo design studies (see Ref. 6, pp. 29-30; Ref. 8, pp. 34-36). Monte Carlo calculations, simulating the assay of uranium-aluminum ingots of varying heights, showed the delayed neutron response from the ^{252}Cf irradiation to be linear to within the accuracy of the calculation (about $\pm 3\%$) (see Ref. 6, pp. 29-30). To anticipate the performance of the Shuffler in assaying the different material types at SRP, we made additional calculations in which the sample diameter, density, and matrix material varied.

The expected response (delayed neutron counts/gram ^{235}U) was not noticeably affected by changes in the sample diameter. Presumably, the response uniformity for diameter changes results from the higher penetrability of the smaller samples

being compensated for by the increased multiplication in the larger samples.

The response did show a slight dependence on the sample density, with the lower density samples having a larger response because of their increased penetrability by neutrons. A correction factor based upon the weight measurement at the beginning of the assay and the height measurement extracted from the ratio of the responses of the side and bottom neutron detectors (see Ref. 6, pp. 29-30) can be used to achieve a uniform response with density. One SRP material type containing lithium fluoride was thought initially to be potentially troublesome because of the possibility of neutron absorption in the lithium (${}^6\text{Li}$). However because the energy of the interrogating neutrons is above the region where the lithium absorption cross section is appreciable, this lithium-bearing material has a response consistent with that calculated for the pure uranium-aluminum ingots.

To summarize, the Monte Carlo calculations indicate that measurements of the ${}^{235}\text{U}$ content for the materials at SRP should be accurate to at least 2%.

2. Mechanical Hardware

The complete Shuffler system (Fig. 27) comprises two main frame units: the source storage unit (A) and counting unit (B). Both units are aligned and connected by a 12-in.- (30.5-cm-) thick polyethylene shield (C). Each main unit will weigh ~ 4000 lb (1816 kg). Caster wheels inside wheel wells provide mobility for check-out and installation.

Attached to the outside panel of the storage unit is the source drive motor (D) and the Teleflex cable extension tube (E). The ${}^{252}\text{Cf}$ source (F) is connected to the Teleflex cable (G) by an "oilite" bronze transition piece (H). The source drive motor is a dc stepping motor, 200 steps per revolution, with an attached shift encoder. The source is 900 mg (2×10^9 n/s) of ${}^{252}\text{Cf}$ doubly encapsulated in stainless steel. The source and cable travel through a Teflon plastic tube (I) which is captive inside a stainless steel pipe (J). The Teflon tube is canted 3° from horizontal through the storage unit. It assumes a slight bend

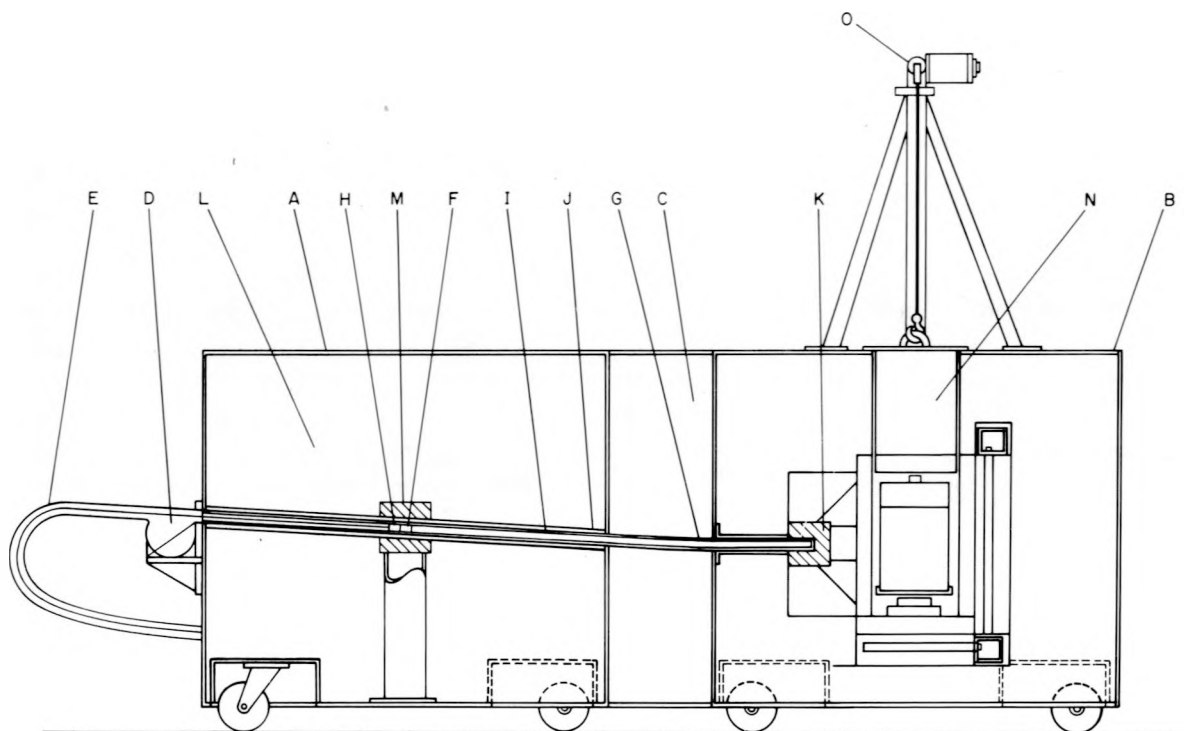


Fig. 27.
Complete ${}^{252}\text{Cf}$ Shuffler unit.

through the shield between the units and is horizontal inside the counting unit where it terminates inside a 6-in. (152.4-mm) right cylinder of tungsten (K). This prevents line-of-sight between the source storage and irradiation positions and aids in reducing source-related neutron backgrounds.

The storage and counting units both have 0.5 in. (12.7 mm) of lead shielding over the entire inside surface of the outer steel panels. This lead is lined with 0.032-in.- (0.81-mm-) thick cadmium sheet. The bulk of the shielding (L) inside both units is high-density (0.94 g/cm³) polyethylene except at positions adjacent to the storage (M) and irradiation (K) tungsten shields where lithium- and boron-loaded polyethylene has been installed. The boron- and lithium-loaded polyethylene reduces the secondary build-up of 2.2-MeV gamma rays from hydrogen capture of source neutrons. The sample cavity access door (N) is essentially an 86-lb (39-kg) shield plug lifted up and transferred to the side by the special jib crane (O) used for sample insertion and removal. An interlock between the access door and the source drive unit prevents the source from moving to the irradiation position when the door is lifted.

The sample cavity (A), as shown in Fig. 28, is 9.5 in. (241 mm) in diameter and 17 in. (432 mm) high. All surfaces facing into the cavity are lined with 0.032-in.- (0.81-mm-) thick cadmium sheet. Directly under the cadmium is 0.265-in.- (6.7-mm-) thick Boral plate (B) (boron carbide-loaded aluminum). Next in line radially is a 2-in.- (50.8-mm-) thick iron moderator-reflector (C) which supports the source tailoring reflectors (D), side plates (E), nickel moderator (F), and tungsten shield (G). The sample can is carried inside an aluminum canister (H), which in turn is lifted by the sample jib crane (I) and lowered onto the sample cavity turntable (J). The long lifting handle (K) bayonets into the top cover of the canister and provides ease of reach down into the sample cavity for connection of the jib crane to the canister.

The large counting array (L) around the sample cavity contains 16 ³He neutron detectors (M). The detectors have a 21-in. (533.4-mm) active length and 1.0-in. (25.4-mm) diam. They are filled with a gas mixture that has a low gamma sensitivity (Ref. 8, p. 39). The top junction box (N) contains two preamplifiers and the high-voltage distribution

network. Connectors for high voltage, preamp power, signal out, and test are provided at two places on the periphery of the top junction box. The box cover (O) is gasketed and hermetically sealed. A dessicant inside the junction box ensures a dry atmosphere for the electronics. Access to the detector ring is provided through the top cover (P) of the counting main frame unit. The bottom detector bank (Q) contains six ³He detectors. The bottom junction box contains three preamplifier boards; one for the bottom bank of neutron detectors and two for the flux monitors inside the cavity (S). Connectors through the box provide high voltage for the neutron detectors, common high voltage for the two flux monitors, a common dc power for all three preamplifiers, separate inputs for both flux monitors, signal out from each flux monitor, a common "test" connection, and a signal output from the bottom detector bank. Access is provided for removal of the bottom detector bank through the end of the counting main frame.

Figure 29 shows the load cells (used for sample weighing) incorporated into the sample rotation unit (A). Three load cells (B) are equally spaced below the turntable near the outer radius. The load is transmitted from turntable to load cell via Kel-F plastic buttons (C). The turntable is raised and lowered relative to the load cells by a cam (E) driven by a gearhead motor (F). The turntable rotates at 7.73 rpm.

3. Electronics Hardware

Control of the ²⁵²Cf Shuffler for SRP will be accomplished with a Digital Equipment Corp. LSI-11 microcomputer. The LSI-11 microcomputer uses a 16-bit word with a memory size of 32k words. The availability of a large number of commercial peripheral interfaces to support the LSI-11 was a major criterion in the selection of this computer. In addition, the software capabilities permit the development of programs that use high-level languages, such as FORTRAN IV.

Electronics hardware development for the Shuffler is based on the use of commercial components wherever possible and on limiting design development to those components that are not commercially available. Shuffler components and operations that will be controlled by the LSI-11 are:

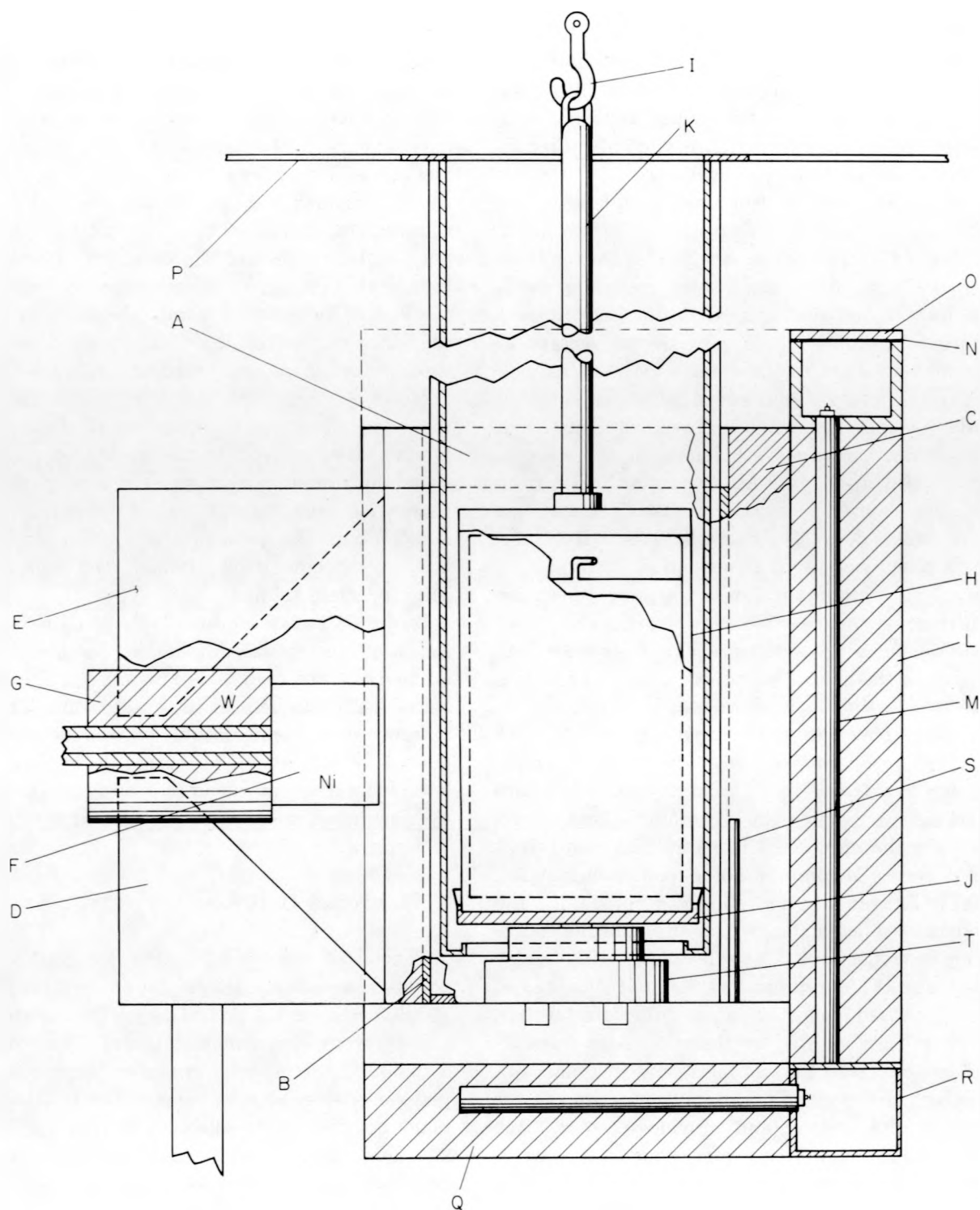


Fig. 28.
Schematic of the Shuffler interrogation-counting well.

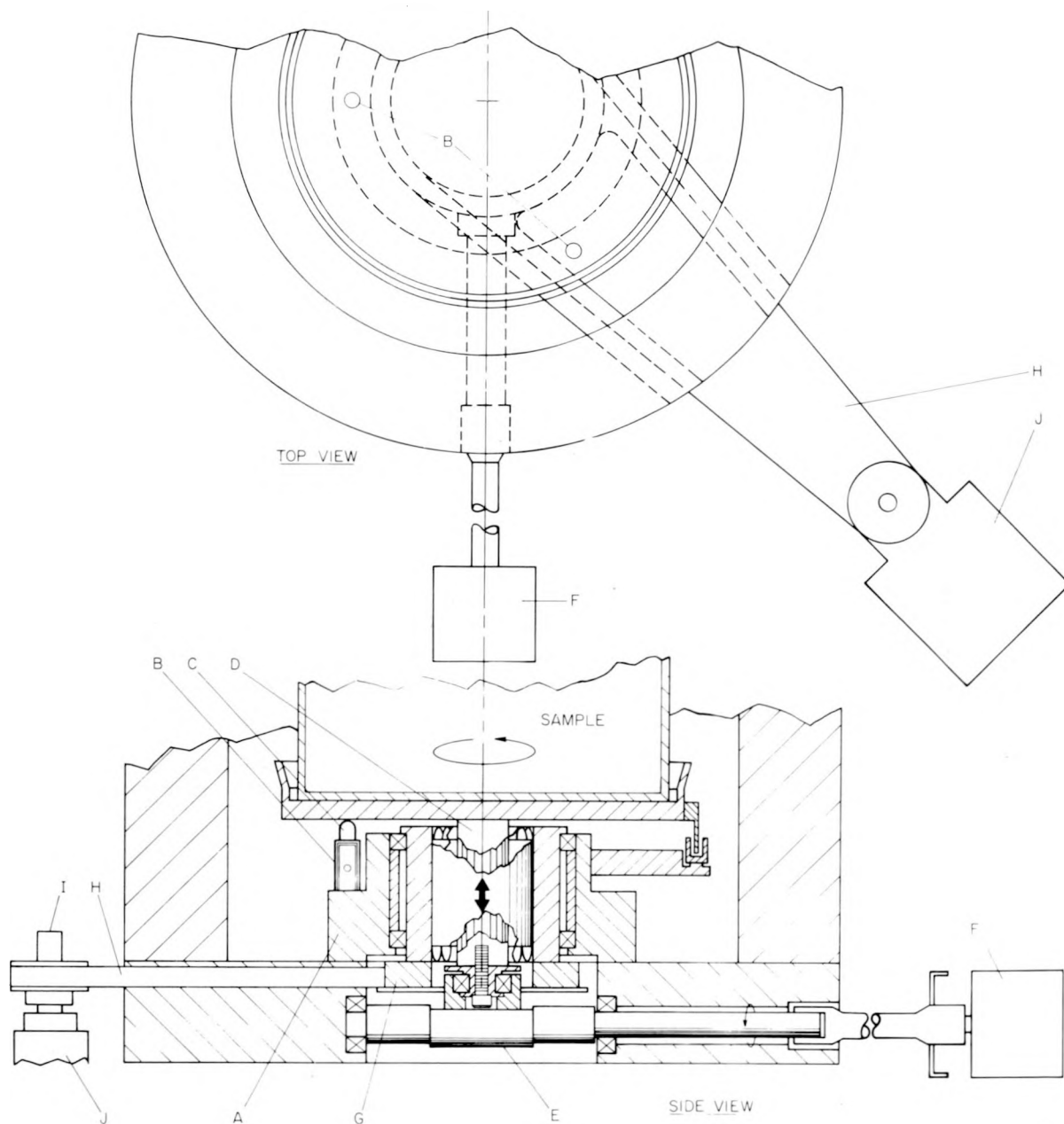


Fig. 29.
Turntable and weighing unit for the Shuffler.

- Load cells for sample weight,
- Sample rotation platform,
- Elevation and lowering of the rotation platform,
- Position of the assay chamber door (open/closed),
- Transfer and positioning of the ^{252}Cf source,
- Control and readout of the counting scalers, and
- Temperature measurements.

The above components and functions are supported largely by commercial devices, except for the scalers and an encoder board used for source transfer. The scaler board has six scalers, each having a 24-bit counting capacity (16 777 215) with an internal software-controlled test function to check each scaler. The unit was developed to permit the counting electronics to be interfaced directly to the

LSI-11, providing a compact package that supports the full scaler requirement of the Shuffler. The encoder board is used to record the stepping motor movement during source transfer and uses optical isolation with an up/down counter that is immune to noise transients from the source transfer system. The interface for each device is designed to permit local control for individual testing of system components.

The peripheral support hardware for the LSI-11-based Shuffler consists of a CRT terminal with a detached keyboard, a hardcopy terminal, and a dual floppy disk. With the exception of the keyboard, all electronic components will be housed outside the sample assay room and will be located in an environmentally controlled equipment rack. The keyboard will be located near the Shuffler assay instrument to permit easy access for system control during sample assays. The purpose of locating equipment outside the assay room is to prevent the possibility of contamination and to permit easy access for maintenance and repair.

Software development to control the Shuffler is based on the use of FORTRAN IV. In addition, structured programming concepts are being applied for the development of more understandable and easier code programming. The software control system consists of a set of commands called by a supervisory routine through one-letter keyboard calls. The general FORTRAN subroutines consist of assay and data reduction, disk input/output of data, calibration routines, and system diagnostic routines.

The Shuffler is controlled with special assembly language driver packages that are FORTRAN-callable. The drivers are machine-dependent and permit faster, more efficient control of the Shuffler.

D. ^{252}Cf Shuffler Assay System for the Fluorinel Reprocessing Facility (H. O. Menlove and G. W. Eccleston)

A neutron interrogation assay system that uses the ^{252}Cf Shuffler principle⁵ is being designed for the measurement of waste canisters and spent fuel packages at the proposed Fluorinel Reprocessing Facility to be operated by ACC.¹³ The assay system is to be used for accountability of ^{235}U in waste canisters after reprocessing. The interrogator will be

an integral part of the facility and the design requires close interaction between LASL, ACC, and Ralph M. Parsons, the architect engineer. At present, design details corresponding to the placement and general location of the Shuffler have been specified, reviewed, and are nearing final stages of the Title II portion of design.

In striving for improved nuclear material control and accounting, ACC has decided to use an NDA assay system to determine the ^{235}U content in the waste canisters from the fluorinel dissolver. The sludge is nonuniform and contains large quantities of zirconium, fission products, and thermal-neutron poisons such as boron and cadmium (Ref. 6, pp. 31-34). This material is generated in the dissolution of high-enrichment spent fuel elements.

The heterogeneities and high gamma-ray background ($\sim 10^3$ R/h) from the fission products preclude the use of passive gamma-ray assay techniques. Also, the large quantities of boron and cadmium neutron poisons preclude the use of thermal-neutron interrogation techniques. For these reasons, an assay system employing fast-neutron interrogation seemed most appropriate.

During the NDA measurement, the waste will be contained in a metal canister roughly 12 cm in diameter and 61 cm long, normally containing <300 g of ^{235}U . Samples containing >300 g of uranium will be recycled through the dissolution process after measurement.

In addition to the waste canister measurement, the same assay instrument will be used to measure the ^{235}U content of spent fuel assemblies before dissolution. This fuel package analysis serves as an additional criticality check and is not a requirement for facility operations.

