

LA--9373-PR

DE83 002917

LA-9373-PR  
Progress Report

UC-15  
Issued: September 1982

## Safeguards and Security Status Report

August 1981—January 1982

Compiled by  
J. P. Shipley

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Los Alamos, New Mexico 87545

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**SAFEGUARDS AND SECURITY STATUS REPORT**  
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**ABSTRACT**

From August 1981 through January 1982, the Los Alamos Safeguards and Security Program was involved in many activities that are described in the four parts of this report: Nuclear Facility Safeguards Support, Security Development and Support, Safeguards Technology Development, and International Support. Part 1 covers those efforts of direct assistance to the Department of Energy (DOE) and the Nuclear Regulatory Commission licensee facilities. This assistance varies from consultation on materials accounting problems, through development of specialized techniques and devices, to comprehensive participation in the design and implementation of advanced safeguards systems. In addition, a series of training courses in various aspects of safeguards helps make the technology more accessible to those who must apply it. Part 2 concerns a relatively new set of activities at Los Alamos aimed at the security of information and computer systems. The focus this period has been on furthering the development of the Computer Security Center, which provides the basis for encouraging and disseminating the emerging technology. Part 3 describes the development efforts that are essential to continued improvements in the practice of safeguards. Although these projects are properly classified as developmental, in every case they are directed ultimately at recognized problems that commonly occur in operating facilities. Finally, Part 4 covers international safeguards activities, including both support to the International Atomic Energy Agency and bilateral exchanges. In addition, enrichment plant safeguards, especially those concerning the Gaseous Centrifuge Enrichment Plant, required a significant portion of our resources. These efforts are beginning to provide substantial returns on our investment in technology transfer. Not only has the level of safeguards effectiveness been increased, but we also are receiving the benefits of field experience in operating environments.

Most projects described here were sponsored by the DOE Office of Safeguards and Security. However, safeguards activities that have other sponsors also are reported.

## PART I. NUCLEAR FACILITY SAFEGUARDS SUPPORT

### I. OAK RIDGE NATIONAL LABORATORY (ORNL)/Y-12

#### A. Safeguards Systems Support (*H. A. Dayem, J. L. Sapir, and C. A. Ostenak, Q-4*)

Los Alamos is providing safeguards systems support to ORNL/Y-12 in two areas: accountability in the UF<sub>6</sub>-to-uranium-metal bomb-reduction process and estimation of in-process inventory in the secondary-extraction columns.

For the bomb-reduction process, our objectives are to recommend upgraded measurement techniques and accountability procedures and to formulate expected inventory difference (ID) behavior and estimate the ID variance. Because information from ORNL is insufficient to carry out these activities, we requested additional information.

For the secondary-extraction columns, Los Alamos is undertaking combined measurement and modeling efforts to develop a pulsed-column in-process inventory estimator based on a minimum number of measurements. The measurement efforts are described in Sec. I.B.

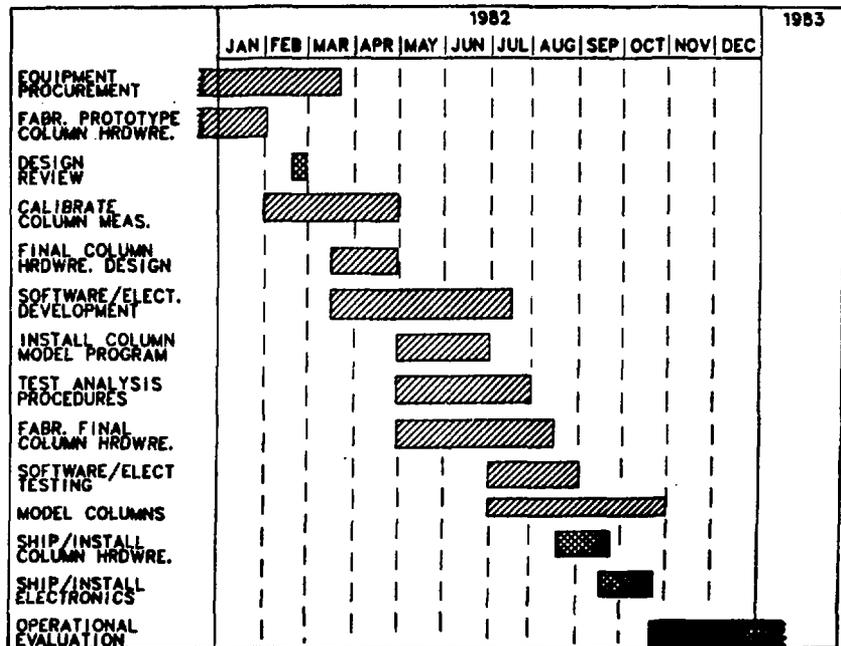
Data are being assembled on the operating characteristics of the Y-12 scrap-recovery secondary-extraction pulsed columns. We will model these columns