Two measurement locations (see Fig. 30) exist in the Shuffler; one is for assay of waste canisters and the other is for assay of spent fuel. The low ^{235}U content of waste canisters and their fixed geometry allow the interrogation chamber to be sized to the canister diameter. The large size of fuel elements requires an assay tube of larger (50.8 cm) diameter. The fuel elements may vary both in size and shape, so various off-center positions can be used when lowering samples through the chambers for measurement. A sample counted at various chamber positions (relative to the detectors and interrogation source) could give a range of counting

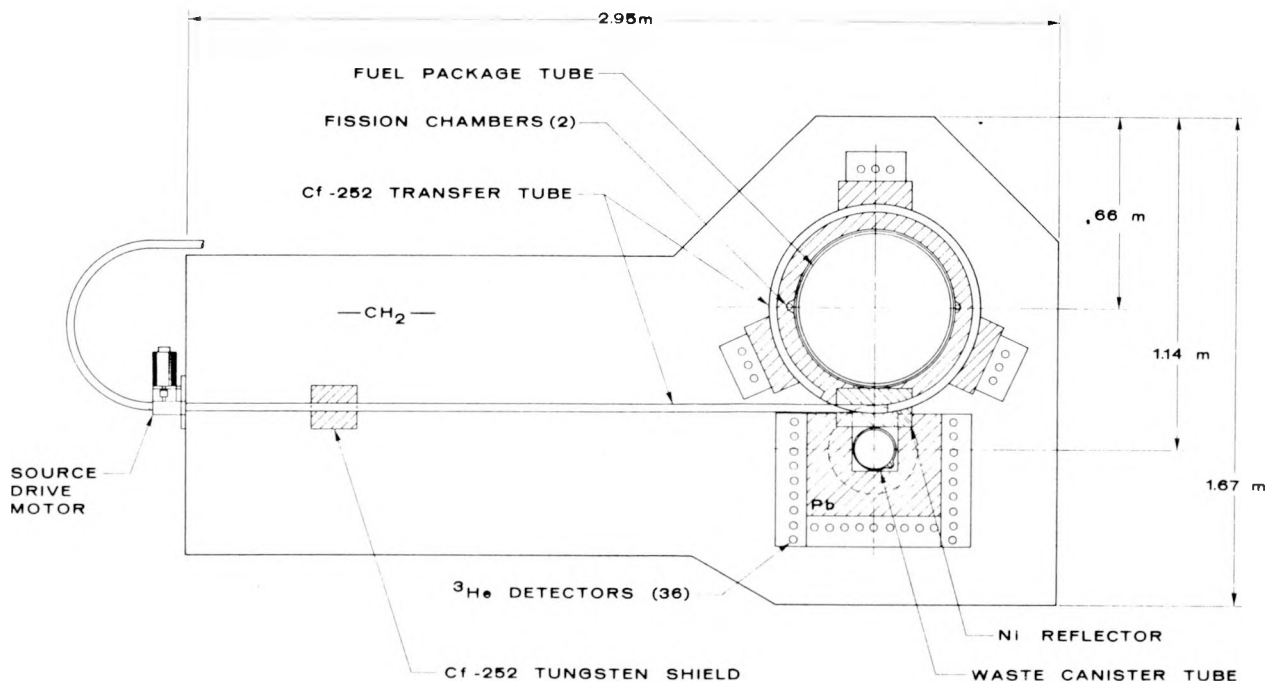


Fig. 30.
Neutron interrogator of the Shuffler type for the Fluorinel Reprocessing Facility in Idaho. There are separate interrogation chambers for the spent fuel assemblies (large tube) and the waste canisters (small tube).

values and the net effect could be an induced bias in the assay. In addition, the hot-cell handling crane and attachments prevent the samples from being rotated during the measurement, thereby introducing a possible assay bias.

Geometric and matrix effects from the spent fuel elements are being mitigated by two methods. First, the ^{252}Cf source will be cycled around the circumference of the interrogation chamber, thereby providing a sample irradiation from all sides of the chamber and decreasing matrix effects and variations from different sample positions in the tube. Second, the three delayed neutron detector banks for the fuel element are located symmetrically around the chamber, $\sim 120^\circ$ apart. The movement of a sample away from one detector bank will produce a lower count rate in that bank, but the other detector banks will have a compensating effect through increased count rates. The purpose is to produce a measured assay independent of fuel element location or movement within the interrogation tube.

The relatively small size of waste canisters does not require source rotation to reduce geometric or matrix effects.

E. HPFL Fuel Rod Scanner (M. S. Krick and H. O. Menlove)

The Hanford Engineering Development Laboratory (HEDL) is procuring an intermediate fuel rod scanner and is planning to obtain an advanced scanner. Performance specifications for these scanners were described in Ref. 6. LASL is assisting HEDL by performing specific tests to evaluate proposed measurement schemes. In particular, the stability of NaI(Tl) detector systems is being investigated relative to gas proportional counter detection systems. NaI(Tl) detectors are being studied in the following areas:

- Stability with respect to temperature changes, with and without electronic stabilization;

- Short-term stability with respect to sudden count rate changes;
- Long-term stability following a sudden count rate increase.

The effect of temperature and humidity changes on NaI(Tl) detectors, proportional counters, and associated electronic modules will be studied in an environment chamber. The chamber features automatic cycling with a temperature range of -73°C to 177°C and a relative humidity range from

20-95%. The 70 x 70 x 55-cm chamber can accommodate a NaI(Tl) or proportional counter detection system and a NIM bin.

High-pressure (8 atm), xenon-filled proportional counters (with and without bismuth linings) have been purchased for evaluation as fission-product gamma-ray detectors. Also, bismuth-lined, argon-filled proportional counters are being acquired for evaluation as moderate-efficiency, high-stability gamma-ray detectors.

V. DETECTOR AND ELECTRONICS DEVELOPMENT

A. Conceptual Design of Selected Instrumentation for an International Safeguards System at the Portsmouth Gas Centrifuge Enrichment Plant (J. W. Tape, M. P. Baker, and R. B. Walton)

A 5-yr effort by the US Working Group on Enrichment Plant Safeguards and meetings of representatives from various DOE contractors in March and April 1978 have resulted in the identification of instruments which could or should form the basis of an international perimeter safeguards system around a large-scale centrifuge facility.

The ground rules for an international safeguards inspection system at enrichment facilities generally limit the inspectors' access to the perimeter of the plant. This constraint changes a relatively easy material balance verification and cascade surveillance problem into a difficult task. In addition, inspectors of a US centrifuge facility such as the Portsmouth Gas Centrifuge Enrichment Plant (GCEP) will not be allowed to see certain classified packages, such as incoming parts and outgoing cascade waste.

The purpose of a perimeter safeguards system is to ensure that the enrichment plant is being operated in its declared mode in three ways: verification of the declared stream material balance, detection of the production of high-enriched uranium (HEU), and detection of undeclared feed, product, or tails streams through the perimeter. The instruments under consideration here have application to the first two strategies. (Instrumentation for the detection of undeclared feed, product, or tails streams is the responsibility of LASL's Group Q-2 and of Sandia Laboratories.)

In-line enrichment monitors for feed, product, and tails streams are equivalent to the continual presence of an inspector in the Feed and Withdrawal (F/W) building and provide a 100% sampling plan. Data from these monitors, when combined with mass measurements of the cylinders as they are loaded and emptied, provide near real-time material balance information for the facility. The continuous-inspection feature of the in-line meters is especially important for centrifuge enrichment plants because of their short equilibrium times. The final material balance data, derived from more accurate mass spectrometer samples and weighings, will confirm the less accurate but more timely in-line data. The in-line monitor also should deter such strategies as the introduction of higher than normal enrichment feed into the cascade by use of shielded feed cylinders.

The ability to detect the production of HEU is the best possible deterrent for safeguarding an enrichment plant; however, ground rules for the perimeter system do not make the detection of HEU production easy. One possible detection technique is the use of area radiation monitors placed outside the perimeter. Another technique relies on the fact that waste streams penetrating the perimeter will contain uranium with enrichments indicative of the enrichments inside the cascade. Thus HEU production could be detected even if the plant operator makes an error and allows to pass through the perimeter any waste material that contains uranium with an enrichment above the allowed value. All of the waste streams provide an opportunity for an enrichment measurement, but only the alumina

trap material is expected to contain sufficient quantities of uranium for a feasible assay.

Conceptual designs are given next for instruments that can be applied to the verification of the GCEP declared material balance and the detection of HEU production.

1. In-Line Monitor for Product Assay

The in-line product assay meter is designed to measure the enrichment of the GCEP product liquid UF_6 streams as they are transferred to the load-out cylinders. The measurement is based on the enrichment principle,^{14,15} which states that the count rate of 186-keV gamma rays from ^{235}U is proportional to the enrichment of the uranium in the sample if that sample is thick.

In 1973 an in-line product assay meter was installed by LASL personnel at the Goodyear Atomic Corp. (GAT) gaseous diffusion plant extended range product withdrawal station.¹⁶ (The GAT system included a neutron detection system as well as a gamma-ray system. The neutron signal in this case is proportional to the ^{234}U assay. If the application of minor isotope safeguards technique (MIST) to the declared streams at the GCEP proves to be an important safeguards technique, in-line assay of ^{234}U might also be desirable.¹⁷)

The product monitors at the GCEP will be similar to the one now at GAT. There will be a sample chamber with heaters and shielding for the gamma-ray measurement. A stabilized NaI(Tl) detector will detect the 186-keV gamma rays, and signals from the detector will be processed locally with standard NIM electronics. Each of the 16 product lines will require a separate sample chamber, detector, and local electronics if there is the possibility of having 16 separate product assays; however, a single (or a few) data processing system(s) could serve all 16 measurement stations.

Installation of the in-line sample chambers and heaters should be included in the construction of the F/W building. The detector and shielding take up a relatively small space. Local electronics probably

will consist of a NIM bin for every two detectors. If the F/W building is air-conditioned, no additional environmental control should be required for the local electronics. The power requirements for the detectors and local electronics are not large; however, separate, "clean" power lines used only for electronics are desirable. The data processor might require a more substantial power source (30 amp, 120 V). The cost of a detector, shielding, and local electronics is ~\$8k/station. The data processor(s) might cost \$50-75k, depending on the desired level of sophistication.

The F/W building could be equipped with a large data analysis and control computer that would accept data from all of the F, P, and T assay stations, analyze them, and correlate the assay data with seal information and serial numbers of cylinders on-line as well as load-cell derived weights.

2. In-Line Monitor for Tails Assay

The in-line tails assay meter is identical in principle to the product monitor. The tails assay of liquid UF_6 is monitored in-line as the cylinders are filled. The major difference between the designs of the two systems is in the type of detector employed. A less favorable signal-to-background ratio, caused by the smaller amount of ^{235}U at the tails assay, may preclude the use of NaI(Tl) detectors. If it is assumed that this will be the case and that there will be no dramatic developments in room-temperature gamma-detector technology (e.g., CdTe) then liquid-nitrogen-cooled germanium detectors will be required. Each detector will have a 45-cm-diam x 70-cm-high nitrogen dewar associated with it. A liquid nitrogen supply must be available for the (approximately) weekly filling of the detectors' dewars.

The precision of the NaI(Tl)-based product meter at GAT is 0.15% in a 10-min assay. From this number plus the expected tails assay and the ratio of germanium resolution to NaI(Tl) resolution, the estimated assay precision at the tails is 0.6% for a similar assay time.

*"Thick" means a thickness greater than a few mean free path lengths for 186-keV gamma rays.

The cost of each tails assay station (detector plus local electronics) is ~\$15k. The data processor(s) used for the product might also be able to handle the tails instruments.

3. In-Line Monitor for Feed Assay

The in-line enrichment meter for the feed stream at the GCEP is designed to determine the assay of the gas-phase feed to the process buildings. The enrichment principle employed in the product and tails monitors will not work for the gas-phase UF_6 unless a large thickness of gas is assayed (>1 m); however, the gas-phase enrichment can be determined by combining measurements of the ^{235}U concentration and the uranium concentration in the gas. The ^{235}U concentration can be determined from a passive rate measurement of 186-keV gamma rays emitted by ^{235}U .

If a gamma detector views a gas cell of volume V , containing UF_6 with density ρ and enrichment I , then the counting rate of 186-keV gammas is given by:

$$R(186) = I \cdot \rho_{\text{UF}_6} \cdot V \cdot \frac{M_{\text{U}}}{M_{\text{UF}_6}} \cdot \Gamma \cdot P_d \cdot \epsilon_d, \quad (8)$$

where $M_{\text{U}}/M_{\text{UF}_6}$ is the fraction of uranium in UF_6 , Γ is the number of 186-keV gammas emitted per gram of ^{235}U per second, P_d is the probability that a decay occurring in the gas volume will produce a gamma that strikes the detector, and ϵ_d is the probability that the gamma will be detected. Equation (8) can be simplified to:

$$R(186) = I \cdot \rho_{\text{UF}_6} \cdot k, \quad (9)$$

where k is a constant. The derived enrichment is given by:

$$I = \frac{R(186)}{\rho_{\text{UF}_6} \cdot k}. \quad (10)$$

The constant k can be determined from a calibration measurement, $R(186)$ is measured, and ρ_{UF_6} must be determined.

The feed-gas pressure is high enough for attenuation (absorption) of gamma-rays in the gas to be a

problem. Thus the actual counting rate of 186-keV gammas will be less than expected on the basis of Eq. (8). The rate loss can be corrected by a standard technique employed in NDAs, provided the average attenuation of 186-keV gamma rays transmitted through the sample can be determined.¹⁸ The attenuation by the gas can be calculated from pressure measurements if the gas is known to be pure UF_6 , or it can be measured by an external source of photons transmitted through the gas sample into the detector. The attenuation measurement has the added advantage of providing an independent indication of the UF_6 gas density. Therefore, a simultaneous measurement of the 186-keV gamma rate and the count rate of gamma rays from an external source transmitted through the gas cell provides enough information to determine the enrichment of the gas-phase UF_6 . The physical characteristics of the gas-phase monitor should be similar to those of the product and tails monitors. A sample chamber 5 cm thick and 10 cm in diameter and equipped with pressure and temperature transducers should provide a volume of gas that can be assayed easily. The chamber probably would sample the main feed flow continuously and remain at the same pressure. A well-shielded NaI(Tl) detector is probably adequate for this application. If a germanium detector must be used, extra space will be required for the liquid-nitrogen dewar. The electronics and processor needs will be similar to those of the product and tails monitors. The cost will be similar to that of the product monitor if a NaI(Tl) detector is used and will be similar to the cost of the tails monitor if a germanium detector is used. An additional ~\$2-3k will be needed for pressure and temperature devices.

Calculations indicate that an enrichment assay of ~1% RSD can be obtained in 10-min assay periods for pressures characteristic of the GCEP feed.

4. Trap Material Enrichment Meter

The trap material enrichment meter is designed to screen outgoing alumina trap material for enrichments larger than the declared top product of the facility and possibly to provide information on the average uranium content of the trap material for material balance purposes.

A number of assay techniques might provide the data required for the enrichment measurement of

large volumes of relatively low-density uranium-bearing waste. One of these methods is described here and other methods are mentioned briefly. The size and cost of the instrument are determined mainly by the size of the trap material containers and not by the particulars of the technique.

Compounds of uranium and fluorine emit neutrons from two sources: (1) α, n reactions in which an alpha particle from uranium decay interacts with a ^{19}F nucleus and emits a neutron and (2) spontaneous fission neutrons. For the most part, in natural uranium (not recycle), α, n neutrons are produced by the decay of ^{234}U and ^{238}U , whereas spontaneous fission neutrons arise almost solely from the fission of ^{238}U nuclei.¹⁹ Thermal-neutron counting techniques can be used to distinguish between α, n neutrons and fission neutrons. An assay device that can detect both the total neutron emission of a container and the spontaneous fission neutrons will allow the determination of the ratio of $^{234}\text{U}/^{238}\text{U}$ which is approximately proportional to the ^{235}U enrichment of the sample. If the uranium to be assayed contains no ^{232}U or ^{236}U , and if the ^{234}U -to- ^{238}U ratio is assumed to be approximately constant, $\sim 1/125$, and if small (few per cent) contributions to the n yield are ignored, then the total neutron yield of UF_6 and the spontaneous fission yield are given by:

$$N_T \approx 4.8 \times 10^3 I_{235} M_U + 28 M_U \quad (11)$$

$$N_{SF} \approx 15 M_U (1 - I_{235}) \quad (12)$$

where N_T , N_{SF} are in units of neutrons/second; M_U is in kilograms, and I_{235} is the enrichment (fractional). Combining Eqs. (11) and (12) results in the following expression for the enrichment:

$$I_{235} \approx \frac{(N_T/N_{SF}) - 1.87}{(N_T/N_{SF}) + 320} \quad (13)$$

Figure 31 shows plots against I_{235} of N_T , N_{SF} , and the ratio N_T/N_{SF} . The values for N_T and N_{SF} were calculated for 5-kg masses; the ratio is independent of the sample mass.

A 4π well counter constructed of ^3He proportional counters, polyethylene moderator, and polyethylene or water shielding with a neutron detection ef-

iciency of 20% can probably verify the assay of 5 kg of UF_6 contained in 55-gal drums of trap material. Experimental tests on smaller samples should verify the principle and establish the expected accuracy. A measurement with an RSD of 10% should be sufficient for IAEA perimeter safeguards.

The approximate cost of a well counter for 55-gal drums is \$100k. The electronics package is small and can be made quite rugged. Some sort of barrel-handling system will be required in conjunction with this instrument and will add to the cost.

The above technique will not work in the presence of varying or unknown amounts of ^{232}U . If recycle uranium is expected, a combination of an active neutron interrogation technique to determine the ^{235}U and a passive coincidence counting method for ^{238}U will have to be employed. The physical scale and cost of an active/passive instrument would be somewhat greater than the passive unit just described.

Other assay techniques might be applied to the trap material problem, such as combinations of gamma-ray and neutron methods; however, a considerable amount of R&D will be required to demonstrate their feasibility.

5. Area Radiation Monitors

Area radiation monitors have the potential to detect the clandestine production of HEU, as both the gamma-ray and neutron emission rates from UF_6 increase with increasing enrichment (see Fig. 31). Detection of HEU production is easy if the detectors can be placed inside the cascade and their performance monitored from time to time. However, under the current ground rules for protection of classified technology, international inspectors will not be allowed access to the cascades. Nevertheless, an agreement might be reached between the facility operator and the inspector to allow the inspector access to the roof area of the declared process buildings for placement and periodic check of area radiation monitors. (Detectors also should be placed on other large buildings inside the perimeter to monitor undeclared enrichment in those areas.)

The poor penetrability of the primary gamma rays from uranium rules out the use of gamma detectors for area monitors on a roof. Neutron detection schemes have a dual advantage in that the radiation

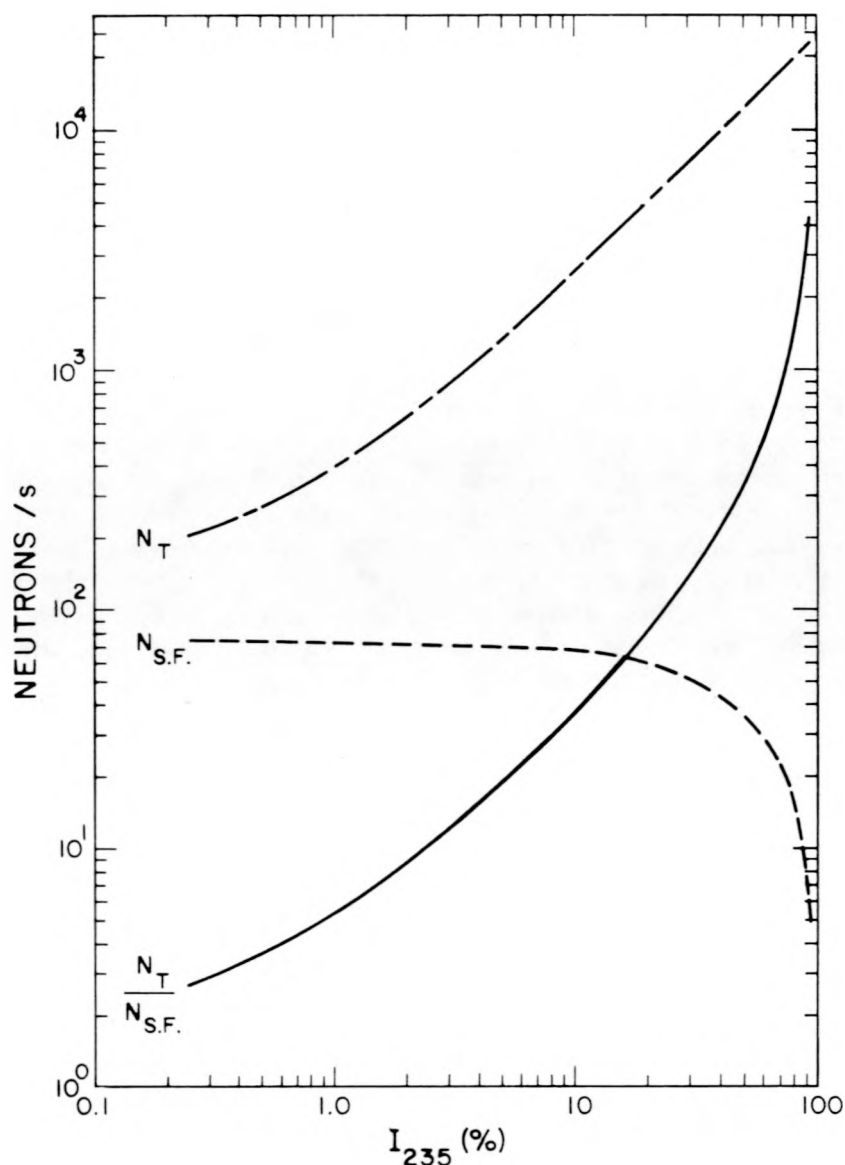


Fig. 31.