to determine their uranium concentration profiles and total uranium inventories, then compare these calculations with the measured and column dump data. The modeling and measurement results will be used to determine the minimum number of measurements required and to develop an estimator based on these measurements.

#### B. Nondestructive Assay (NDA) Measurements for Inventory of Highly Enriched Uranium (HEU) in Solvent-Extraction Pulsed Columns (*P. Russo, T. Marks, M. Stephens, and R. Strittmatter, Q-1*)

1. Introduction. Information from in-plant measurements of pulsed columns with portable NDA equipment<sup>1,2</sup> is being used with recommendations from facility personnel to design a prototype NDA system for inventory of HEU in solvent-extraction pulsed columns. This system will be designed and fabricated at Los Alamos and will be evaluated at the Y-12 facility for recovery of HEU. A tentative schedule for completion of design, fabrication, testing, and preliminary evaluation is shown in Fig. 1.

Fig. 1. Tentative schedule (calendar year 1982) for design, fabrication, testing, and evaluation of the Los Alamos system for inventory of Y-12 pulsed columns.



The prototype system will consist of an array of six shielded NaI(Tl) detectors that mount simultaneously along the 9-m length of any one of the three pulsed columns in each of two secondary solvent-extraction systems at the Y-12 facility. The six signals from the NaI(Tl) detectors will be multiplexed (with routing) into a single CAMAC analog-to-digital converter (ADC). The spectra will be stored separately by a CAMAC-based programmable multichannel analyzer (MCA) system, which will automatically analyze uranium concentration at the six vertical locations along the column and will compute the concentration profile in the vertical dimension. The HEU inventory for each secondary solvent-extraction system will be deduced from the uranium concentration profiles of the three columns in each system (see Fig. 2).

**2. Equipment.** Figure 3 shows the required electronics and a single shielded detector unit for the prototype system. At the right, a single detector (with preamplifier tube base) is shown on top of the table. The detector mounts inside the horizontal, cylindrical collimator/shield, which attaches to a holder that clamps to the column. The collimator/shield is lead encased in stainless steel, and the clamp and holder are stainless steel. The

collimator is 7.5 cm long and 2.5 cm in diameter. The mechanical assembly is clamped to a section of stainless steel pipe designed to simulate the Y-12 column configuration. (This simulated column section will be filled with well-characterized uranium solutions for calibration purposes.) A similar clamping mechanism is proposed for the Y-12 installation. Thirty-six clamps (six per column on each of the six columns) will be required for the test phase to accommodate the six shielded detectors. The pulsed-column measurements will be performed one column at a time during the test period.

The detectors are 3.2- by 1.3-cm-thick NaI(Tl) crystals integrally mounted to 12-pin photomultiplier tubes, 3.8 cm in diameter. Power for the preamplifiers in the tube bases is tapped from the high voltage to minimize the number of cables. Each detector will have a radioisotopic source ( $^{241}\text{Am}$ ) mounted near the crystal, in fixed geometry, for count-rate loss correction and gain stabilization.