Neutron emission rates for a 5-kg mass of UF_6 vs ^{235}U enrichment. The dot-dashed curve labeled N_T is the total number of neutrons emitted per second. The dashed curve labeled $N_{S.F.}$ is the number of spontaneous fission neutrons emitted per second. Solid curve is the (mass-independent) ratio of total neutrons emitted to spontaneous fission neutrons emitted.

penetrates matter relatively easily (and thus is difficult to shield) and the detectors can be made up of large arrays of simple detectors.

Some work on the detection of low-density, large-area sources already has been done. Measurements of neutron fluxes near and in the operating diffusion cascade at Oak Ridge have been reported by Walton.¹⁷ Field measurements supported by calcula-

tions will be required to test the principle of using area neutron monitors to monitor the cascade enrichment for centrifuge facilities.

The proposed roof-top monitors might be placed in pairs; one at the product end and the other at the tails end of each "train." (A train consists of six centrifuge cascades.) The measured neutron emission rates could then be compared (correlated)

between product and tails detectors for each train and between product detectors and tails detectors for different trains. The detector plus background shield could consist of a 1.2 x 1.8-m slab of polyethylene, 0.3 m thick and with 64 ^3He tubes embedded in it. The detector would weigh ~ 636 kg (1400 lb) with a load of 294 kg/m^2 (60 lb/ft^2). The

electronics required to operate the detectors are relatively simple. Data recording and transmission to a central processing station outside the fence would be more complex. Each detector would cost $\sim \$50\text{k}$. The size and number of ^3He tubes might be reduced without sacrificing too much sensitivity and would lower the cost accordingly.

VI. STANDARDS AND MEASUREMENT CONTROLS

A. HgI_2 Low-Energy Photon Detectors (J. W. Tape)

Room-temperature semiconductor detectors with moderate energy resolution have potential safeguards applications where portability or ruggedness are required. One candidate detector material, CdTe, was reported on previously (Ref. 1, pp. 27ff). CdTe detectors have a relatively high leakage current and are therefore useful only for the detection of photons with energies $> 100 \text{ keV}$. On the other hand, the small useful depletion depth (owing to charge trapping effects) of CdTe limits its use above a few hundred keV. HgI_2 detectors suffer from a similar limitation on thickness because of charge collection difficulties; however, the low leakage current of HgI_2 crystals allows their use as a low-energy (x-ray) detector with possible application in portable L-edge densitometers or x-ray fluorescence units for total elemental analysis of well-characterized, product-type nuclear materials.

An experimental HgI_2 detector obtained from EG&G/Santa Barbara, was evaluated for safeguards application. The detector, 16 mm^2 in area and 0.170 mm thick, operates at a bias of 400 to 600 V. A low noise, TC-161D Ge(Li)-type preamplifier was purchased from Tennelec, Inc., to be used in conjunction with this detector. A spectrum from a ^{241}Am source obtained with this system is shown in Fig. 32. Pulse shape analysis is not an important requirement for HgI_2 when low-energy x rays are detected and it was not employed here (see Ref. 1, p. 27). The detector can partially resolve the neptunium L x rays at 13.944 keV and the multiplet at 17.8 keV , making it a potential replacement for NaI(Tl) detectors and gas proportional counters in this energy region. The high average Z and density ($\rho \sim 6.7 \text{ g/cm}^3$) of HgI_2 result in a high efficiency per unit

volume which helps to compensate for the small detector volumes available at this time.

Evaluation of mercuric iodide has begun, but much work remains to be done, especially in the areas of producing reliable, rugged detectors. Reference 20 contains a review of the state of the art for both HgI_2 and CdTe.

B. Development of LSI-11 Interfaces (G. Eccleston, S. Bourret, and E. Gallegos)

The purpose of the hardware development is to supplement commercially available interfaces by providing increased capabilities for safeguards instrumentation. The construction of hardware devices is accompanied by the development of FORTRAN-callable assembly language software drivers. The software enables the interface to be incorporated easily into a system by anyone with a knowledge of FORTRAN.

A scaler board has been completed. It consists of a quad board and plugs directly into the Q-bus of the LSI-11 microcomputer. The scaler board has six separate channels, each having a 24-bit (16, 777, 215) count capacity. The scaler inputs are TTL-compatible and operate in a ripple mode. A count is registered on the transition of the pulse from a low to a high state. The minimum pulse width for counting is 0.2 s and the pulse pair resolution of any scaler is 0.1 s . A universal clear strobe is used to zero all the counters simultaneously. Control of the individual scalers to enable/disable counting or to read their contents is accomplished through a control status register. An additional test feature permits scaler incrementing to test the counting and readout functions of the counter.

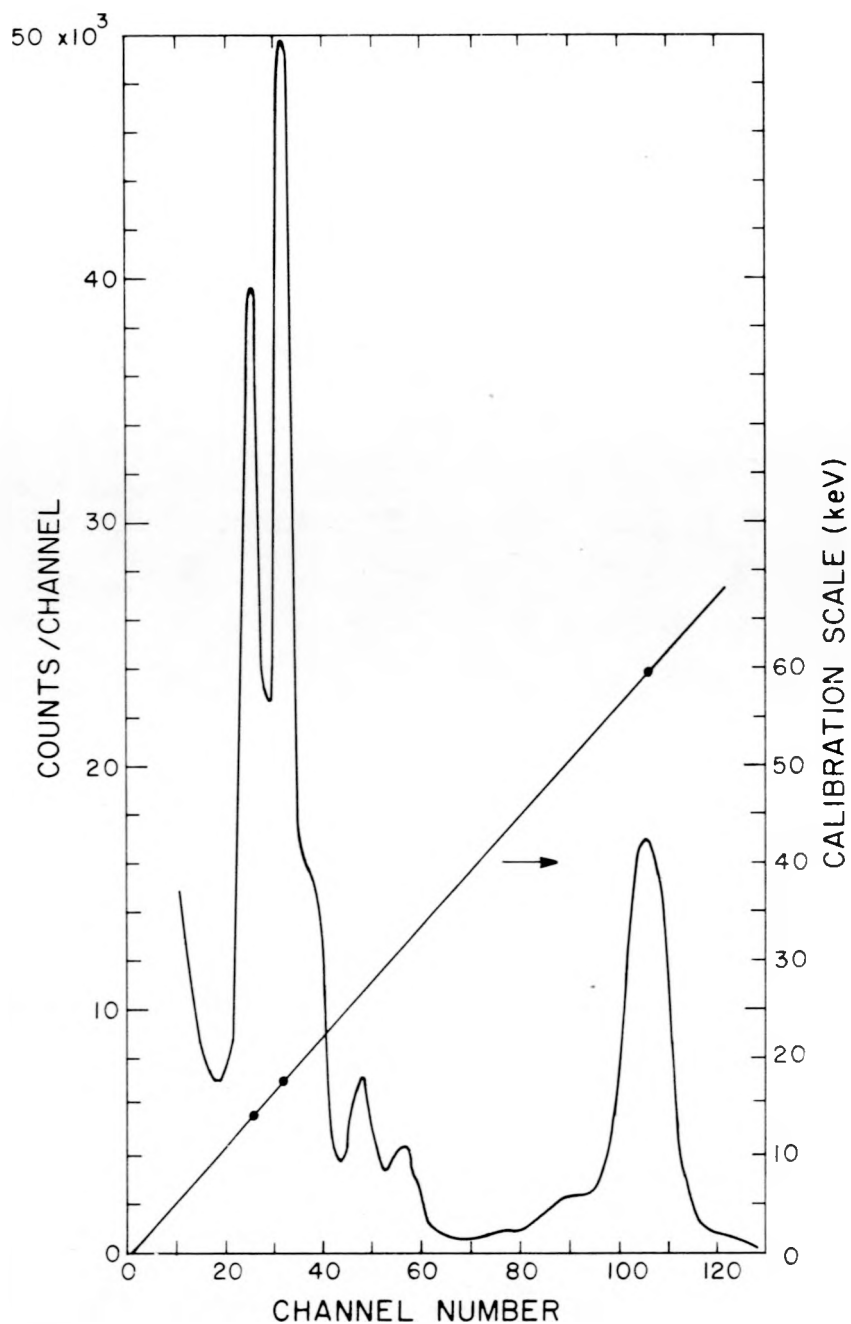


Fig. 32.
Spectrum obtained from ^{241}Am source by experimental HgI_2 detector.

The FORTRAN-callable scaler driver accomplishes all communications required to control the scaler board. Complete control of the scalers is obtained with the following four FORTRAN calls:

- | | | | |
|-------------------|---|----------------|--|
| 1) CALL CSCAL | ; | CLEAR SCALERS | |
| 2) CALL ESCAL (X) | ; | ENABLE SCALERS | |
| 3) CALL RSCAL | | | |
| (N, DATA) | ; | READ A SCALER | |
| 4) CALL TSCAL | | | |
| (INC) | ; | TEST INCREMENT | |
| | | A SCALER | |

The clear scaler (CSCAL) call requires no input parameters and all units are cleared. The enable command requires an octal pattern (x) to specify which scalers are to be enabled (1) or disabled (0). For example, octal value 35, which is binary (011 101), will enable scalers 5, 4, 3, and 1. The read call (RSCAL) obtains the count from only one scaler (N) per call and the data are double-integer format (INTEGER #4). Scaler testing is accomplished with the TSCAL call, where INC is the integer number of times the scaler is to be incremented. When scalers are tested they must be disabled from counting (CALL ESCAL (0)).

C. Californium Shuffler Source-Transfer Testing (G. W. Eccleston, J. Swansen, S. Bourret, E. Gallegos, and D. Garcia)

Two experimental test beds (TB1, TB2) have been constructed to study the source transfer system used in the ^{252}Cf Shuffler. The objective of the study is to simulate a 10-yr life on the mechanical transfer system and to select materials and equipment capable of supporting this long-term operation. The test beds simulate the source-transfer system to be used in the SRP Shuffler (TB1) and in the Fluorinel Reprocessing Plant Shuffler (TB2) (see Pt. 1, Secs. IV-C and D).

A previous Shuffler used a Teflon transfer tube to guide the ^{252}Cf source between the irradiations and storage positions. The Teflon flaked in this Shuffler and contributed to failure of a LED position sensor by blocking the transmittal of light. At present, a Teflon tube (TB1) and a 316 stainless steel tube (TB2) are being used to guide the source. In addition, a stepping motor (TB1) and a pancake motor (TB2) are being tested individually to determine their comparative capabilities for rapidly and accurately transferring a source between two specified positions. The reliability of the motor optical encoders is being tested, and more reliable position sensors are being developed.

The SRP Shuffler has a source-transfer distance of 1032 m. The stepping motor transfer time (TB1) varies between ~ 0.4 and 1.0 s, depending on the ramp rates for starting and stopping. The test source is a stainless steel copy of the actual ^{252}Cf source and is not radioactive. This source is attached to a Teleflex cable by an oil-sintered coupling and is

driven by a Teleflex gear through a bushing connected to the shaft of the stepping motor. So far, more than 100 000 cycles (264 km of travel) have been completed without problems. A 10-yr life under continual full operational conditions requires 30 samples/day, 50 cycles/sample for 365 days/yr—a total of 5.5 million cycles.

The test bed for the Fluorinel Reprocessing Plant Shuffler (TB2) requires a Teleflex cable ~ 4 m long. The irradiation scheme varies, depending on whether spent fuel or waste canisters are being assayed. In both situations, an initial fast transfer of 180 cm is required between the storage position and the start of the irradiation position. Transfer times as low as 0.45 s have been obtained with the dc pancake motor. In this system the motor shaft is connected to the Teleflex gear wheel through a set of ball bearings which has less friction than the bushing arrangement on TB1 and contributes to the faster transit time. Testing has been directed principally toward the motor interface, Teleflex position sensors, and long-term cycling of the cable, with no source mockup attached. The cable, which is self-lubricating, has been run $>1\text{M}$ cycles (3657 km) with no appreciable wear in the stainless steel tubing. Not surprisingly the Teleflex gear motor shows signs of wear along the edges of the helical teeth because of the large number of cycles completed under extreme accelerating and decelerating conditions.

D. Neutron Well Coincidence Counter Electronics Fabricated by EG&G (C. Spirio and J. Swansen)

An updated version of the neutron well coincidence counter-shift register electronics packaging is almost complete. This second-generation system includes a microprocessor (Motorola 6800) that serves as an interface between the shift register/counter electronics and the HP-97 calculator, an external HP-97 calculator (with hard-copy printer) that extends the calculation ability of the microprocessor, an RS-232 interface that allows data stored in the microprocessor memory to be transmitted to a computer or terminal. This new electronics package uses the latest state-of-the-art, multiwire, printed cards for the high-density packaging of the shift-register system and microprocessor system.

Features carried over from the first version are: highly stable 2-kV power supply, a six-channel amplifier-discriminator signal conditioning board, a display panel for data, and the essential front and rear panel controls and test points.

The units are undergoing final assembly, with checkout and testing to follow in June 1978. Three of the units are planned for IAEA applications to replace the original portable shift-register units now in use by the IAEA.

E. Evaluation of SILENA Multichannel Analyzer (K. Kaieda, J. L. Parker, and S. T. Hsue)

We have evaluated the SILENA MCA to study (1) the resolution of the full-energy spectral peaks and (2) the losses to the full-energy spectral peaks as a function of count rate. The SILENA analyzer has a built-in amplifier and high-voltage supply suitable for high-resolution detectors, making it a full spectroscopic package. It has the pile-up rejection feature for high count rate cases and can be either dc or dcr (ac coupled with restorer) coupled with the ADC input. In addition, it is compactly packaged for inspection purposes and is complete with cassette tape for data storage.

The following measurements were made on several fixed sources (^{133}Ba , ^{54}Mn , ^{22}Na) and a movable ^{57}Co source in front of a Ge(Li) detector by using the built-in amplifier and high-voltage supply of the SILENA unit. The count rates to the amplifier were varied from 2700 to 30 000 cps by changing the position of the movable source. The resolution was measured at full width half maximum (FWHM) for the 1274-keV energy peak of ^{22}Na with an approximate precision of $>10\%$. Full-energy spectral peak areas were determined by subtracting straight-line backgrounds for the 356-keV (^{133}Ba), 511-keV (^{22}Na), 834-keV (^{54}Mn), and 1274-keV (^{22}Na) peaks. The precision of the peak areas was $>1\%$.

Figure 33 shows the FWHM resolution in keV and the full-energy peak areas for the various energies normalized to that observed at 2700 cps as a function of different count rate. The most important

observation in that the FWHM increases rapidly with increasing count rates, and at 30 000 cps, it is between 8 and 11 keV. Also shown in Fig. 33 are the losses to the full-energy spectral peak when either dc or dcr coupling is used. Losses should be the same for all the peaks, and for dcr coupling the losses follow the naturally expected trend quite closely although the losses for the different peaks are not quite as constant as expected. With dcr coupling the fractional losses are more severe and, as the rate increases, the lowest energy peak loses an increasingly larger fraction than the high-energy peaks. The reason lies in the degradation of FWHM resolution noted above, along with increasingly severe low-energy tailing. The combination of these two factors makes it difficult to define a region of interest that includes all (or even a constant fraction) of the peak areas.

Figure 34 gives results taken with an amplifier time constant of $2\ \mu\text{s}$, dc coupling, with and without pile-up rejection. The resolution deteriorates more rapidly than with the $1\text{-}\mu\text{s}$ time constant (Fig. 33), and the pile-up rejection does not improve the resolution within precision. The amplifier was blocked whenever the count rate exceeded 20 000 cps. Obviously, the rate-related effects are far more severe than those obtained with $1\text{-}\mu\text{s}$ time constants. Similar results are obtained on the four SILENA units currently at LASL.

The apparent cause of most of the degradation of resolution and peak shape with increasing rate lies in the limitations of the built-in amplifier, for when a high-quality external spectroscopy amplifier is used (dc coupling) the FWHM resolution increases by only $\sim 10\%$ for rates up to 30 000 cps, with little or no tailing.

In summary, the SILENA analyzer is a useful, self-contained system for gamma-ray spectroscopy. However, in view of its limitations, we suggest that $1\text{-}\mu\text{s}$ amplifier time constants and dc coupling be used and that gross count rates be restricted to 10 000 cps or less if accurate quantitative results are required. If higher rates are to be encountered, the use of a high-quality separate amplifier will improve the quality of the acquired spectra and simplify the accurate extraction of the desired information.

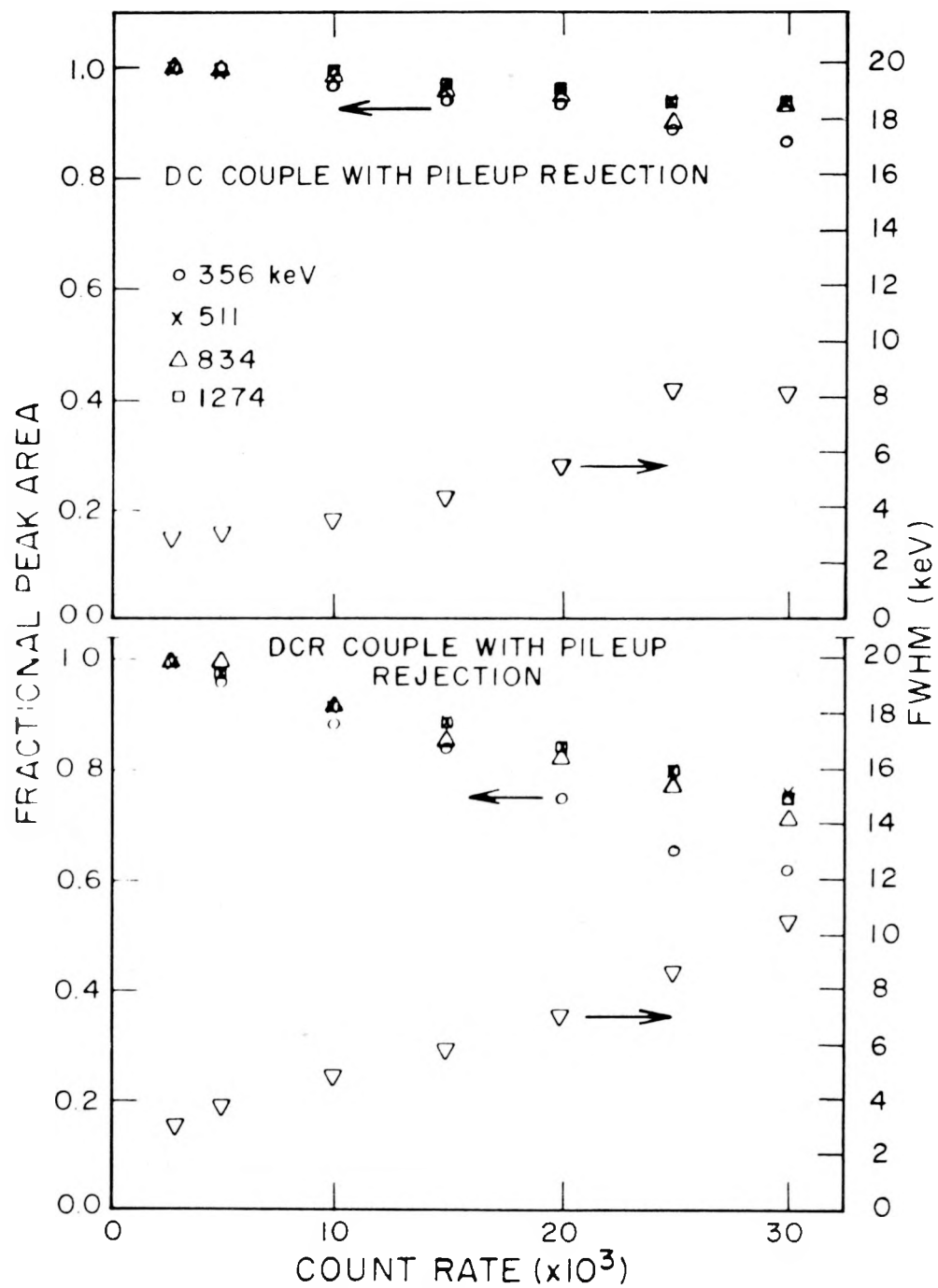


Fig. 33.
Fractional peak areas and FWHM in keV as a function of count rate for an amplifier time constant of $1 \mu s$.

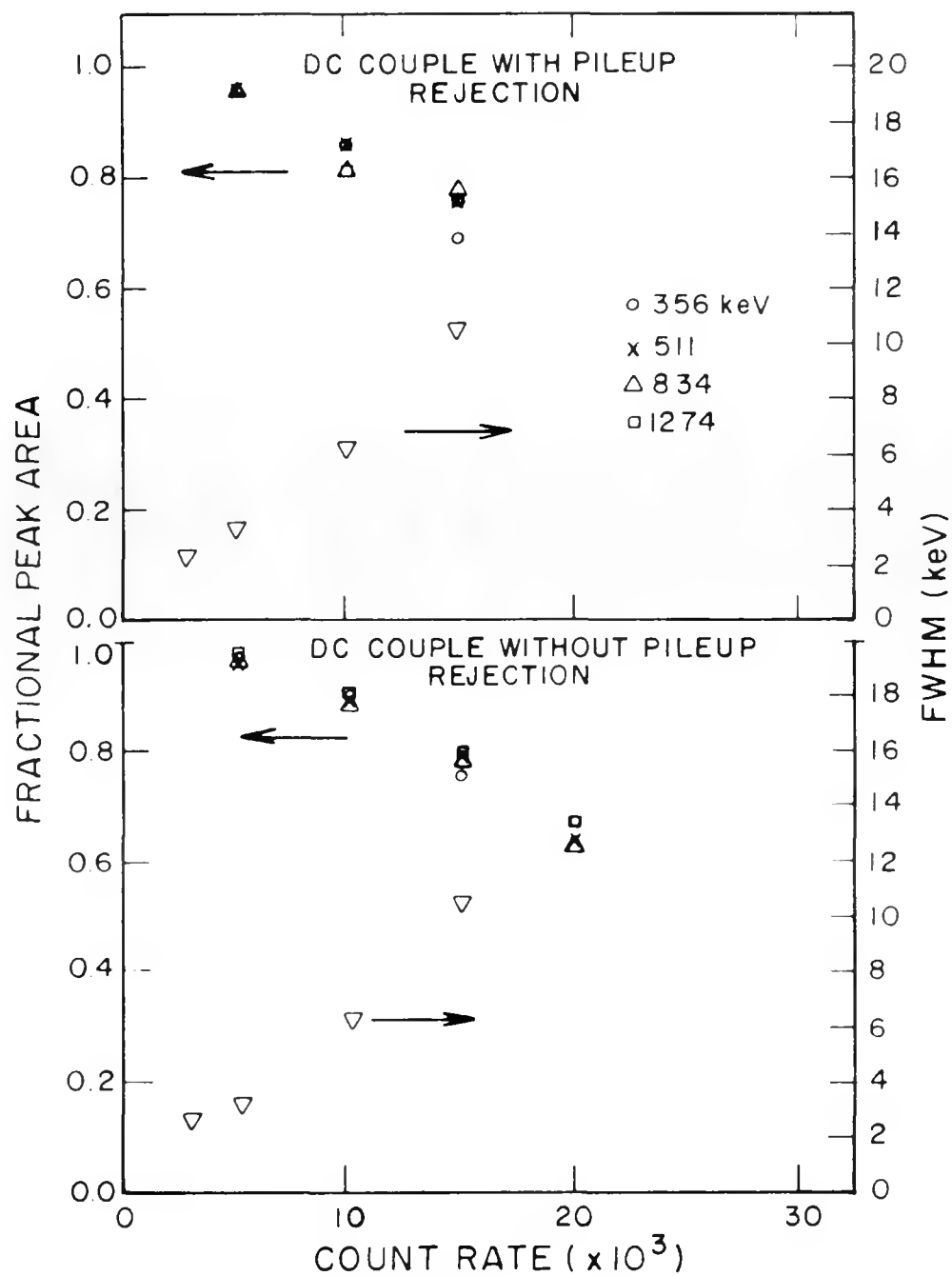


Fig. 34.
 Fractional peak areas and FWHM in keV as a function of count rate for an amplifier time constant of $2 \mu\text{s}$.

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

1. DYMAC Plutonium Solution Assay System

A system based on transmission-corrected gamma-ray analysis is being developed for on-line analysis of plutonium solution samples for the DYMAC project. Five such assay stations will be installed in the new plutonium facility under the DYMAC project. Each station has its own detection system and data reduction computer which transmits assay results to the Eclipse accounting and materials control computing system. Each station also will communicate with a number of load cells and with a balance used in the computation of plutonium concentration. The stations will be calibrated and program-loaded by a movable control cart containing a microprocessor display system with a terminal, paper-tape reader and monitor scope. This operational strategy provides protection against tampering with the data reduction systems of these measurement stations.

The initial system with one assay station and the control cart has been completed and additional assay units are being assembled. The cart is shown in Fig. 35 and the assay unit in Fig. 36. Preliminary tests indicate that the system will meet all engineering expectations.

2. Segmented Gamma Scanner for the IAEA

A segmented gamma scanner (SGS) was completed and sent to Casaccia, Italy, for test and evaluation by the IAEA. A central processing unit is used for two assay units; one unit can size samples and the other can be used for large barrels. LASL will assist in the setup of the instrument at Casaccia and the training of IAEA personnel in its use.

3. Barrel-Scanner Modifications

Preliminary engineering has started for a more compact, self-contained gamma-assay unit for barrels as large as 55 gal. The unit will be similar to the SGS barrel-handler that was shipped to the IAEA but will have its own processor and display system.



Fig. 35.

Plutonium solution assay system control cart containing microprocessor display system with a terminal, paper-tape reader, and monitor scope.

The mechanical portions will be microprocessor-controlled, with a minicomputer used for data collection, analysis, and computations. This unit will require less floor space than the present SGS barrel system.

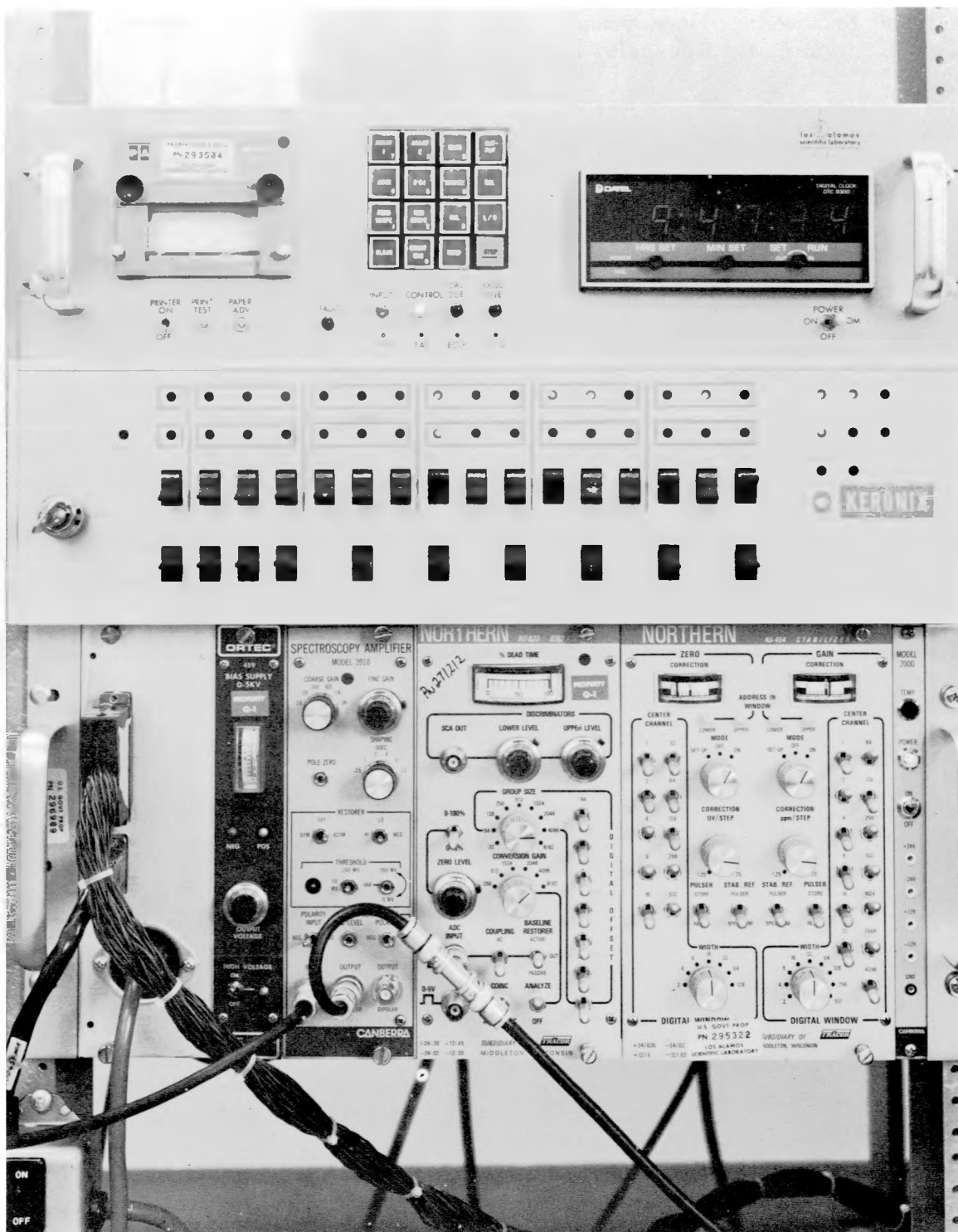


Fig. 36.
Electronic rack for the DYMAC plutonium solution assay system.

VII. TRAINING AND TECHNOLOGY TRANSFER

A. Nondestructive Assay of ^{235}U in Ten 55-gal Barrels (T. W. Crane and E. Medina)

The prototype barrel-counter test setup (Ref. 8, p. 10) was used to assay for Group CMB-8 ten 55-gal barrels containing HEU (93.15% ^{235}U dispersed in combustible waste. The prototype NDA unit employs thermal-neutron interrogation and delayed neutron counting to measure fissile materials in a ^{252}Cf Shuffler configuration. Because no 55-gal-barrel calibration standard was available at the time, the results were extrapolated from an assay of a 30-gal-barrel standard (Ref. 6, pp. 52-54) located in the center of the chamber. This central positioning was expected to result in a low bias for the larger barrels because the material in the bottom of the chamber yields a lower response (Ref. 7, p. 12). Designs for future systems will minimize the position-dependent biases (Ref. 8, p. 10).

Contents will be recovered from the waste barrels that were assayed. The first steps in the process are incineration and subsequent assay of the ash with the CMB-8 random driver. As recovery measurements become available, a calibration curve can be obtained and future assays of combustible waste can be made with improved accuracy.

B. Accelerator Activities (J. D. Brandenberger and E. L. Adams)

We have used the Q-1 Van de Graaff accelerator-based small sample assay station (SSAS) for two routine small sample assay projects as well as in preliminary development of a set of large working standards for use with the CMB-8 random driver.

This random driver is being used at the LASL uranium recovery facility for the assay of bulk uranium samples. It needs a set of large ($\sim 10^4 \text{ cm}^3$) working standards with the same chemical composition as the materials being assayed. CMB-8 has selected several such samples to be made into working or secondary standards. Our procedure is to divide each large sample into 50 to 60 small samples, each of which is assayed on the SSAS. After we have

made a uranium determination on each of the small samples, the material will be returned to the larger container, which will then be a secondary standard. The precisions will be good because the statistical uncertainty on the reconstituted sample is the same as if the sum of the assay counts from each sample had been taken on the large sample itself. The use of small samples will result in minimal biases from the geometrical effects of neutron attenuation and multiplication. We effect an accuracy of 1% for the secondary standards. A set of nine small standards has been ordered from CMB-1. The large standards we develop will be available to support the anticipated large-sample work of Group Q-1.

Work on the assay of large samples as (1) a backup assay system for the random driver and (2) to support assay and standards development for the californium Shuffler is reported in Ref. 6, p. 51. A neutron flux-flattening lead and steel cavity, required for the large-sample assays, has been designed and is to be fabricated soon. It will increase the flux at the sample and give a more uniform irradiation over the sample volume.

C. Alternative Approach for Determining an Assay Value from a Calibration Function (T. W. Crane)

For NDA systems in general and particularly for neutron assay systems such as the fuel rod scanner, the random driver, and the ^{252}Cf Shuffler, the assayed SNM mass is based upon calibration data. The calibration procedure consists of assaying a set of standards in the instrument to determine the system's response as a function of the SNM mass. A calibration function is used to relate the system's response (R) to the mass (M) of the sample being assayed: $R = f(M, a_1, a_2, \dots, a_p)$, where f is the calibration function and a_1, a_2, \dots, a_p are parameters determined by a best fit to the calibration data.

Once the NDA instrument is calibrated, the SNM mass of a sample is obtained from the observed response by using the inverse of calibration function:

$M = F(R, a_1, a_2, \dots, a_p)$. The estimated uncertainty in the mass, S_M , can be calculated by the formula:

$$S_M^2 = \left(\frac{\partial F}{\partial R} \right)^2 S_R^2 + \sum_{i=1}^p \left(\frac{\partial F}{\partial a_i} \right)^2 S_{a_i}^2 + \sum_{i=1}^p \sum_{\substack{k=1 \\ i \neq k}}^p \text{cov}(a_i, a_k) \left(\frac{\partial F}{\partial a_i} \right) \left(\frac{\partial F}{\partial a_k} \right),$$

where S_R is the uncertainty in the response R , S_{a_i} is the uncertainty in the parameter a_i , and $\text{cov}(a_i, a_k)$ is the covariance between the parameters a_i and a_k . A complete discussion on calibration and error estimation for NDA systems is given in Ref. 21.

Implicit in the above discussion is the assumption that the calibration function can be inverted to obtain a formula for the mass. And, in practice, an invertible calibration function is selected; however, this requirement unnecessarily restricts the use of some otherwise suitable calibration functions. Three potential calibration functions are given in Table V. The first two functions are discussed in Ref. 21 for NDA applications, and Ref. 22 gives a physical interpretation for the second function. The third function in Table V is an extension of the second, with an additional parameter to include multiplication effects. This third function gives good fits to a series of calculations of the response for samples containing 0-22 kg ^{235}U . Neither of the first two functions could provide an adequate fit over the entire mass range. As noted in Table V, the third function does not have an inverse that can be expressed in closed form.

When the calibration function cannot be inverted, an alternative approach for determining the mass M of the sample being assayed is to find the value of M by a systematic guessing procedure for which the calibration function $f(M, a_1, a_2, \dots, a_p)$ yields the

observed response R . A variety of iterative schemes employing extrapolation or interpolation can be used to converge rapidly on the desired mass value (see Ref. 23). The numerical algorithms are quite simple and work just as well for functions that can be inverted.

The estimated uncertainty in the mass is obtained by using the familiar calculus "chain rule" to obtain the partial derivatives of the inverse function (F) in terms of the partial derivatives of the calibration function (f). The relationship between the derivatives is given by:

$$\frac{\partial F}{\partial R} = \left(\frac{\partial f}{\partial M} \right)^{-1}$$

and

$$\frac{\partial F}{\partial a_i} = - \left(\frac{\partial f}{\partial M} \right)^{-1} \left(\frac{\partial f}{\partial a_i} \right).$$

These two mathematical identities for the partial derivatives can be substituted into the previous expression for the mass uncertainty. The resulting formula for the uncertainty in the mass, S_M , becomes:

$$S_M^2 = \left(\frac{\partial f}{\partial M} \right)^{-2} \left\{ S_R^2 + \sum_{i=1}^p \left(\frac{\partial f}{\partial a_i} \right)^2 S_{a_i}^2 + \sum_{i=1}^p \sum_{\substack{k=1 \\ i \neq k}}^p \text{cov}(a_i, a_k) \left(\frac{\partial f}{\partial a_i} \right) \left(\frac{\partial f}{\partial a_k} \right) \right\}.$$

Thus, the calibration function need not be inverted to obtain the assayed mass value and its uncertainty.

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

Table VI 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 V

EXAMPLES OF POTENTIAL CALIBRATION FUNCTIONS AND THEIR INVERSE FUNCTIONS

Calibration function (f)	Inverse function (F)
1) $R = a + bM + cM^2$	$M = ([b^2 + 4c(R - a)]^{1/2} - b)/2c$
2) $R = a(1 - e^{-bM})$	$M = -\frac{1}{b} \ln(1 - R/a)$
3) $R = a(1 - e^{-bM})/(1 - cM)$	$M = ?$

TABLE VI
VISITS AND TECHNICAL DATA TRANSMITTALS
(Group Q-1)

<u>Technical Data Transmittal</u>		
<u>Date (1978)</u>	<u>Organization</u>	<u>Subject</u>
1-4	Rockwell/Hanford	Least squares fitting procedure for neutron coincidence counter used during In-Plant Safeguards School.
1-9	ANL	Drawings for SGS.
1-9	IRT Corp.	Blueprints and equipment lists for LASL random driver.
1-10	DOE/OSS	Copies of "Gamma-Ray Measurements with the Segmented Gamma Scan."
1-16	Nuclear Engineering Dept., Univ. of Michigan	Consultation on irradiation history correction in the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio method to determine burnup.
1-17	Union Carbide	Specifications for the SGS.
1-19	Canberra	La-7059-M, Operator's Manual, SGS.
1-26	ISPRA (Reilly)	LA-6849-PR, applications of absorption edge densitometry.
1-31	Union Carbide	Bid specification for automated SGS assay system.
1-31	ANL	LASL drawings for HEDL and FFTF fuel rod handling device.
2-1	DOE/OSS	Copy of the Safeguards Technology Training Program manual "In-Plant Nondestructive Assay Instrumentation."
2-7	National Nuclear Corp.	Design documentation on the compact HLNCC.
2-7	ACC	Short-term action items: transfer of ^{252}Cf source; calculate radiation dose estimates; operational sequence for data handling and software interface.

TABLE VI (cont)

<u>Date (1978)</u>	<u>Organization</u>	<u>Subject</u>
2-16	ORNL	Technical data on the effects of spiking on NDA of SNM.
2-18	IRT Corp.	Information on Shuffler assay system, LA-6007-M, LASL drawing 68Y-155522.
2-24	Battelle	Information on Segmented Gamma Scan Assay System, LA-7059-M, drawings.
2-28	CMB-8	Results of gamma-ray assay for ^{235}U in four cans of rags.
3-3	NBS	LA-7059-M, "Gamma-Ray Measurements with the Segmented Gamma Scan." design.
3-3	Jomar Systems	Design package for random driver.
3-3	Battelle	Drawings for barrel-handling system for SGS unit.
3-28	Westinghouse Hanford	Cost breakdowns—electronics for random driver.
4-17	National Nuclear Corp.	Design data on new random driver for plutonium.

Visitors to LASL Group Q-1, Domestic

<u>Date (1978)</u>	<u>Name & Organization</u>	<u>Subject</u>
1-11	G. Entine/Radiation. Monitoring Devices, Inc.	CdTe detectors.
1-11/1-12	W. Echo/ACC H. Lawroski/ACC R. Swanson/ACC J. Jones/DOE/ID W. Spalding/Parsons K. Parsons/Parsons D. Silliker/Parsons	Neutron Interrogator meeting.

TABLE VI (cont)

<u>Date (1978)</u>	<u>Name & Organization</u>	<u>Subject</u>
2-6/2-8	K. McMurdo/Savannah River P. Makahon/Savannah River	SRL densitometer.
2-10	G. Bates/Sandia	Organization of Sandia Training Program.
3-17	L. Scheinmann/State Dept.	Safeguards overview.
3-20	J. Burkhardt/Savannah River	Enrichment plant safeguards.
3-21	J. Krupa/Idaho Falls K. Neishmidt/Idaho Falls	Waste measurements.
4-25	L. Brenner/DOE/OSS	Safeguards discussions.

Group Q-1 Travel

<u>Date (1978)</u>	<u>Name</u>	<u>Destination</u>	<u>Subject</u>
1-10	C. Spirio	ANL/Canberra	SGS.
1-16	J. Tape	Ft. Lauderdale	ASTM Committee.
2-7	C. Hatcher	Dallas	ISPO Coordinators meeting.
2-10	H. Menlove	Salt Lake City	Presented paper: 2nd Int. Conf. Nondestructive Evaluation in the Nuclear Industry.
2-11	S. T. Hsue	Ann Arbor, Mich. Traverse City, Mich. Zion, Ill. Chicago Jackson, Miss.	J. Lee, Univ. of Michigan; Big Rock Point, spent fuel assay; Zion Power Station, spent fuel assay; Commonwealth Edison, spent fuel assay; Consumers Power, spent fuel assay.
2-11	J. Parker	Salt Lake City	Presented paper: 2nd Int. Conf. on Nondestructive Evaluation in the Nuclear Industry.
2-12	D. Langner	Salt Lake City	2nd Int. Conf. on Nondestructive Evaluation in the Nuclear Industry.