The MCA display cathode-ray tube (CRT) and keypad are shown in the upper portion of the rack in Fig. 3. Adjacent to the CRT is a minirate for CAMAC modules. The capability for multiplexing, routing, and digitizing up to 16-detector inputs can be implemented

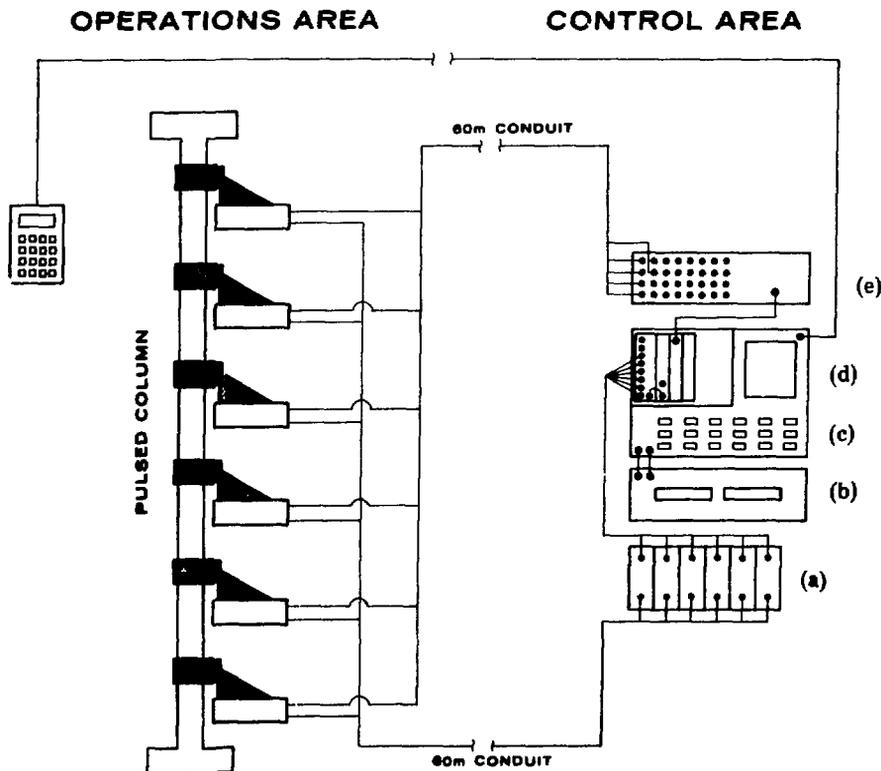
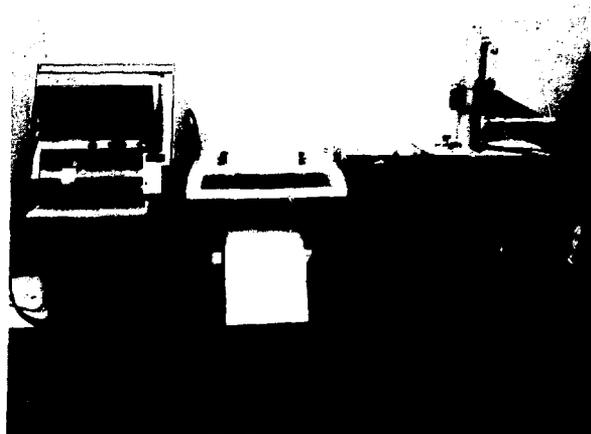


Fig. 2. The pulsed-column measurement system design. Six shielded NaI(Tl) detectors are shown (at left) mounted to the pulsed column. The signal and high-voltage cables from the detector preamplifiers (located in the photomultiplier tube bases) run through approximately 60 m of shielded conduit to the control area outside the operations area. The electronics in the control area include (bottom to top) the linear amplifiers (a); a dual floppy disk unit (b); the programmable MCA keypad and CRT (c); a CAMAC minirate (d), containing, left to right, an 8-input multiplexer-router, an ADC, the power supply interface, and a live timer; and a 32-channel, programmable, CAMAC-based high-voltage power supply (e).