TABLE VI (cont)

Date (1978)	Name	Destination	Subject
2-13	J. Phillips	Traverse City, Mich. Zion, Ill. Chicago Jackson, Mich.	Big Rock Point, spent fuel assay; Zion Power Station, spent fuel assay; Commonwealth Edison, spent fuel assay; Consumers Power Co., spent fuel assay.
2-14	C. Hatcher	DOE/SS, Washington, D.C. Commonwealth Edison, Chicago Consumers Power, Jackson, Miss.	Safeguards programs. Spent fuel assay. Spent fuel assay.
2-14	R. Walton	Washington, D.C.	Review of topics, Tokai trip.
2-24	H. Menlove	Tokai-mura, Japan	Discuss measurement technology for safeguarding plant.
2-27	K. Johnson	Oak Ridge	Random driver assay unit.
3-8	J. Tape	Oak Ridge	Enrichment plant—safeguards meeting.
3-13	T. Canada	Anaheim, Cal.	Invited paper, ACS meeting.
3-16	D. Langner	Savannah River Lab. Augusta, GA	Alternative fuel cycles.
3-19	S. Beach	Zion, Ill. Jackson, Miss.	Spent fuel assay. Spent fuel assay.
3-23	K. Johnson	Las Vegas, Nev.	INMM Standards Committee.
4-5	M. Baker J. Tape	Albuquerque, N.M.	Sandia, enrichment plant safeguards meeting.
4-12	S. Beach	Big Rock Point, Traverse City, Mich.	Spent fuel assay.
4-22	H. Menlove	Allied Chemical WEC, Idaho Falls	ICPP neutron interrogator project.
4-23	S. Beach	Salt Lake City Idaho Falls Chicago	Possible instrumentation vendors. ICPP Shuffler. Zion reactor, spent fuel assay.
4-24	G. Eccleston	Allied Chemical WEC, Idaho Falls, ID	ICPP neutron interrogator project.

TABLE VI (cont)

Date (1978)	Name	Destination	Subject
4-24	J. Phillips	Taipei, Taiwan	Taiwan research reactor.
4-25	S. T. Hsue	Zion Power Station, Chicago	Spent fuel assay.

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. Fast Critical Assembly Plutonium Storage Vault Monitor (J. T. Caldwell, P. E. Fehlau, and A. A. Robba)

The international safeguarding of any FCA facility probably will include a vault area that contains 1000 kg or more of plutonium in the form of plates or other fuel mockup. Consequently, the study of different reactor configurations will involve considerable traffic between the vault and the other facility areas.

One possible way to detect gross diversions of plutonium from such a vault is to place an array of neutron detectors on the vault's walls and ceiling. A single neutron detector would not sense a distant diversion of plutonium, but an array of detectors

monitoring the entire vault would detect diversions from any arbitrary location.

In a storage vault we used a portable ^3He neutron detector to make measurements typical of those to be expected at an FCA. The detector could measure both fast (efficiency-optimized for fission spectrum) and thermal-neutron fluxes. Figure 37 is a schematic of the vault and gives measured thermal and fast-neutron flux values for various locations. All values are given in multiples of the cosmic-ray background at 2000-m (6500-ft) elevation (Pajarito Site, Los Alamos). The plutonium storage racks were solid concrete "tables" ~1.4 m high and 1.4 m thick. Holes in each table at ~0.6-m intervals permit the insertion of a plutonium-containing canister.

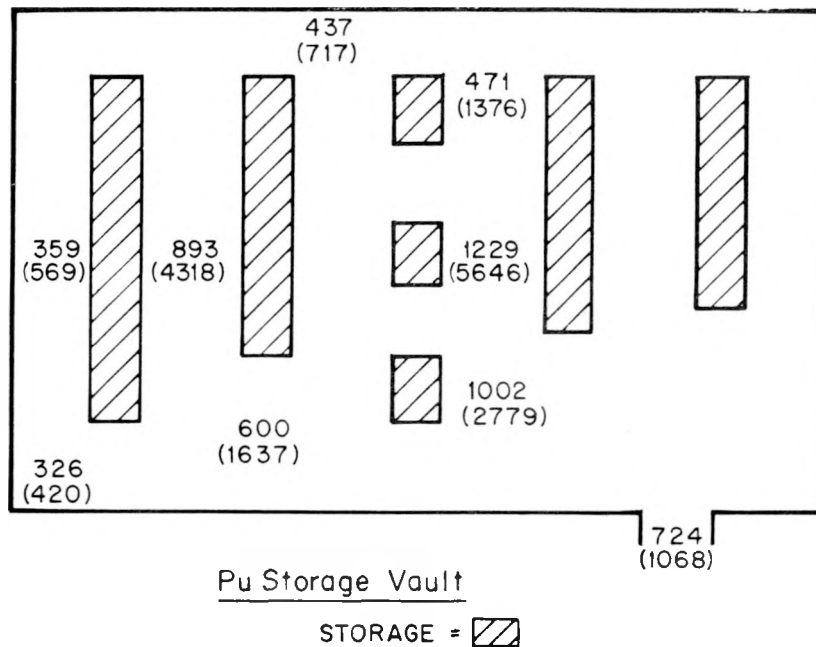


Fig. 37.

Relative thermal and fast neutron backgrounds measured in a typical plutonium storage vault.

This highly moderated geometry (and the thick concrete and surrounding dirt walls, floor, and roof) results in a reasonably uniform thermal-neutron flux around the vault. Measurements were made only ~1.4 m from the floor. At this level, thermal fluxes varied only by a factor of 3.8 at the locations indicated in Fig. 37. At the ceiling level (~3 m), the thermal flux probably would be even more uniform. The corresponding fast fluxes varied by a factor of 13.4 in the same locations.

On the basis of our initial measurements, we propose an evaluation of a storage-vault monitor consisting of a bare ^3He detector grid (sensitive only to thermal flux) on the ceiling and a limited number of detectors at lower wall levels. More systematic measurements within such a vault (with experimental movement of plutonium) are required to develop and determine the sensitivity of such a system.

The system has key limitations. For instance, the common practice of storing startup neutron sources (Pu-Be or ^{252}Cf) in such vaults would have to be eliminated because these sources have the neutron

output equivalent of tens to hundreds of kilograms of plutonium. The uncontrolled removal or insertion of such sources in a vault would totally circumvent the system. In fact, to be effective this system would require strict control over all plutonium removal and storage within the vault and a knowledge of the type and neutron output of materials being removed or stored. With strict entry/removal control, the system might have excellent sensitivity. With proper calibration, flux measurements at the $\leq 0.5\%$ credibility level could be made routinely with ^3He tubes. For a vault having a nominal storage capacity of 1000 kg plutonium, this would translate roughly into ~5 kg plutonium sensitivity.

This type of system probably will not be useful within the critical assembly area itself. There the flux is sensitive to the multiplication condition and can vary by an order of magnitude during shutdown conditions, depending on shield configurations, etc. Because of low spontaneous fission yields in the uranium isotope, the system would not be effective in monitoring uranium fuels.

B. Rapid NDA Scanning System for FCA Plutonium Fuel Verification (J. T. Caldwell, P. E. Fehlau, S. W. France, and A. A. Robba)

Because of the large number of fuel pieces in the inventory and their disposition inside reactor fuel elements, relatively rapid, collective inventory verification techniques are needed at FCAs. A specific system being investigated for the purpose is based on rapid gamma and neutron scanning of fuel-containing reactor drawers. The system is designed to scan one or two typical fuel drawers per minute with a sensitivity adequate to detect the presence or absence of a 2.54-cm-long segment of plutonium fuel plate in a drawer containing up to 2.84 m of effective plutonium fuel plate length, together with a variety of coolant, structural, and fertile mockup materials also in the drawer.

The technique assumes that the fuel plates are well characterized but does not require that all plates have the same ^{240}Pu , ^{241}Am , etc., isotopic content. An example of a typical fuel inventory is shown in Table VII. Table VIII shows the measured (or calculated) neutron and isotope-specific gamma intensities for a sample of the fuel plates listed in Table VII. The neutron and gamma line intensities are normalized to the values for plutonium/uranium/molybdenum fuel with 11.56% ^{240}Pu .

Table VIII shows that each of the five classes of fuel has a characteristic signature when the three independent quantities (total neutron, ^{239}Pu γ , ^{241}Am γ) are considered. Furthermore, on the basis of our experimental measurements with 25 separate fuel plates taken from among the first four classes listed in Table VII, the uniformity in the signature from plate to plate within a class is quite good, probably

because of the excellent quality control required in the manufacture of such plates. The plutonium/uranium/molybdenum plates have a neutron output consistent with 100% spontaneous fission and the plutonium/aluminum plates have an additional aluminum α, n component ($\sim 70\%$ additional for the 4.50% ^{240}Pu plates and $\sim 50\%$ additional for the 22.33% ^{240}Pu plates).

A conceptual design of the scanning apparatus required to perform the fuel drawer verification measurement is shown in Fig. 38. Fuel drawers are loaded on a conveyor belt that transports them first through a collimated fast-neutron detector and subsequently past a collimated intrinsic germanium or Ge(Li) gamma detector. For ease and accuracy of drawer identification, we recommend that each drawer be tagged with a grocery-store-type laser-scan identification label which could also contain information on the drawer's current plutonium content. A photocell/laser sensing system would read the label, and the drawer ID and the operator's statement of drawer content could be routed to a microprocessor or minicomputer-based data acquisition system.

The observed "signature" for each 25.4 mm of drawer length (as determined by the neutron output and the two isotopic gamma lines) would then be compared to the operator's stated values. A simple go-no-go indication (say, a green or red light) would be activated to indicate the drawer's passing or not passing the signature check. For a drawer to pass the signature check, each 25.4 mm of the drawer would have to check against the expected values.

The IAEA inspector would then return all "passing" drawers to the reactor assembly (or to a certified vault area) and further examine all nonpassing drawers. Count rates for the conceptual design shown in Fig. 38 (both neutron and gamma) are such that a statistically reliable signature for 25.4 mm of any plutonium fuel plate listed in Table VII can be obtained in <1 s. The signature for the combination of any two fuel types (two rows of fuel plates in a drawer) is also obtained statistically in <1 s per 25.4 mm.

A consideration of Table VIII shows that statistical accuracies on each attribute measurement need be only $\pm 10\%$ to verify a signature. For most cases an even poorer statistical accuracy would suffice.

TABLE VII

TYPICAL ISOTOPIC VALUES FOR VARIOUS PLUTONIUM FUELS IN AN FCA INVENTORY

Fuel Type	^{239}Pu (%)	^{240}Pu (%)	^{241}Pu (%)	^{241}Am (%)	g Pu/cm
Pu/Al	95.25	4.50	0.20	0.24	13.4
Pu/U/Mo	90.80	8.66	0.51	0.46	7.9
Pu/U/Mo	87.00	11.56	1.20	0.59	12.2
Pu/Al	74.20	22.33	2.86	1.80	13.9
Pu/U/Mo	68.70	26.40	3.39	2.19	14.8

TABLE VIII

**MEASURED NEUTRON AND GAMMA LINE RESPONSES FOR
A TYPICAL VARIETY OF FCA PLUTONIUM FUEL**

Fuel Type	^{240}Pu (%)	Relative Total Neutrons/cm	Relative Total ^{239}Pu (414 keV) γ/cm	Relative Total ^{241}Am (662 keV) γ/cm
Pu/Al	4.50	0.73 ± 0.07	(1.26 ± 0.13)	(0.42 ± 0.04)
Pu/U/Mo	8.66	0.47 ± 0.05	(0.67 ± 0.07)	(0.49 ± 0.05)
Pu/U/Mo	11.56	1.00 ± 0.10	1.00 ± 0.10	1.00 ± 0.10
Pu/Al	22.33	3.17 ± 0.32	0.96 ± 0.10	3.44 ± 0.30
Pu/U/Mo	26.40	(2.75 ± 0.28)	(0.95 ± 0.10)	(4.47 ± 0.45)

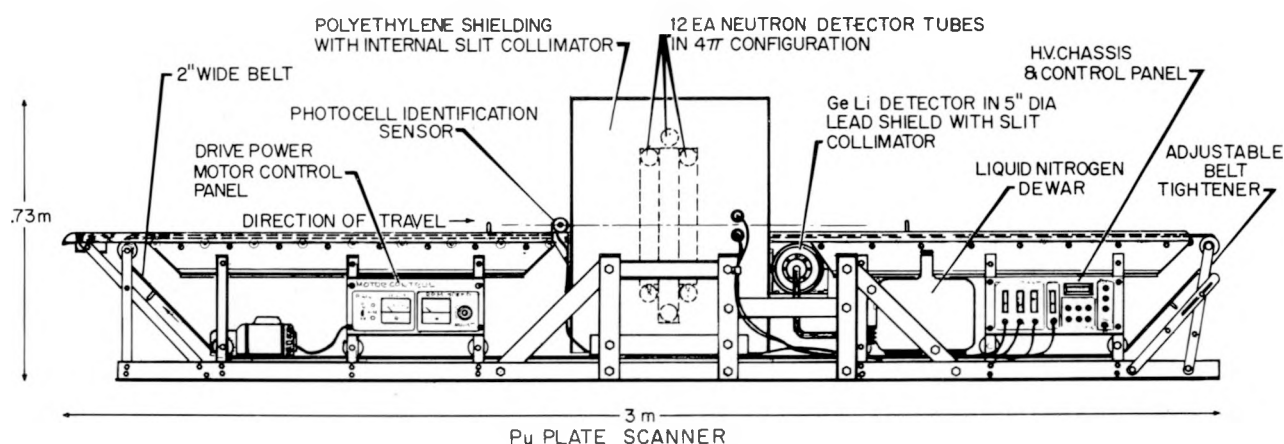


Fig. 38.

Schematic of proposed neutron/gamma rapid scanning instrument for FCA reactor drawers containing plutonium fuel.

The proposed system is more difficult to subvert than are verifications based on either neutron or gamma signatures alone. Subversion is also made more difficult by requiring a signature check on each 25.4 mm of drawer length as opposed to a signature check of an integral drawer.

C. Zero Power Plutonium Reactor (ZPPR) Neutron Portal Monitor (P. E. Fehlau)

Sandia has proposed a neutron detection capability for the Zero Power Plutonium Reactor (ZPPR) personnel portal monitor. We made measurements with a portal detector to compare its

sensitivity for neutron detection to that for gammas. The results indicated a sensitivity of ~ 4 g of ^{240}Pu , which corresponds to 40 g of plutonium in a reference ZPPR fuel plate. The detector cost would be $\sim \$16\text{k}$. We plan to determine the detector's gamma sensitivity for shielded plutonium and the effect of placing neutron-poison shielding around the plutonium.

D. Random Driver (P. E. Fehlau and J. T. Caldwell)

The random driver was examined for its application as a material pass-through for ZPPR. With the

help of D. Lee, Q-1, measurements were made in a random driver to be installed in TA-55. The sensitivity was estimated at 4 g of ^{239}Pu for a unit of the size needed. We also will investigate the use of a National Nuclear Corp. random-driver unit.

E. Centrifuge Enrichment Facility Safeguards (J. T. Caldwell, E. J. Dowdy, P. E. Fahlau, W. E. Kunz, and C. E. Moss)

We completed a conceptual design for a large vehicle monitor, also known as the tunnel detector. UF_6 is detected from passive neutron emissions with an estimated sensitivity of ~ 1 kg UF_6 unshielded. This type of system is intended primarily to monitor railroad freight cars and tractor-trailer rigs. Scaled-down versions also could be used for small-truck and passenger vehicle traffic.

Several meetings have been held with LASL, Sandia Laboratories, Union Carbide Corp., and other interested organizations. The principal focus has been the impact of safeguards on the design of the proposed Portsmouth centrifuge enrichment plant. The engineering design of that facility will be "frozen" in June or July 1978.

Technical studies also have been revived (or initiated) in the areas of cylinder verification meters, shipping dock monitors (especially in the tradeoffs between a large-package monitor and a total vehicle monitor), mass spectroscopy sample studies, and miscellaneous trap/waste/plumbing monitors. In addition, we have been instrumental in formulating the overall safeguards system for Portsmouth and have attempted to formulate ground rules where none existed and clarify those that were ambiguous.

F. Real-Time Inventory (N. Nicholson, C. D. Ethridge,* and T. H. Kuckertz*)

1. Shelf Monitors

The Intel 8748 microcomputer shelf monitor was debugged and interfaced with the Nova 3 minicomputer. Two shelf monitors were successfully tied into the Nova 3. Each unit accumulates data from its gamma detector and scale, stores the data until the

computer requests transmission of the information, resets both counters, and resumes counting. Long-term stability tests were made to check the consistency and operation of the system. Problems with software in the shelf detector were corrected. Shelf monitors were debugged through use of the Prompt-48 as a modified in-circuit emulator. The newly acquired Biomation Logic Analyzer also helped in debugging the software.

Group E-2 is updating the printed circuit artwork and is reducing the board size to allow four-can monitoring in the same size package originally planned for one can. Sixteen monitors containing the updated artwork will be fabricated for the 16-can system planned for demonstration at TA-18.

Initial testing of the shelf monitors interfaced to the Nova 3 indicates the need for an accurate timer to correlate the gamma counts and scale counts of a shelf monitor string comprising 127 monitors. The shelf-monitor software and the flexibility of the Intel 8748 microcomputer have been exploited to generate a time monitor occupying one address on a shelf monitor string. The unit reports a 4-byte value of time to the Nova 3. The software has been written and assembled and is being debugged.

Group E-5 received the in-circuit emulator, ICE-48, which will decrease software/hardware check-out time. The ICE-48 couples the unit being tested into the software control of the Intel MDS (Micro-Development System).

2. Verification Station Development

A CAMAC-based nuclear ADC is being used to collect gamma spectra obtained from an NaI detector. The spectra have been stored in 512 memory locations and plotted with the Data General terminal. A Versatec printer/plotter will improve the speed and quality of the plotting capability. Several gamma sources were used to calibrate the energy region of interest, and a linear calibration-curve-fitting routine that uses least squares has been developed. By means of these calibrations and a crude peak-fitting routine, the energy of major peaks of unknown samples can be obtained. We are investigating techniques that will permit computer control of the gain and offset of the nuclear ADC.

*Group E-5.

Analog inputs to the ADC allow for small variations in both the gain and offset of the conversion from analog signals to digital information.

3. Nova 3 Software Development

We have prepared documentation for ~90 programs and subprograms that were written for the Real-Time Inventory program. In addition, device drivers for the CAMAC system and the various modules are being perfected.

4. Proposed TA-55 Shelf Monitoring System

The Real-Time Inventory shelf system soon will be tested with a 16-detector array at Pajarito Site. A 200-detector system has been proposed for one of the rooms in the plutonium storage vault at TA-55, and tentative concurrence has been obtained from CMB-11.

A final detector configuration for the proposed TA-55 detector array is being completed. To reduce the time and effort required for wiring and detector installation, an attempt will be made to prewire a vault room before any plutonium is permanently stored there.

G. Sandia/Rockwell Plutonium Protection System (N. Nicholson and T. H. Kuckertz*)

A demonstration of the Sandia/Rockwell plutonium protection system is being installed at Rockwell and is expected to go hot by the middle of July 1978. The 3-month operational demonstration should be completed by mid-October. An additional two months are scheduled for solving problems discovered during the operational phase. The system should be dismantled and returned to Sandia by the end of December 1978.

The Phase I or "basis" verification station is functioning well and confirms the presence of plutonium (by nuclear detection techniques) within the sealed

Sandia overpacks as they enter the Sandia plutonium storage system. The primary objective of the demonstration is to evaluate the impact of the Sandia system on the operation of the Rockwell plutonium vault. The principal interest in the verification station is in the complexity and duration of the verification procedure. Technical aspects of the verification are of secondary interest. Because of the operational emphasis of the demonstration, the basic verification station will satisfy all functional requirements necessary for a thorough evaluation and will therefore remain in the Sandia system through the remainder of the demonstration period.

Work continues on an advanced verification station that can be used in a Sandia-type plutonium storage system. The station will be designed to give a more quantitative value, or verification, of the contained plutonium than does the basic station, which provides a confirmation of plutonium. Measurements will be made in a neutron well-counter-type geometry containing four banks of neutron detectors tied into a reduced variance logic system and a high-level coincidence counter. Also, a precision balance will give the weight of the plutonium plus both inner and outer containers. We are also investigating the application of a microcomputer to analyze the gamma spectra obtained from an NaI detector. An unsophisticated analysis, not requiring extensive memory or software, may be capable of identifying the characteristic gamma lines of plutonium. Although the measurement would not be quantitative, the technique could determine whether the principal gamma emitter was plutonium; if it was not plutonium, the energy of the main gamma lines of the sample in question would be supplied. With an appropriate data base, the radioactive material also could be identified.