Fig. 3. Assembly of equipment for pulsed-column inventory system. The prototype system will include (left, top) a CAMAC-based programmable MCA with CRT display, keypad, and CAMAC modules for multiplexing (with routing) and digitizing the linear signals. Stabilized linear amplifiers (left, bottom) will be used on each detector. Spectra will be stored on floppy diskettes (left, middle). Hard copy will be generated at a main terminal (center) in the control area. A remote terminal (right of the analyzer keypad) will be located in the operations area. Each of six detectors with tube base preamplifier (top of table at left) will fit into a shield that mounts onto the pulsed column (top of table at right). The section of column is the sample cell designed for the calibration measurements. Beneath this cell are the electronics for the calibration measurements, consisting of a NIM-powered linear amplifier (left), ADC (center), and compact portable MCA.



within the limits of the 8-slot crate. (Expansion to 48-detector inputs can be accomplished by adding a second CAMAC crate. Analyzer memory can be expanded accordingly.)

Beneath the MCA keypad (Fig. 3) is a dual, double-density floppy disk unit for software and data storage. The bin beneath this unit contains five stabilized linear amplifiers and a high-voltage power supply. A hard-copy terminal is to the right of the electronics rack, and a portable (hand-held) terminal, appropriate for remote-control operation, is adjacent to the MCA keypad.

A programmable, CAMAC-compatible power supply with 32 independent high-voltage outputs (shown in Fig. 2 but not in Fig. 3) is planned to eliminate the need for manual adjustment of the gains on individual detectors following power-up or during extended operation or substantial temperature variations. Manual adjustment of gain is inconvenient but feasible in a six-detector system and impractical in larger systems.

Hardware for the prototype system will be designed and assembled for operation with six detectors. However, the system is immediately expandable to 32 detectors by adding the extra (26) detectors and linear amplifiers plus one extra CAMAC crate and ADC and three multiplexers. Expansion to 48 detectors requires another high-voltage power supply and ADC, two multiplexers, and the extra (16) detectors and linear amplifiers. The available memory and the software design will be compatible with these hardware expansion capabilities.

**3. Calibration.** The 186-keV gamma ray of  $^{235}\text{U}$  will be used to assay uranium concentration in the HEU solutions. The calibration will be determined by calcula-

tions and verified empirically using solutions of HEU from the Los Alamos Group CMB-8 enriched uranium recovery facility. The sample cell designed for this purpose (upper right, Fig. 3) is fitted with stainless steel sieve plates in the lower half, positioned at 2.5-cm intervals by stainless steel stator rods to reproduce the Y-12 column configuration. Because the calculated calibration does not include the effects of the plates and rods, the verification measurements will be performed on the upper half of the cell. An empirical correction for the plates and rods will be applied, based on the measurements on the lower half of the cell.

The cell will be filled with solutions of 93%-enriched uranium stored at the CMB-8 facility. A sample will be withdrawn from the cell and assayed by the CMB-8 uranium solution assay system<sup>3</sup> to give a reference value. Selected samples also will be analyzed destructively. The measurements will cover a range of uranium concentration from 10 to 300 g/l.

Calibration measurements will be made by compact electronics (see the lower shelf of the table in Fig. 3) consisting of a nuclear instrument module (NIM) bin that powers a programmable MCA and the ADC, a stabilized amplifier, and high-voltage power. The shielded detector and the amplifier are the same units to be shipped to Y-12. An  $^{241}\text{Am}$  source will be used for gain stabilization and count-rate loss corrections. A normalization measurement using a small HEU foil mounted on a tungsten holder that inserts into a slot in the collimator/shield will be performed before each solution assay measurement. Background will be measured before each solution assay with a solid piece of tungsten (6 mm thick) inserted in the slot.

4. **Proposed Assembly and Test Procedures.** We proposed that all electronics downstream of the tube-base preamplifiers be located outside the Y-12 operations area to minimize the effects of the hostile plant environments. Therefore, approximately 60 m of cable for the signal and high voltage will be required for each detector. We further proposed that each detector be mounted at one of six column locations (one per column) accessible to the operator's reach from a given floor level. (There are four such levels in the operations area.) The two secondary solvent-extraction systems are separated by 3 to 4 m. Thus, only a few meters of cable will give each detector the flexibility to be positioned on any one of the six columns at some height accessible from the appropriate floor level.

Inventory measurements with the test system will be performed column by column, and the operator will initiate each column measurement at the control terminal. Count times for measurements of a given column will be, typically, less than 5 min. Following measurements on a given column, each shielded detector will be removed from its mounting bracket on the column and positioned in the mounting bracket on the next column. Removing, shifting, and repositioning a detector should require about 1 min. A portable control terminal will be carried by the operator to minimize the time required to initiate each assay. A plug-in connection (to the central processing unit interface) for this terminal should be located at each floor level in the operations area.

The inventory in the secondary solvent-extraction system includes significant amounts of HEU in solution volumes in the plumbing external to the columns. The solution volume of each external line and the vertical column location of the feed to each line will be used, with measured column concentration profiles, to deduce the inventory in the external portions of the system. The accuracy of the (deduced) external inventory and that of the column inventory will depend on the operations procedures followed at the time of the concentration measurements. Possible procedures may be grouped into four categories:

- (1) Concentration measurements are performed during pulsed operation.
- (2) Concentration measurements are performed following simultaneous shutdown of column and pulsing.
- (3) Concentration measurements are performed after column shutdown during pulsing.

- (4) Concentration measurements are performed after column shutdown and following a period of continued pulsing for phase disengagement.

The deduced inventory in the external plumbing is least susceptible to error under procedure 1, whereas procedures 3 and 4 effectively erase the information (concentration profile during operation) required to deduce the inventory. Procedure 2 might be effective with an additional, substantial empirical effort to obtain accurate deductions of external inventory. However, this procedure, and any that involves measuring the static columns, introduces other complications. A static column has a well-defined interface at which a large, discontinuous change in the measured concentration profile is observed.<sup>1,2</sup> Therefore, inventories obtained from measurements performed on the static column require measurements close to and on either side of the interface. Because the location of the interface is variable, such measurements require repositioning the detector(s) on either side of the interface. This repositioning likely involves two separate measurements with the same detector mounted on a movable bracket on the glass portion of the column where the interface is visible. This approach can be used with procedures 2 and 4, but it adds complexity to the manual effort. Furthermore, the interface probably will be below the (upper 2 m) glass section of the pulsed column with complete phase disengagement (procedure 4), so it could not be located visually. Therefore, the inventory measurements should be performed during pulsed-column operation as defined by procedure 1.

5. **Evaluation of Inventory Capability.** The concentration profile for each column based on the six measured concentrations can be accomplished by constructing a smooth curve through the measurement results plotted vs column height. In addition, pulsed-column models<sup>4</sup> will be used to deduce column inventories from measurement results.

The models can, in principle, determine the column inventory during pulsed operation from as few as three concentration measurements plus the flow rates on the input product and waste streams at the time of measurement. Success in these applications could reduce by a factor of 2 the number of detectors required to measure a single column. In all cases, the solution volumes and vertical locations of the feed to each external line in the secondary solvent-extraction system are required to

obtain the system inventory. Column dump experiments during the evaluation period at Y-12 will aid in assessing the accuracy of the inventory capability of the measurement system. A detailed plan of test procedures during operation for evaluation of the inventory capability of the Los Alamos system will be prepared jointly by Los Alamos and Y-12.

### C. Automated Closed-Loop Process Control (R. Strittmatter, T. Marks, and P. Russo, Q-1)

During the March 1981 Los Alamos visit to Y-12, the intermediate evaporators at the Y-12 HEU recovery facility were identified by Y-12 personnel as the source of one of three materials accounting problems to be treated with highest priority. The intermediate evaporator is a recirculating, long-tube, vertical, steam-jacketed evaporator with a 13-cm-diam heat exchanger. The recirculation return loop is a 9-cm-diam stainless steel pipe. The intermediate evaporator concentrates the uranium solution that feeds the secondary solvent-extraction system. During inventory, the evaporator is drained, and an estimated 500 g of uranium is left as holdup. However, as a result of the chemical nature of the primary solvent-extraction product, which is the input to the intermediate evaporator, uranium can precipitate and collect on the evaporator walls and bottom if the evaporation process is not properly controlled. In one case, 9 kg of uranium was removed from an evaporator after improper operation. The immediate accounting concern is the ability to measure holdup in the evaporator after drainage. The real desire, however, is to improve the process control so that uranium precipitation does not occur, thus alleviating the major materials accounting concern for the evaporators. The evaporator now is operated manually, based on sample hydrometer readings.