A computer-based verification station would have definite advantages. Interfacing it to another system, such as the Sandia protection system, would be simple. Furthermore, the station would be a stand-alone instrument and could be used in all future portal security systems. Any change or addition of detectors or devices could be accommodated by changing the software rather than redesigning the hardwired logic modules.

*Group E-5.

H. Reduced Variance Logic Redesign (E. J. Dowdy and C. D. Ethridge*)

Group E-2 is wire-wrapping two circuit boards. The first board contains the counting logic; the second contains the microcomputer/microprocessor calculating system. Group E-5 has written and is assembling the new data multiplexing routine software. After the new display routine is written, the existing software will be merged with the new routines for check-out. Board two will be debugged after the total package is assembled and after board one is tested.

I. Hand-Held SNM Monitors (W. E. Kunz and D. R. Millegan)

At the request of DOE/OSS we evaluated the detection sensitivity of National Nuclear Corp.'s new version, HM-3, of the hand-held SNM monitor. The HM-3 uses an NaI(Tl) crystal of 38-mm diam and 19-mm length. The crystal is only one-half the length of that used in the earlier version, yet it easily exceeds our proposed detection sensitivity standards.

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

The second SNM portal monitor was installed at TA-55. Of five 10-g ^{235}U test spheres for portal monitors fabricated by CMB-6, three were given to E-1 for routine portal monitor tests.

Group E-5.

K. NRC Portable Monitor (P. E. Fehlau)

We have completed NRC Task 6, which involved carrying out an inspection specified by a procedure drawn from inspection techniques discussed in the Task 5 report. A test procedure document was written and used to conduct a trial on the National Nuclear Corp. portal monitor at TA-55. From the results of this trial, we were able to modify the procedure and use it on the IRT Corp. portal monitor at T-55. The procedure was tested by David Woodwell. Group E-1, who is responsible for performance testing of portal monitors at LASL. The procedure document and the results of the trials are included in the Task 6 report.

NRC reimbursable R-174 is now complete and task reports will be condensed into a LASL report.

L. IAEA Task E-21 (Q-2 Staff)

The paper study of possibilities for performing irradiated fuel monitoring has been completed, and a report has been written. Copies were given to IAEA representatives.

Work continues on the Cerenkov light-detection scheme for spent fuel elements, with EG&G/LA developing the hardware. A visit was made to the Omega West reactor, and arrangements have been made for EG&G personnel to make preliminary measurements on Omega West irradiated fuel. We will provide calculational support for their measurements.

PART 3

SAFEGUARDS SUBSYSTEM DEVELOPMENT AND EVALUATION

GROUP Q-3

Ronald H. Augustson, Group Leader

John J. Malanify, Alternate 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 real-time 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 real-time 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 (called DYMAC/TA-55) is being integrated into the new LASL Plutonium Processing Facility at TA-55 to serve as a working demonstration program. 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 cost-effective manner.

The DYMAC/TA-55 program falls into three phases. Phase I, already completed, was a pilot

program of research and development and in-plant testing of NDA instruments coupled with an interactive computer at the LASL DP Site facility. In Phase II, currently under way, the technology and equipment developed in Phase I is being integrated into the new plutonium facility. DYMAC/TA-55 is a full-scale, plantwide DYMAC system with NDA measurement stations located in each unit-process accounting area, interactive computer terminals for data entry and retrieval, and real-time accountability and control of all SNM in the facility. In Phase III, the data collected during operation of the four DYMAC subsystems will be evaluated in depth, with particular attention to effectiveness of material control, reliability, operational ease, costs, and applicability to other facilities.

The DYMAC program began official operation at the new Plutonium Processing Facility. On January 19, 1978, the nuclear materials officer (NMO) received a shipment of depleted uranium and entered it into the DYMAC inventory, thereby initiating DYMAC operation. The first shipment of plutonium entered the facility on February 9. Figure 39 shows the initial shipment being wheeled into the vault. Regular shipments of plutonium have been arriving at the facility regularly each working day since February 9. The NMO meets the shipment at the loading dock and escorts it into the facility's receiving area, where he enters each item into the DYMAC inventory and places it in the vault.

The first two processes to begin operation in the new facility were Advanced Carbide Fuels and Metal Fabrication. The Advanced Carbide Fuels process moved into the ²³⁹Pu R&D Wing in November 1977 and began sampling depleted uranium on January 20, 1978. On March 13, a formal shakedown period began (with depleted uranium)



Fig. 39.
The NMO wheels the first shipment of plutonium into the vault.

and will continue until June, when plutonium will replace the uranium. The Metal Fabrication process moved into the entire 300 Wing in March and began full operation on April 24.

The DYMAC computer has been available on request to the NMO and to the needs of the two processes. The rest of the time the programmers use it for further system development. By April 30 the DYMAC inventory contained 661 items, most of them located in the vault.

The DYMAC computer now has 40 ports for use by terminals and instruments. By the end of April 1978, 24 terminals in the facility and cold-support building and 10 electronic balances were connected to the computer. The two thermal-neutron coincidence counters (TNCs) in the casting receipt area of the Metal Fabrication process began operation; neither is yet on-line.

Increased demand for computer time has caused system development to give way to the needs of regular processing. The programming staff does much of its development before 8 a.m. and after 5 p.m. The lack of computer time also affects the training of DYMAC supervisors and technicians. Therefore, Group Q-3 has ordered a second Eclipse computer to handle all demands outside of normal processing and to serve as backup to the present computer.

I. DEVELOPMENT AND INSTALLATION

A. Communication System (K. A. Lindsey and V. S. Reams)

Seven new Teleray terminals were checked out. They will be installed as soon as the next scheduled process occupies its assigned laboratory space at the Plutonium Facility.

Forty on-line computer ports are now available. Each time the DYMAC system is brought up, the programmer specifies the ports to be used for terminal and instrument connections. Such flexibility is useful for system development and training requirements. During the next reporting period, all 128 on-line computer ports will be connected to the DYMAC system.

B. Balance (M. M. Stephens)

Twenty 5.5-kg capacity Arbor electronic balances are installed in gloveboxes at the new facility and their corresponding readout units are connected to the DYMAC computer.

The manufacturer delivered three 15-kg Mettler electronic balances. One is installed in a glovebox in the Metal Fabrication wing and is connected to the DYMAC computer.

Accuracy and precision data for these balances are being gathered and will be discussed in the next progress report.

C. Thermal-Neutron Coincidence Counter (TNC)
(R. S. Marshall, J. P. Gonzales, N. Baron, and W. R. Severe)

1. Status of Installations

Sixteen TNCs are to be installed at the new facility. Their major mechanical components have been fabricated and are in possession of Group Q-3. DYMAC personnel installed one TNC in the ^{238}Pu R&D Wing, another in the ^{239}Pu R&D Wing, and two in the Metal Fabrication Wing. Seven TNCs have been installed, of which six are operational.

The two TNCs in the Metal Fabrication Wing are horizontal units with counting wells that protrude from the glovebox side, as shown in Fig. 40. The large metal enclosure above the electronics unit houses the horizontal counting well, ^3He detector

tube array, and polyethylene-cadmium shielding. These TNCs are designed to assay large items such as plutonium metal components. The horizontal orientation of the TNCs and their roll-in sample holders simplifies the transfer of heavy, bulky pieces into the counting well. Figure 41 shows the sample holder in the rolled-out position. Like all TNCs in the facility, these are dual range instruments; a high-efficiency mode is used for assaying plutonium items of <1 kg, and a low-efficiency mode is used for assaying plutonium items of >1 kg.

In the Recovery Wing, six TNCs are being installed in gloveboxes. Because these gloveboxes are being readied to receive plutonium, the components had to be installed before the gloveboxes were sealed. DYMAC personnel also installed the counting well, sample chamber, and Teleflex cable in the gloveboxes, and on the outside they connected

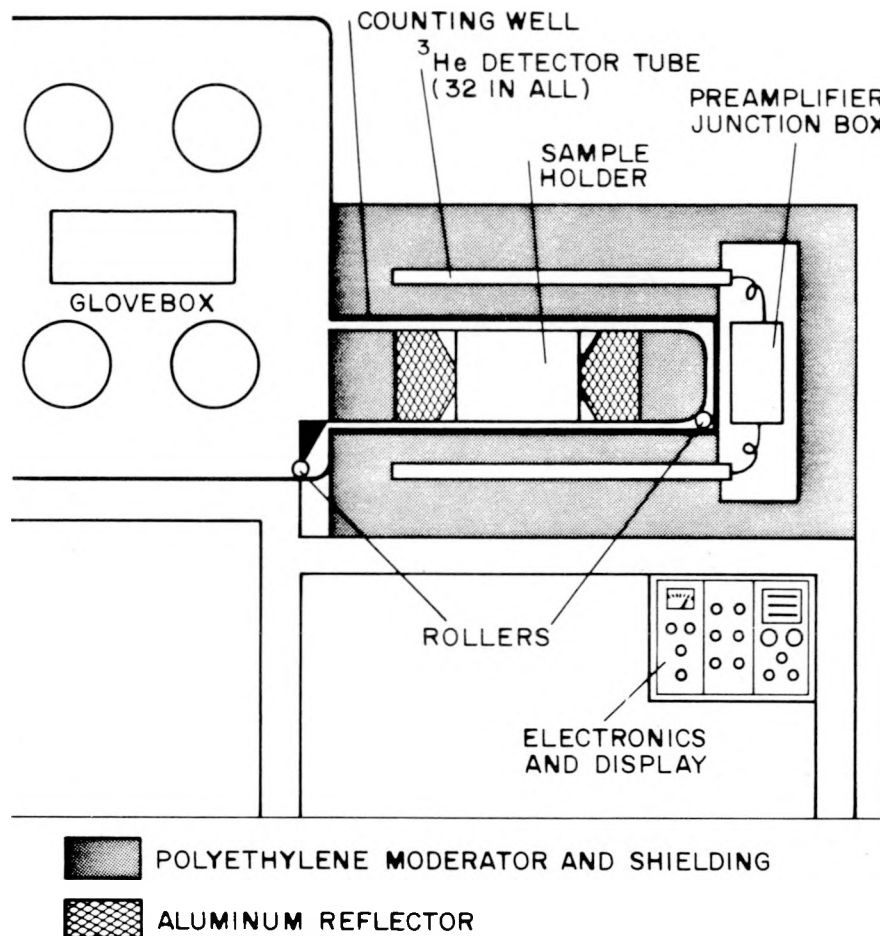


Fig. 40.
Horizontally oriented TNC.



Fig. 41.
Sample holder for horizontally oriented TNC.

the drive motor and pulleys and associated guide tubes that affect the glovebox seal. The detector tube assembly, shielding, and electronics (which do not affect the glovebox integrity) are being assembled in another location before installing them. The six Recovery Wing TNCs should be operational by July 1978.

DYMAC personnel made a destructive test to determine the life span of the Teleflex cable and pulleys that raise and lower the TNC sample chambers. The test assembly (Fig. 42) has a dc motor and controller, square aluminum pulley boxes that house 5.08-cm-diam pulleys, and a Teleflex cable that supports a lead brick load. The assembly automatically raised and lowered a varying load of bricks ~1 m before the cable broke. Cable failure resulted more from the number of flexures than from load. During testing, the cable failed after ~4500 cycles. Consequently, we designated cable replacement at 2500 cycles, which corresponds to a 1- or 2-yr interval, depending on frequency of use.

2. Checkout and Deadtime Measurements

To evaluate the performance of the TNC electronics packages, we ran a series of tests to check deadtime and spatial sensitivity. For the electronic deadtime determination, we calculated deadtime parameters τ from measurements (defined in Ref. 8, p. 76) for the TNC electronics located at gloveboxes 307 and 310; the respective measurements were 3.58×10^{-6} s and 3.48×10^{-6} s—values that are being used to correct the counting data for deadtime. To determine the spatial sensitivity of the same TNC detectors, we placed a ^{252}Cf source at varying locations within the counting well. The variation in the vertical sensitivity is only $\pm 0.75\%$ for locations ranging from 82.55 to 96.52 cm above the entry position of the TNC well. Thus, the assay results are insensitive to the fill height of material within the sample container.

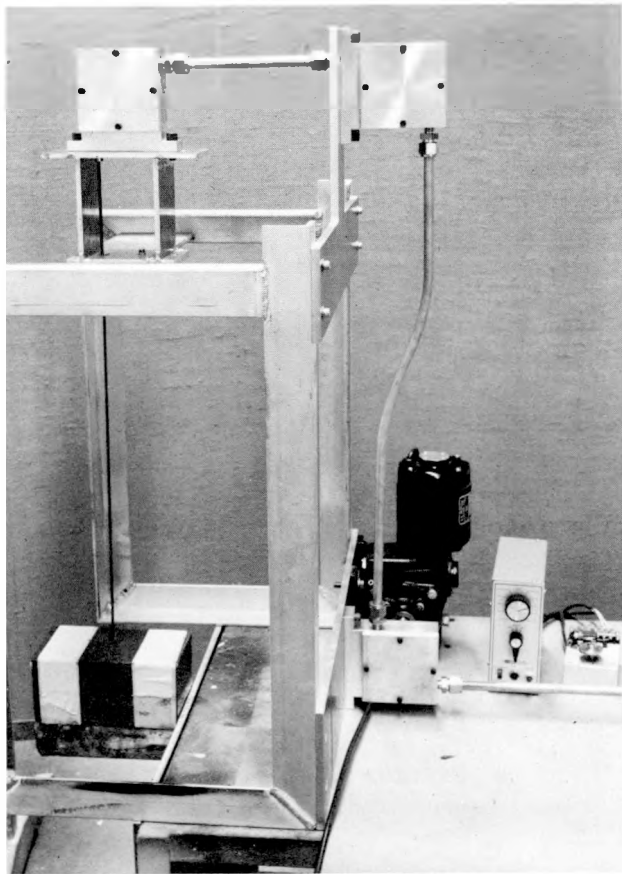


Fig. 42.
Teleflex cable test assembly.

3. Status of THENCS Unit

The thermal-neutron coincidence counting system (THENCS) is described in Ref. 8, pp. 72-75. During the present reporting period, the hardware interfacing between the Motorola 6800 microprocessor and the TNC's coincidence circuit was built. The FORTRAN software and associated assembly language programming are essentially complete for one of the two types of TNCs designed for use in the new facility. These THENCS units will provide the data analysis function for all DYMAC TNCs.

4. Measurement of Scrap in 300 Wing

DYMAC measurements were begun in the Metal Fabrication Wing with the two fully operational TNCs installed there. Production began in April

1978, and the TNCs were used to measure a wide variety of waste and scrap materials.

D. Count Room (R. H. Augustson)

DYMAC personnel have installed three NDA instruments in the TA-55 count room. The large barrel counter and the 2-L counter were moved from DP Site and reassembled at TA-55. The two TNCs will be calibrated and made fully operational during the next reporting period. In addition, an SGS, on loan from Group Q-1, is in the count room and will be functioning soon.

E. Solution Assay Instrument (SAI) (D. G. Shirk and J. L. Parker)

The usual procedure for recovering plutonium from scrap material is to dissolve the material in a solution and purify the solution by ion exchange and solvent extraction techniques. For accountability purposes, the plutonium must be measured in solution form; however, it is seldom feasible to measure an entire batch of solution. Instead, one establishes a mass ratio. First, the operator measures the mass of a 25-ml aliquot with an electronic balance. Next, he obtains a measurement of the solution mass in the holding tank by means of the pressure transducer system (described in the next section). Then the SAI software calculates the mass ratio, which is the ratio of tank mass to aliquot mass. The operator places the aliquot into the SAI sample chamber to determine the number of grams of plutonium in the aliquot. To determine the number of grams of plutonium in the holding tank, the SAI software multiplies the number of grams in the aliquot by the mass ratio. The SAI responds to a data request from Eclipse with two numbers: the grams of plutonium in the holding tank and the measurement uncertainty.

Group Q-1 designed and constructed a prototype SAI to assay plutonium solutions by gamma-ray spectroscopy. The SAI consists of six major hardware components, as shown in Fig. 43. A Data General-compatible minicomputer with 32k, 16-bit word memory; central operator console; NIM electronic modules; General Electric Terminet terminal and storage oscilloscope (located on a mobile

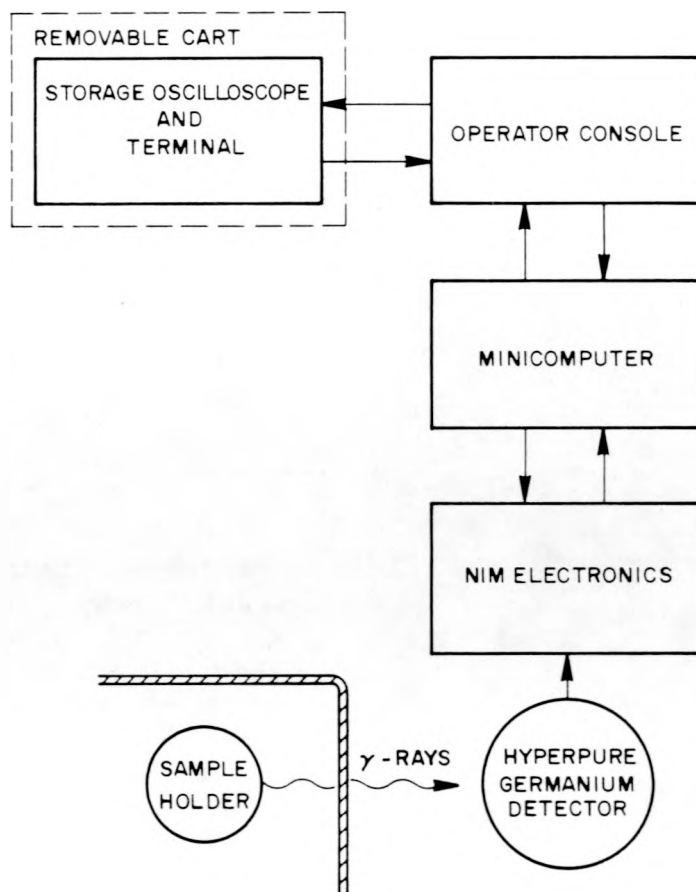


Fig. 43.
Components of the solution assay instrument.

cart); a hyperpure germanium detector; and a sample holder with pneumatic tungsten shutter (located in the glovebox). Q-1 also provided a basic operating system that allows the resident program to control the operator console, analog-to-digital converter, calendar clock, and graphics display for the storage oscilloscope.

Groups Q-1 and Q-3 formulated a test-and-evaluation program for the prototype. The program has two primary functions: first, to ascertain the functional reliability of the numerous components and their suitability for fabrication; second, to ensure that the applied-physics principles involved in the preliminary production code are correct for measurement problems that may arise. The first function involved writing many diagnostic routines in assembly language to allow direct communication with the various hardware devices. Group Q-3 ran these diagnostic routines on each component of the

prototype instrument to ensure its integrity before fabricating duplicate systems. The second function of the test-and-evaluation program involved testing the applied-physics assumptions and their corresponding algorithms. A series of three experiments were conducted to test these assumptions and algorithms. The results of these experiments, shown in Fig. 44, indicate that the SAI can measure (with >1% precision) concentrations of plutonium solutions that vary from 0 to 500 g/l. In these experiments, ^{76}Se and uranium were substituted for plutonium in the solutions.

The preliminary production code (in assembly language) requires a minimal amount of operator intervention for assaying a sample of plutonium solution. The code contains warning diagnostics to indicate improper operator procedures and provides other diagnostics to monitor instrument performance. If a malfunction occurs that would yield an

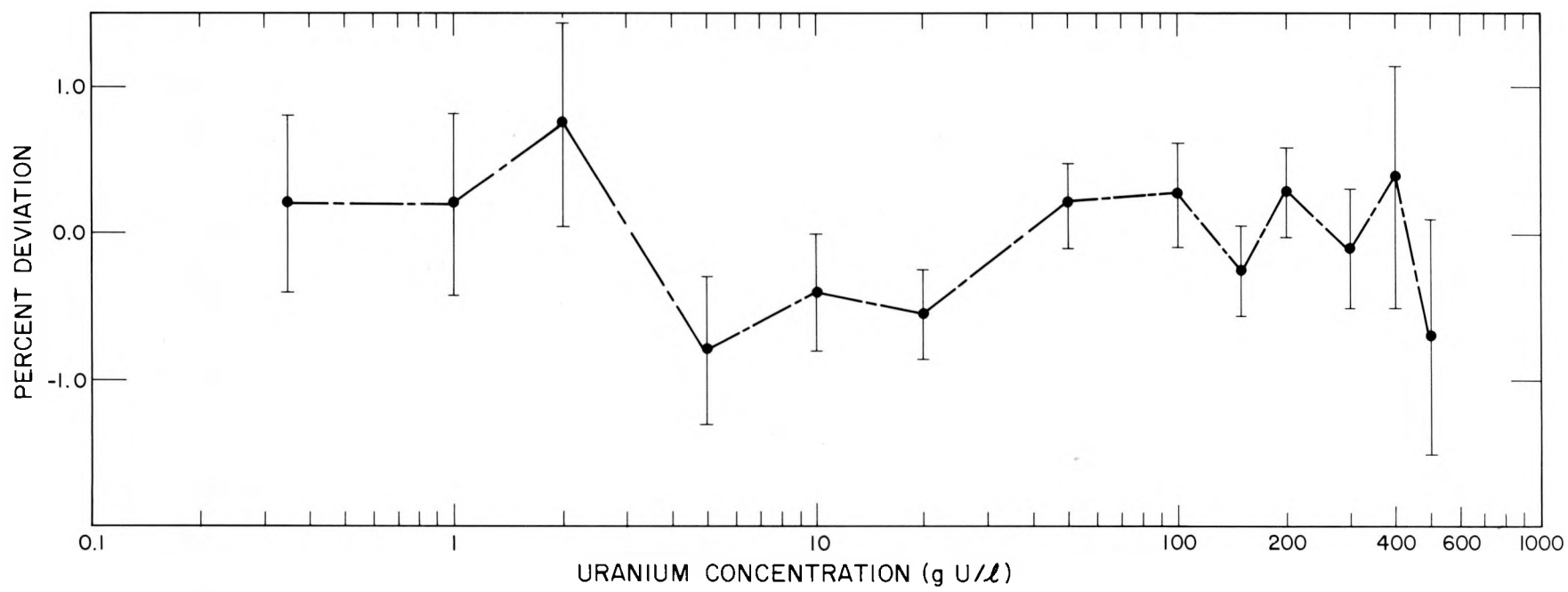


Fig. 44.
Fractional deviation of corrected rate per unit activity from the average.

erroneous result, the software renders the instrument inoperable and instructs the operator to get help.