Manual operation of the evaporator system involves transferring a solution sample from the return loop to the at-line hydrometer for a 10% reading on solution density. The process of transfer and measurement requires at least 1 min, a delay that can allow the evaporation process to proceed beyond its optimum end point before the appropriate inlet and outlet valves are adjusted. The consequences of overconcentration, in addition to uranium precipitation, include possible safety hazards.

A NaI(Tl) probe was used to perform a series of 1-min measurements of uranium concentration in the secondary intermediate evaporator solutions in March 1981.

The probe was held up to the solution return loop pipe. These 1-min passive assays based on the 186-keV gamma ray demonstrated that a very nearly real-time measurement (in 10 s) could provide a 1% result on  $^{235}\text{U}$  concentration. For constant  $^{235}\text{U}$  enrichment, this approach offers possible significant improvement over the present operations technique.

A prototype instrument designed to provide process control and improved materials accounting for the Y-12 intermediate evaporators has been assembled from existing equipment. This prototype uses the NaI(Tl) detector fixture fabricated for the Y-12 solvent-extraction column measurements, a microprocessor-based data-acquisition and systems-control device developed for the gas-phase  $\text{UF}_6$  enrichment monitor, and commercial nuclear electronics. The system has the potential of providing near-real-time uranium concentration data and automated control of valves regulating the input and output flows of the evaporator (see Fig. 4).

We proposed that a valve manifold be assembled to test the prototype system and that the system be

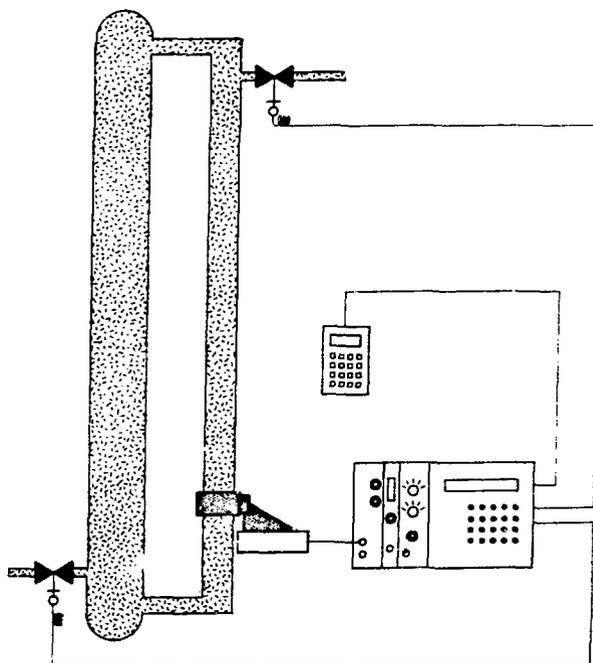


Fig. 4. The prototype control system for intermediate evaporator. A shielded NaI(Tl) detector is mounted to the evaporator return-loop pipe. The electronics (mounted nearby in an environmental cabinet) consist (left to right) of a NIM-powered linear amplifier, high-voltage supply, ADC, and programmable MCA. A portable terminal with LCD would be used by the operator to interact with the system and would provide near-real-time-measurement results for operation in the manual mode.

designed to operate either manually or automatically. Because testing the Y-12 pulsed-column inventory system involves concurrent testing of the essential prototype instrument, we suggested that the tentative work sched-

ule for the pulsed-column system be applied also to the prototype evaporator control system. A separate evaluation plan for the evaporator control system will be prepared by Los Alamos and Y-12.

## II. HANFORD ENGINEERING DEVELOPMENT LABORATORY (HEDL)

### A. Fuels and Materials Examination Facility (FMEF) (*R. G. Gutmacher, A. L. Baker, K. C. Greenough, E. A. Kern, R. S. Marshall, and D. P. Martinez, Q-4*)

Assistance to HEDL in safeguards systems design has focused on the FMEF and the Secure Automated Fabrication (SAF) process line. Computer modeling and simulation of the SAF process were initiated as essential steps in the safeguards system design. The SAF process line will fabricate 6 MT of mixed-oxide (MOX) fuel annually for the Fast Flux Test Facility (FFTF) and for breeder reactors such as Clinch River.