Q-3 technicians fabricated one complete set of printed-circuit boards for the instrument and used the test-and-evaluation program to check their reliability. The central operator consoles also are being fabricated and checked out.

Group Q-3 will install five SAIs in the 400 Wing (the only wing to receive them). Each SAI will be located near a row of as many as 8 to 10 solution tanks. The SAI can assay only one tank at a time. The sample holder and the pneumatic tungsten shutter are in place within the glovebox and the hyperpure germanium detector is installed beneath the glovebox. The system should be operational by June 15, 1978.

F. Pressure Transducer System (W. R. Severe)

Our new approach to weighing tanks of plutonium solution for the SAI is to use strain-gauge pressure transducers. Each tank will have one pressure transducer associated with it, located outside the glovebox near its top. The transducer measures the solution pressure at the tank bottom via a dip-tube bubbler system, as shown in Fig. 45. Because the solution tanks have nearly constant cross-sectional area, the pressure at the bottom of the tank is directly related to the solution weight. Figure 46 is a plot

of the electronic output of the pressure transducer vs solution weight for a typical solution tank. The maximum nonlinearity of response for these data is ~94 g of solution weight.

The pressure transducer system should be an excellent replacement for the load cell system, which has been abandoned. It uses the same electronics packages as the load cell system. Moreover, it offers an improvement by providing solution weights based on a single measurement instead of the two by-difference measurements required by the load cell system.

Group Q-3 is procuring and assembling the components for the pressure transducer system. We expect the installation of the transducers on 42 tanks to be complete by September 1978.

G. Hot Glovebox Measurements (R. H. Augustson and R. Siebelist)

Thirty gloveboxes were moved into the new facility from the processing area at DP Site. They were thoroughly cleaned before the move and contained only residual quantities of plutonium. A DYMAC team with NDA equipment will measure this residual material once the hot gloveboxes are installed.

The NDA equipment used by the team consists of a portable collimated NaI gamma-ray detector and an MCA. The team has measured eight gloveboxes,

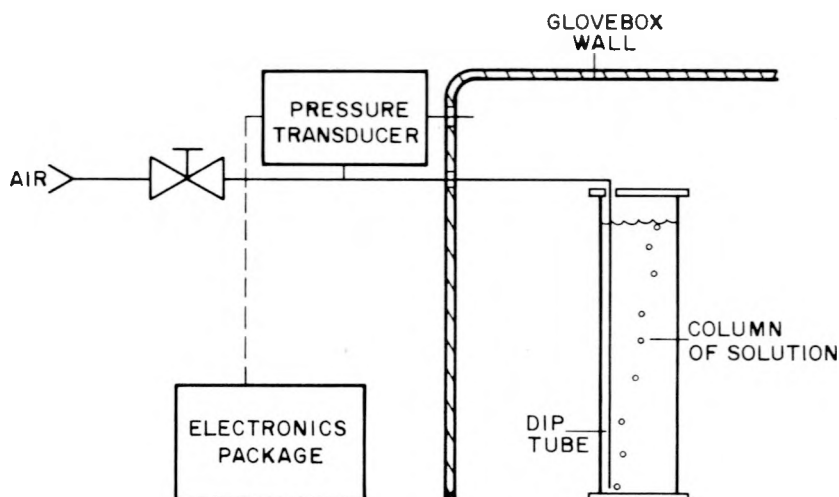


Fig. 45.
Pressure transducer system.

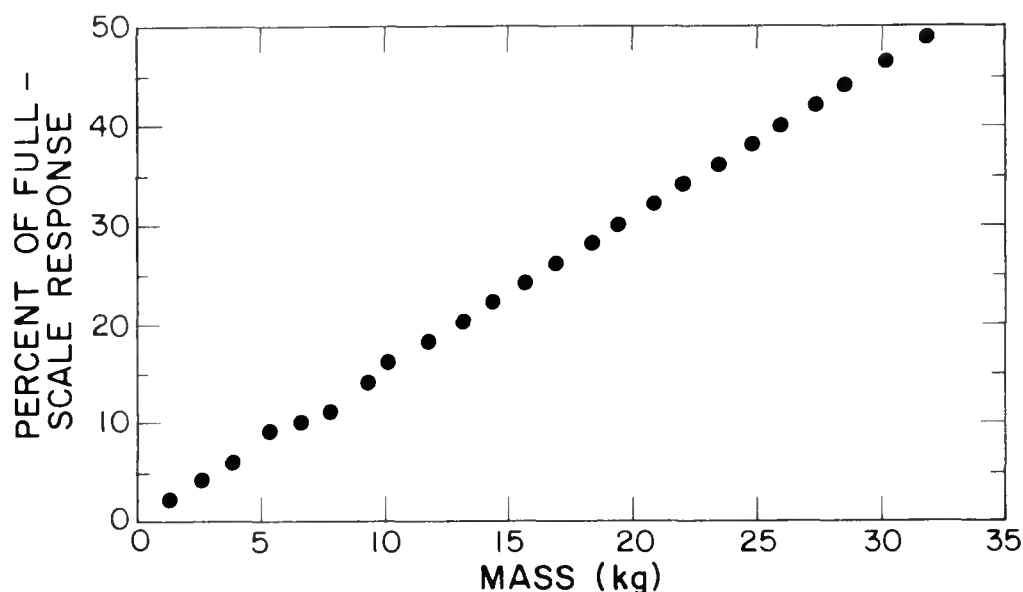


Fig. 46.
Response vs mass.

with the total residual material amounting to 44 ± 22 g ^{239}Pu . The measurements will be repeated regularly when processing begins in the gloveboxes. Thus the DYMAC system can establish a history of plutonium holdup in each glovebox.

H. DYMAC System Software (R. F. Ford,* J. Hagen, J. K. Wooten,* E. P. Elkins, W. H. Dorin,* T. A. Cordova, and W. M. Olson***)**

The DYMAC program began operation on January 19, 1978, when the NMO received the first shipment of nuclear material at the new facility. During the reporting period, the DYMAC system ran 45 processing days and handled 2178 transactions. No SNM differences occurred during comparison of the physical inventory with both the DYMAC general ledger and with the laboratory-wide LASL accountability system (LAS) for nuclear materials. Although there were no SNM differences between the two accountability systems, DYMAC and LAS did show occasional differences in other fields of the inventory records, such as creation date

of an item, or its location, shelf, item description, measurement code, bulk amount, or bulk units. This lack of agreement is because LAS and DYMAC follow different inventory updating procedures and because LAS does not execute all the transactions in chronological order. In all cases of disagreement, except date, the DYMAC inventory correctly represented the physical inventory.

1. Backup Computer

Experience indicates that the initial plan to use the Nova 840 computer from Phase I is no longer viable. The computer's limited memory and inability to run INFOS, the data base manager, make it useless for backup and training purposes. Originally, we had planned to use the data base manager, written for DYMAC Phase I, at the new facility, but with our more clearly defined resource needs for backup and training, we find that INFOS is indispensable. We plan to purchase an Eclipse C330 that will include the following equipment:

- Eclipse C330 with 80k words of memory, memory allocation and protect (MAP), and expansion chassis;
- 20-megabyte top loader disk system;
- magnetic tape unit;

*Group E-5.

**Group AADP-2.

***Group CMB-11.

- 2 system CRTs;
- real-time clock;
- 165 character/second line printer; and
- communication chassis with asynchronous line multiplexer ALM-16 and four 4-line, 20-mA interfaces.

2. General Software Modifications

Changes to the DYMAC software continue as the system grows to accommodate the facility's processing and accountability needs. Changes made during the reporting period include an expanded measurement control program, additional off-line reports, and improved user functions.

The measurement control program now includes a table of weight and count standards required for maintenance of the NDA instruments. Another program update includes TNC maintenance.

The DYMAC system generates two new off-line reports. One report sorts and sums the inventory by location and by shelf to aid in taking physical inventory. The other sorts and sums the inventory by special designator.

Improvements to user functions will clarify messages, shorten system response time, and facilitate data entry. For example, when the user wants to correct an erroneous entry on the display screen, he can hit the delete key to erase one character at a time or the back-slash key to erase the entire line. Another improvement is response time-outs that release the system from a terminal request if the user lets 5 min elapse before responding and from an instrument readout if the response takes >1 min. The system thus becomes more available to other users.

3. Advanced Carbide Fuels: Transaction Refinement

The transaction sequence for the Advanced Carbide Fuels process in the ^{239}Pu R&D Wing (Ref. 6, p. 75) has been altered. The two changes to the list (shown in Fig. 47) were the addition of option 12, REDUCE BRIQUETTES, which adjusts the bulk value of a briquette lot to account for a 19% bulk weight loss in the reduction furnace, and the reordering of transactions 8 through 16 to more closely resemble the order of physical processing.

```
<<< SELECT OPTION >>>
1...RECEIVE FROM MANAGEMENT ROOM
2...TAKE SAMPLE
3...RETURN FEED STOCK TO MANAGEMENT ROOM
4...SEND SAMPLE OR SCRAP
5...PREPARE NEW BATCH
6...ADD CARBON AND UO2 TO PUO2 BATCH
7...TRANSFER IN GLOVEBOX LINE (WITH MEASUREMENT)
8...REDESCRIBE BATCH
9...WEIGH SCRAP
10...SEND BRIQUETTES TO REDUCTION FURNACE
11...RECEIVE AND VERIFY BRIQUETTES
12...REDUCE BRIQUETTES
13...BLEND LOT
14...SEND SINTERED PELLETS TO GRINDING
15...RECEIVE SINTERED PELLETS
16...WEIGH AND SEND FINISHED PELLETS
? _____
```

Fig. 47.

Advanced Carbide Fuels transaction sequence.

```
<<< SELECT OPTION >>>
1...RECEIVE SAMPLE PELLET
2...APPORTION PELLET TO PIECES, SCRAP POTS
3...COLLECT PELLET PIECES TO BE SCRAPPED
4...MEASURE SCRAP
5...SEND SCRAP TO RECOVERY
6...WEIGH UNPROCESSED PELLETS TO DETERMINE MIP
? _____
```

Fig. 48.

Metallography transaction sequence.

4. Metallography Transactions

DYMAC programmers defined a transaction sequence (shown in Fig. 48) for the metallography process, which occupies a portion of the ^{239}Pu R&D Wing. The transactions will be ready when Metallography begins processing in May 1978. Most input to the metallography process is pellet samples from the Advanced Carbide Fuels process. Metallography technicians cut each pellet in half. One half is kept as a reserve sample and the other is processed. The scrap includes sweepings, grit paper, cloths and wipes, etching solution, and mounting materials.

The material value for each pellet half is <1 g, the minimum accountable unit for plutonium. Normally, each half would not appear on the book inventory, but DYMAC will account for the plutonium to the milligram.

5. Metal Fabrication Transactions

DYMAC programmers defined and wrote the transactions for the metal fabrication process, which occupies the entire 300 Wing. There are three option lists, corresponding to the material management room, fabrication process, and supervisory capabilities.

The 300 Wing material management room is a unit process whose option list is shown in Fig. 49a. Options 2 and 3 follow material sent to and received from the fabrication unit process. Options 1 and 4 follow material sent to and received from other processes in the facility, such as the vault, counting room, or Recovery Wing. Option 5 follows material as it moves within the material management room.

The option list for the fabrication process (shown in Fig. 49b) gives the process operator a choice as he performs the various process steps. The transactions can call for a measurement or write a MIP transaction where required.

The supervisory option list (shown in Fig. 49c) requires a password for access. Options 1 and 2 allow a supervisor to send and receive material without its going through the material management room. Material of that sort will not go through the conveyor system either because of its size or because it is sealed and can be entered into the glovebox line through one of the many hoods in the processing area. Options 3 and 4 are transactions whose sensitivity requires the attention of a supervisor.

```
<<< SELECT OPTION >>>
  1...RECEIVE FROM OUTSIDE 300 WING
  2...SEND TO PROCESSING
  3...RECEIVE FROM PROCESSING
  4...SEND OUTSIDE 300 WING
  5...SHELF TRANSFER
?_____
```

a)

```
<<< SELECT OPTION >>>
  1...RECEIVE FROM MANAGEMENT ROOM
  2...RECEIVE FROM WITHIN FABRICATION PROCESS
  3...SEND ENTIRE LOT WITHIN FABRICATION PROCESS (MEASURE AND
    AUTOMATICALLY MIP THE DIFFERENCE)
  4...SEND ENTIRE LOT WITHIN FABRICATION PROCESS (NO MEASUREMENT
    OR CHANGE OF SPECIAL DESIGNATOR)
  5...COMBINE OR DIVIDE LOTS
  6...SEND ENTIRE LOT TO ISO PRESS ROOM (NO MEASUREMENT)
  7...RECEIVE ENTIRE LOT FROM ISO PRESS ROOM
  8...MEASURE IN TNC
  9...CHANGE SHELF
 10...CHANGE ITEM DESCRIPTION
 11...CHANGE BULK
 12...RECEIVE AT ISO PRESS ROOM
 13...SEND ENTIRE LOT FROM ISO PRESS ROOM (NO MEASUREMENT)
 14...BULK WEIGHT VERIFICATION
?_____
```

b)

```
<<< SELECT OPTION >>>
  1...SEND FROM PROCESS (NOT VIA MANAGEMENT ROOM)
  2...RECEIVE INTO PROCESS (NOT VIA MANAGEMENT ROOM)
  3...CLEAR LOT TO MIP
  4...WEIGH AND APPLY FACTOR
?_____
```

c)

Fig. 49.
Metal fabrication transactions.

6. Streamlining of NMO Transactions

The NMO has two shipping/receiving accounts, one in the count room and the other in the vault. Identical option lists are associated with each, and are shown in Fig. 50. We modified Option 1 to make it repetitive. Now the NMO and his staff can receive a series of items consecutively without having to return to the program start. No longer do user-number, password, receiving account, process, and option selection have to be entered for each item. At the end of each RECEIVE transaction, the system asks whether the user plans to receive another item. If the user answers yes, he can begin entering specific information about the next item. Option 6 also has been modified for repetitive SEND within the facility. As with Option 1, this transaction allows the operator to send a series of items without having to return each time to the program start.

Option 8 enables the NMO to make corrections. He selects it when he detects an entry error made while receiving an item from an external account. (Data entry errors occur most frequently when an item is first entered into the data base.) He makes the correction through account 7ER, which has been set up exclusively for error corrections.

In the vault transaction sequence (shown in Fig. 51), the repetitive transactions have been added to Options 1 and 2. Transactions 3 and 4 have been added. In the transaction sequence for account 777 (shown in Fig. 52), the NMO clearing account, Options 8 and 9 were added. The GENERAL CORRECTIVE TRANSACTION allows the NMO to correct input mistakes. The WRITE IN FROM ACCOUNT 991 transaction allows the NMO to adjust the station balance for normal operating losses.

```
<<< SELECT OPTION >>>
1...RECEIVE EXTERNAL SHIPMENT INTO TA-55
2...SEND EXTERNAL SHIPMENT FROM TA-55
3...ITEMIZE A SHIPMENT
4...COMBINE ITEMS TO FORM A SHIPMENT
5...CLEAR NON-PHYSICAL ITEM TO ACCOUNT 777
6...SEND ITEM WITHIN TA-55
7...RECEIVE ITEM FROM WITHIN TA-55
8...EXTERNAL TRANSACTION CORRECTION
? _____
```

Fig. 50.

NMO shipping and receiving transactions.

```
<<< SELECT OPTION >>>
1...RECEIVE INTERNAL ITEM
2...SEND INTERNAL ITEM
3...GENERAL EDIT OF NON-NAME ATTRIBUTES
4...TRANSFER TO ACCOUNT 777 (NO PHYSICAL MOVE)
? _____
```

Fig. 51.

NMO vault transactions.

```
<<< SELECT OPTION >>>
1...CHANGE ENRICHMENT
2...DECAY
3...NORMAL OPERATING LOSS
4...WRITE-OFF TO MUF
5...DEGRADATION
6...WRITE-OFF SHIPPER-RECEIVER DIFFERENCE
7...TRANSFER MATERIAL BACK TO PROCESS CUSTODIAN
8...GENERAL CORRECTIVE TRANSACTION
9...WRITE IN FROM ACCOUNT 991
? _____
```

Fig. 52.

NMO transactions for the clearing account.

II. OPERATIONS

A. History and Status of DYMAC at the Plutonium Processing Facility During the Reporting Period (D. C. Amsden, C. E. Nordeen,* R. J. Herbst,* and D. R. Harbur*)

1. NMO Shipments and Receipts

Nuclear material entered the Plutonium Processing Facility for the first time on January 19 (~5 months later than scheduled). Eight bottles of depleted uranium were received at the loading dock by the NMO, who entered them as the first items in the DYMAC inventory. He took them to the vault and they were called up the next morning to be used by the Advanced Carbide Fuels process.

The first shipment of plutonium arrived at the facility on February 9. Full security and safety measures began on that day. The NMO entered the shipment into the DYMAC inventory and took the items to the vault. Since that day, he and his staff have been receiving regular shipments of plutonium for the vault as holdings at DP Site are transferred to the new facility. The plutonium transfer will not be completed until Fall 1978.

Early shipments from DP Site did not always conform to the stricter accounting methods at the new facility. Sometimes several items were grouped under one name—this is not allowed in the DYMAC system because control of individual items is lost. Sometimes the information was incomplete and the NMO had to resolve the problems before entering the items into the DYMAC system. All recent shipments have conformed to DYMAC requirements.

During the reporting period, as processes began to move into the new facility, the DYMAC system was brought up for a few hours during the work day as needed. The rest of the time it was used by the programmers for development. Even when the system was operating with the real data base, programmers did development in background mode. This limited to five the number of terminals that could be used at once. When all were in use simultaneously, system response time was slow, with a longer wait between prompts and displays. DYMAC personnel will reallocate system resources to improve the system response.

The DYMAC system provides diagnostics for data entry that help screen out errors. The readily available on-line reports help the NMO locate and correct mistakes. The off-line reports that DYMAC produces at the end of every working day provide the NMO a printed inventory of every item in the facility by location. The end-of-day current inventory (by material type) is useful for locating errors in the inventory—a careful scan turns up errors that he can correct. The daily inventory printout provides, for example, the locations of measurement standards that need to be returned to the vault at the end of the working day.

2. Advanced Carbide Fuels

The Advanced Carbide Fuels section moved into the Plutonium Processing Facility in November 1977 and began sampling depleted uranium on January 20, 1978. The formal shakedown of the equipment, procedures, and process steps began March 13 with depleted uranium, and will continue until June 1.

After June 1, the section will work with plutonium, but before then the staff wants to become familiar with and check out new equipment, to feel comfortable with DYMAC accountability procedures, and to be thoroughly acquainted with the steps involved in making the fuel pellets.

The advantage of depleted uranium during the shakedown period is its physical similarity to the plutonium oxide that will be processed later. In fact, the real material will be 85% depleted uranium and 15% plutonium.

To make pellets out of the oxide feed stock, eight different unit processes are employed, as reported in Ref. 8, pp. 61-62 (grinding and inspection now have been separated into two unit processes): oxide blending, oxide press, reduction furnace, comminute, automatic pellet press, sintering furnace, grinding, and inspection. Two batches of material were processed in sequence through the sintering stage and through six of the unit processes. Neither batch has made it completely through the entire process. For this shakedown period, the batches will not go through the grinding stage.

*Group CMB-11.

The transaction option list for the Advanced Carbide Fuels process has been modified. The most significant oversight in designing the process flow was the failure to recognize all the unit processes that generate measurable scrap. The change was relatively simple: the transaction list needed no modification because a scrap operation already existed there. Instead, the change was made on the direction sheet that tells technicians what transactions to make for each unit process. Instructions to make a scrap transaction were added to several other unit processes. Another problem arose with a prompt that asks the technician to provide a "to" account number—a technically correct but misleading phrase—and almost every mistake the group has made stems from this prompt. It will be reworded so that the technician supplies the proper account number.

During the processing, technicians weighed the oxide with the DYMAC electronic balances, which were not yet on-line. They wrote down the results before making the corresponding transactions. Once the balances are placed on-line, technicians must make each weighing in conjunction with a transaction.

DYMAC is extremely useful to the section supervisor. Its on-line reports help him keep an eye on all the processing. The payoff will be at inventory time when physical inventory-taking will be simple. When a hardcopy terminal is installed next to the supervisor's terminal, random inventories of glovebox locations will be made by obtaining a printout of what should be in a particular glovebox and having the technicians take physical inventory.

3. Metal Fabrication

The Metal Fabrication section moved into the 300 Wing of the new facility and began bringing up plutonium from the vault on April 24, 1978. The next day they began casting plutonium pieces. Metal Fabrication is the second section to occupy the new facility. Its accountability areas are set up differently from those of the Advanced Carbide Fuels section. The entire 300 Wing has been designated as one account. Within the account, seven unit processes have been defined: material management, casting, machining, welding and assembly, inspection, special recovery, and uranium processing. Each of these processes still occupies a

room, or part of a room. Hence, to pinpoint more accurately the whereabouts of a particular location, unit processes have been subdivided into locations (i.e., glovebox lines), which are further subdivided into "shelf" locations. (Shelf is a field in the inventory record originally intended only for vault use.) In the Metal Fabrication section a shelf corresponds to a glovebox, or part of a glovebox (if it is very large), or a bin beneath a glovebox. The Metal Fabrication section does not use the item description field in the inventory record (as does the Advanced Fuels section). Instead, the item ID indicates that information.

The section supervisor is refining the Metal Fabrication transactions on the basis of real operation and says the transactions work quite well.

The first TNCs used in the facility were two used by the Metal Fabrication section. So far they have been used to assay low-content material such as rags and graphite. The two TNCs are located on the sides of gloveboxes, as shown in Fig. 40. Material enters the counting chamber through horizontal elevators.

B. Training (D. C. Amsden, R. H. Augustson, J. Hagen, and W. R. Severe)

Metal Fabrication section personnel took the DYMAC training course a week before moving into the new facility. Fifteen people attended the lecture and terminal training sessions. In a third session a week later, DYMAC personnel guided the trainees through an exercise of measurements and transactions designed to resemble their actual work. Each of two trainee groups used an off-line balance and a terminal to complete the exercise and learned how to use the terminals' on-line reporting capability to determine the location of their items in the inventory.

The first five chapters of the DYMAC User's Manual were written in time for the Metal Fabrication training class. The manual, now in draft form, will be expanded and polished before formal publication. Other chapters will be written about the NMO and the reporting capabilities of the DYMAC system. In addition, a series of appendixes will cover instrument operating procedures, process sheets for each section, error messages, and accountability locations in the facility (e.g., list of unit

processes, locations, accounts). Eventually the User's Manual will contain 200-300 pages of information for anyone using the DYMAC system at the plutonium facility. The manual will be published in a three-ring binder and change pages will be issued periodically.

C. Visitors

A steady stream of people visits the DYMAC project during the year. During this reporting period,

DYMAC personnel met with 19 visitors (16 from US agencies and private industry, 1 from the IAEA, 1 from the United Kingdom, and 1 from Japan) to brief them about the DYMAC system, to review LASL technical support to the IAEA, to discuss an integrated safeguards system using the Plutonium Processing Facility as a focal point, to exchange NDA and neutron coincidence techniques, and to guide the visitors through the plutonium facility.

PART 4

INTEGRATED SAFEGUARDS SYSTEMS AND TECHNOLOGY TRANSFER

GROUP Q-4

R. J. Dietz, Group Leader

J. P. Shipley, Alternate 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 conventional as well as alternative 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. By virtue of its overall cognizance of safeguards technology and needs, Q-4 also continues to provide technical information and assessments of both domestic and foreign fuel cycle facilities to DOE on a high-priority, as-needed basis.

During the past four months the Q-4 effort was evenly divided between the two line items: Safeguards Concept Definition for Fuel Cycle Facilities, and Technology Transfer and Support to DOE/SS, their subcontractors, and the safeguards industry.

In addition to these program status reports, the efforts of Q-4 are reported in journal papers, consultations, briefings, and topical reports at the conclusion of specific tasks. A list of publications issued during the report period is given at the end of this report.

I. SAFEGUARDS CONCEPT DEFINITION FOR FUEL CYCLE FACILITIES

The safeguards concept definition report, "Coordinated Safeguards for Materials Management in a Nitrate-to-Oxide Conversion Facility," has been published as LA-7011 (Ref. 24). The conversion report, outlined in Ref. 6, is the third of a series intended to treat each of the process facility modules characteristic of a complete national fuel cycle. The other two modules, mixed-oxide fuel fabrication and fuel reprocessing, were addressed in Refs. 25 and 26, respectively.

The pivotal role of the conversion process in safeguarding the whole nuclear fuel cycle against subnational diversion, or as part of a national nuclear nonproliferation policy, is underscored by development of the concept of the bonded crucial

facility (BCF). 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, respectively. The expanded complex then incorporates all inventory functions for the materials most attractive for diversion and can provide the opportunity for continuous monitoring of the production and consumption of fissile materials on a wide scale. Depending on the situation, the BCF could be operated under an umbrella of enhanced safeguards that might include multinational control.

The important role of safeguards for enrichment plants has been recognized through a series of four

meetings of representatives from various DOE contractors. The meetings culminated in an initial definition of the DOE/OSS Uranium Enrichment Plant Safeguards Program, which the Union Carbide Corp., Nuclear Division, will manage. Their immediate task is to develop a perimeter safeguards system for the GCEP at Portsmouth, Ohio. A system of perimeter safeguards is intended to assure that the enrichment plant is being operated in the

declared mode by (1) verifying the declared-stream materials balances, (2) detecting clandestine production of high-enriched uranium, and (3) detecting flows of undeclared feed, product, or tails streams through the perimeter. Efforts by Q-4 in this program are directed primarily toward the first of these functions. Centrifuge process design and operating data are being developed for the study.

II. SAFEGUARDS CONCEPTS FOR ALTERNATIVE FUEL CYCLE FACILITIES

Group Q-4 is providing support to the DOE/Nuclear Production Division's (NPD) Alternative Fuel Cycle Technology (AFCT) programs. These programs are administered by the Savannah River Operations Office under the technical guidance of SRL.

The first alternative, now being studied as a follow-on replacement for the conversion process studied in LA-7011, is the coconversion of coprocessed, mixed uranium-plutonium nitrate solutions via the General Electric COPRECAL process. This baseline COPRECAL facility is exactly the same size, in terms of plutonium throughput, as the reference facility described in LA-7011,²⁴ and it has a number of components operating in parallel in each part of the process in order to provide that capacity of 116 kg/day. This feature allows better materials accounting than for a single line, but accounting procedures are complicated by the resulting interdependent measurement sequences and instrument calibration errors.

Modeling of the COPRECAL process is complete to the degree required for materials accounting. The process model already has revealed one important characteristic of the process, namely, that the reasonable 5% variations in the precipitator contents during normal operations can cause 40% variations in product batch size. The ramifications of this discovery are still being discussed, but the preliminary results probably will not be acceptable without some process modification.

The measurements model is generally similar to that employed in the conversion process described in LA-7011.²⁴ However, the material forms and compositions and the process equipment are sufficiently different that Group Q-1 has been asked to provide

guidance on appropriate measurement techniques and capabilities. A draft report on safeguards concepts for the COPRECAL process will be issued soon.

The other current, major effort in this area is the design of safeguards systems for fuel cycle facilities operating under the $^{233}\text{U}/^{232}\text{Th}$ fuel cycle. A thorium-uranium fueled reactor is designed to generate electricity by burning fissile fuel that can contain ^{233}U , ^{235}U , or ^{239}Pu and to convert ^{232}Th to ^{233}U . Thus, two types of fuel are required—the fissile and the fertile. These may be present in the same fuel rods, or the fertile material may be present as a blanket around the fissile core. The use of ^{239}Pu as the fissile fuel will not be considered in this design study.

The fissile fuel consists of UO_2 fuel elements similar to PWR fuel elements for conventional light water reactors (LWRs). Cladding may be Zircaloy or stainless steel. The initial loading of the reactor core (referred to as the prebreeder) uses ^{235}U as the fissile material, with an enrichment of up to 93%. In addition to the 93% enrichment (high enriched), enrichments of 20-30% (medium enriched) and 10% (low enriched) are being considered. The thorium/uranium ratio is nominally between 5 and 10, depending on reactor design. The fuel from the initial loading still contains significant amounts of ^{236}U when the fuel is discharged, and recovery of the ^{235}U value is desirable. However during burnup, significant amounts of ^{236}U , a neutron poison, are produced. Hence, it may be desirable to reprocess the uranium only once; the second discharge would be discarded without reprocessing. Some plutonium also is produced in the initial fissile loading from neutron absorption by ^{238}U as well as by buildup from ^{236}U . The uranium/plutonium ratio will depend

on the ^{235}U enrichment of the fuel. The plutonium produced can be coprocessed with the recovered uranium and used to increase fissile content of the fuel or it can be cycled to high-level liquid waste with the fission products.

The reprocessing of thorium/uranium fuels differs significantly from reprocessing in the uranium-plutonium cycle. The head end for oxide fuel elements is similar to that for LWR fuels, consisting of a chop-leach process. However, for UO_2/ThO_2 fuels traces of HF are required to effect dissolution, and the presence of fluoride must be considered in subsequent analytical schemes.

Two separate solvent-extraction systems are required to process the fissile and fertile fuels. A Thorex (thorium recovery by extraction) process for recovering ^{233}U in the fertile fuel is the more important for safeguards. A conventional Purex (plutonium-uranium recovery by extraction) process for recovering uranium from fissile fuel can also recover plutonium, but the characteristics of the uranium and plutonium are not necessarily similar to LWR fuel.

For the Thorex process the fertile particles are dissolved in a solvent 10-13M in HNO_3 , 0.05M in HF, and 0.1M in $\text{Al}(\text{NO}_3)_3$ (*Thorex solution*). The solution is centrifuged to remove solids that presumably consist primarily of Zircaloy fines and metallic fission-product ingots. The metallic ingots could contain some ^{233}U .

The clarified solution containing uranium, thorium, and fission products is heated and steam-stripped to remove most of the HNO_3 and HF, and the HNO_3 is adjusted to 1M. A three-cycle solvent-

extraction system purifies uranium and thorium. In the first cycle, the high-acid extraction, uranium and thorium are removed from most of the fission products into a solution of 30% TBP in n-dodecane. The feed to the first cycle contains 200-250 g/l of thorium and 5-15 g/l of uranium. The uranium and thorium (and any plutonium and neptunium) are stripped into 0.1M HNO_3 and concentrated to ~250 g/l of thorium. In the second cycle, the low-acid extraction, the HNO_3 is adjusted to 0.1M and uranium and thorium are again extracted into 30% TBP in n-dodecane for further purification from fission products. An acid-strip column then removes most of the thorium. The uranium is then stripped in a third column into 0.01M HNO_3 . This solution is concentrated to ~10 g/l of uranium, sulfamic acid is added, the HNO_3 is adjusted to 3.6M, and uranium is extracted in 5% TBP in n-dodecane (the third cycle of uranium extraction). The uranium is stripped into 0.01M HNO_3 and becomes the uranium product from the Thorex process.

Literature for this study has been obtained from SRP, ORNL, ICPP, General Atomics, and Westinghouse. A reference flowsheet for the reprocessing plant is being developed that is based on low enriched UO_2 (<20% ^{235}U or <13% ^{233}U). A draft of an LA-series report entitled "A Critical Review of Analytical Techniques for Safeguarding the Thorium-Uranium Fuel Cycle" is being reviewed. An outline was prepared for a draft report entitled "Preliminary Management in a Uranium-Thorium Fuel Reprocessing Plant" and work on the report has begun.

III. INTERNATIONAL SAFEGUARDS

Group 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.

Task 3 has been addressed by the issuance of two reports. The first was "Preliminary Concepts for Materials Measurement and Accounting in Critical Facilities,"²⁷ which has been published. The second report, "Concepts for Inventory Verification in Critical Facilities," will shortly be circulated in draft form for review. The contents of the study were presented in detail in the previous status report.⁶

IV. DEVELOPMENT OF SAFEGUARDS DESIGN AND EVALUATION METHODOLOGY

The development and refinement of analysis and evaluation techniques compatible with the decision-analysis framework is continuing. We now have the ability to handle complicated correlations in large amounts of data and to search for diversions occurring in any pattern. The main results were presented in an invited paper, "Decision Analysis for Nuclear Safeguards."²⁸ Further results will be presented at other nationwide meetings during the next reporting period and at the Safeguards System Training Course at LASL, May 9-11, 1978.

Assessment of system security and reliability plays an important role in the design and evaluation of effective safeguards systems. Toward these ends, a series of simulation modules for security characteristics have been developed. A decision framework is established to analyze the risk in terms of the actual or perceived value of what is to be protected. This is followed by examining the threat environment and incorporating countermeasures having sufficient effectiveness to reduce the probability of penetration in each threat area. Conceptual designs are then prepared for potential systems architectures, with protection mechanisms, and this information is incorporated into SECSIM security simulation models. From these a security simulation profile is generated indicating systems strengths and weaknesses. Parameters are changed and the process is repeated until desired requirements are met. Models in this study used to characterize candidate systems security behavior are comprehensive in accommodating the full range of threats and protection levels. They are tailored to each facility by weighting input parameters in accordance with the design configuration and environment selected. This method of analysis is applicable to facilities in the conceptual design, detailed design, construction, and operational stages of evolution. Although the majority of security criteria are common to all facilities, individual specific facility vulnerability data are deliberately suppressed.

The results of these simulations provide top management, security officers, system operators, systems programmers and other center staff with a set of evaluation criteria with which to access their facility for its security vulnerability. They also

provide a basis for determining the effect of design changes for upgrading and improvement. Inter-facility comparisons can be made by using a common set of references.

Emphasis is placed on providing as much structure as possible for the parameters associated with computer security. Barrier evaluation is similar to security in general in which subjective as well as objective judgments must be made to determine effectiveness. The range of 0 to 1 is chosen, where 1 represents a totally effective barrier. The individual facility threat environmental factors are incorporated as weighted inputs along with the corresponding attributes of each barrier, and a numerical value or range of values is assigned. These are then introduced into the appropriate segment of the model and a composite effectiveness is computed for that sublevel. This is repeated for the next event-sequence level until an index is generated in that respective category. A two-dimensional horizontally layered and vertically structured network-decomposition methodology is used.

The algorithms in the models contain branch points allowing for redundant barriers having generally dissimilar characteristics. A range of conditions can be imposed from at least 1 of N to N of N, or exactly M of N, surviving, and the corresponding effectiveness can be determined. A sensitivity analysis shows the change on system effectiveness with changes in individual barrier characteristics. The simulation is capable of being run in real time to give current estimates of the security effectiveness as the threat environment and individual barrier effectiveness change.

Similarly, series of information system architecture reliability simulations, using the Q-4 code RELSIM, have been conducted for the purpose of designing data transmission networks with reduced downtimes. Triply redundant distributed structures exhibit projected downtimes of 0.06 h/yr. These are contrasted with more conventional, doubly redundant, tandem connections having downtimes of 9 h/yr. The overall data acquisition, transmission, processing, and storage system downtime has been decreased to 1.5 h/yr. This level of performance can be attained with commercially available system modules.

Additional RELSIM programs have been coded and run to provide overall system estimates of the probability that a fraction such as 95 out of 100 non-identical system sensors are functioning normally.

Another is a real-time version to be run in operating facilities, allowing updated reliability projections to be computed rapidly as changes occur or anticipated changes in systems architecture are contemplated.

V. TECHNOLOGY TRANSFER AND TECHNICAL SUPPORT

Group Q-4 personnel participated in numerous briefings, visits, and consultations in all the above areas with people from DOE/OSS, Brookhaven-ISPO, Sandia Laboratories/Albuquerque, ORNL, Burns and Roe, Westinghouse, the IAEA, the United Kingdom, and West Germany. In addition, L. E. Scheinman and others of the State Department staff were given formal presentations on Q-4 activities and capabilities.

Direct technical support was provided to DOE in the form of safeguards systems descriptions for the congressional study report, "Barnwell Nuclear Fuels Plant Applicability Study." Work sessions were attended at Barnwell, DOE Headquarters, ORNL, and Sandia Laboratories/Albuquerque. Group Q-4 staff also participated in the development of the program plan for the Fuels Refabrication and Development Program arranged by Battelle-PNL. The program plan will be circulated for review sometime during the next period.

The review of controllable-unit accounting developed by Mound Laboratory was completed in January, as discussed in Ref. 6. A summary of techniques that might be effective in analyzing low-

quality, historical MUF data was prepared in response to queries from DOE/OSS.

As always, the manifold efforts of the Q-4 staff continued to be reported in the technical literature and at national meetings. One staff member chaired a symposium on "Nondestructive and Analytical Chemical Techniques for Nuclear Safeguards" for the American Chemical Society's annual meeting, and another staff member presented an invited paper there on decision analysis. Other staff members reviewed the result of Q-4's systems studies efforts at the 2nd Int. Conf. on Nondestructive Evaluation in the Nuclear Industry²⁹ and at the American Nuclear Society Meeting on the Back End of the LWR Fuel Cycle.³⁰

In addition, Q-4 personnel have participated in writing ANSI standards, including the draft ANSI N15.36, "Materials Control and Quality Assurance," which is being revised for formal review. Safeguards terms have been prepared as part of the work for Writing Group N9.9, which is revising ANSI N1.1, "Glossary of Terms in Nuclear Science and Technology."

VI. MISCELLANEOUS

The increasing responsibilities of Group Q-4 for a widening range of safeguards systems expertise place more and more emphasis on timely and effective communication. Computer graphics support for the group was significant during the reporting period. Some improvements were made to the decision analysis movie for a fuel reprocessing plant, and another movie was generated for the conversion process. Many computer-generated, 35-mm presentation slides were made in support of briefings, presentations, and the Safeguards Systems Training Course.

During this period, two new data analysts were hired by the group and should provide effective support for the computer graphics, modeling, and simulation effort.

Likewise, technical support for modeling, simulation, and data-analysis exercises has become extremely important. The Safeguards System Simulation and Evaluation computer has been placed on-stream. Problems associated with the initial delivery of the two 12-MB disk drives were resolved and the PRIMOS III multi-user executive system installed. The UT-200 emulator hardware and software enable

use of the system for remote job entry to the central computer facility (CCF).

The system has been used extensively in developing and running programs for systems reliability studies as well as for the word processing of reports, and the possibility of locally generated computer graphics is being investigated.

Q-4 staff attended the PRIMOS III Internals course held in Boston. The course dealt intensively

with the internal workings of the executive. Some parity errors in the system will have to be tracked down. Occasional disk read errors also occur. Aside from these few problems, the computer is functioning satisfactorily. The executive is easy to use and provides the flexibility necessary for these applications.

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GLOSSARY

ACC	Allied Chemical Corp.
AFCT	Alternative Fuel Cycle Technology
AWCC	active well coincidence counter
BCF	bonded crucial facility
BWR	boiling water reactor
DYMAC	dynamic materials control
FCA	fast critical assembly
FFTF	Fast Flux Test Facility
FWHM	full width half maximum
GAT	Goodyear Atomic Corp.
GCEP	(Portsmouth) Gas Centrifuge Enrichment Plant
HEDL	Hanford Engineering Development Laboratory
HEU	high-enriched uranium
HLNCC	high-level neutron coincidence counter
HPFL	High Performance Fuel Laboratory
IAEA	International Atomic Energy Agency
ICPP	Idaho Chemical Processing Plant
ISPO	International Safeguards Project Office
LWR	light water reactor
MCA	multichannel analyzer
MTR	materials testing reactor
NBS	National Bureau of Standards
NDA	nondestructive assay
NMO	nuclear materials officer
PWR	pressurized water reactor
SAI	solution assay instrument
SGS	segmented gamma scanner
SNM	special nuclear material
SSAS	small sample assay station
THENCS	thermal-neutron coincidence counting system
TNC	thermal-neutron coincidence counter
ZPPR	Zero Power Plutonium Reactor