The Los Alamos MODEL code was modified to use the SLAM-II, rather than the GASP IV, simulation language. This modification will simplify modeling complex processes, such as the SAF process, and yield process-related information of interest to HEDL in addition to data needed for safeguards.

The pellet grinding and inspection portion of the SAF process was modeled. Computer simulation of 1 yr of operation required 3.5 min on a Prime 750. Data on reliabilities of process equipment and repair times, estimates of holdup in process equipment, times required for setup and routine maintenance, and details of conveyor operation will be incorporated in a refined model when the data are available.

We expect to complete the SAF process model in July 1982, and the documented model and computer program will be available to HEDL. In subsequent work, a measurement model superimposed on the process model

will form the basis for the design of the SAF line safeguards system in FY 83.

Identification and investigation of potential FMEF vulnerabilities are continuing. Holdup in process equipment and potential queuing problems at the secured storage facility were chosen for detailed investigation. We also will assist HEDL in computer security studies.

### B. Wet Scrap Recovery (WSR) (*R. G. Gutmacher, A. L. Baker, K. C. Greenough, E. A. Kern, R. S. Marshall, and D. P. Martinez, Q-4*)

Discussions with HEDL personnel concerned with the WSR operation were held to establish possible future interactions. The WSR will be a pilot plant to dissolve contaminated MOX scrap, purify the solution by solvent extraction, precipitate mixed uranium-plutonium oxalate by the COPRECAL process, and convert the oxalates to oxide. Partial operation of the process with uranium is scheduled for FY 83, and full operation with plutonium will begin in FY 85. The capacity of the WSR operation will be 0.5 kg of MOX scrap/h. HEDL has expressed interest in detailed process modeling, especially of solvent-extraction column inventories, that would aid in development of materials accountability procedures, in selection of in-line or on-line instrumentation, and in implementation of near-real-time materials measurement and accounting techniques.

## III. CONSOLIDATED FUEL REPROCESSING PROGRAM

(*H. A. Dayem, R. G. Gutmacher, E. A. Kern, J. T. Markin, D. P. Martinez, and C. A. Ostenak, Q-4*)

### A. The Hot Experimental Facility (HEF)

A draft report, "Materials Accounting in an FBR Fuels Reprocessing Facility: Optimal Allocation of Measurement Uncertainties," was completed and issued

for review. Comments from the DOE/Office of Safeguards and Security (OSS) and ORNL were incorporated in the final report.<sup>5</sup>

This report is the first in a series, in support of the DOE Consolidated Fuel Reprocessing Program

(CFRP), for developing and evaluating advanced materials measurement and accounting systems (MMASs) for fast-breeder-reactor spent-fuel reprocessing concepts. The study's purpose was to propose an MMAS for the HEF that uses current measurement technology, then to calculate optimal measurement uncertainties that meet specified materials accounting performance goals.

The HEF process has several design features that constrain the application of advanced materials measurement and accounting techniques. The dissolution and feed preparation area, in particular, has novel features that will require additional testing on a laboratory and pilot-plant scale before incorporation into the HEF. These features include the voloxidizer, the continuous dissolver, and the constant-volume accountability tank. Other areas of concern having unknown impact include the estimation of in-process inventories in solvent-extraction contactors and other process vessels, coprocessing, recycle streams, and the inaccessibility of nuclear materials for at-line analysis. We recommend that the HEF design philosophy be revised to permit penetration of the cell wall by sampling lines and to allow location of nondestructive analysis instrumentation in a controlled-access instrument gallery. Such a revision would provide more timely information for process control and materials accounting.

The HEF MMAS combines conventional materials accounting and near-real-time accounting (NRTA) and serves several functions, including process monitoring, domestic safeguards, and international safeguards. It uses sampling and chemical analysis, weight and volume measurements, and NDA instrumentation, supported by data-base management and data analysis techniques. We described a conventional accounting strategy that divides the facility into five materials balance areas (MBAs) and formulated two NRTA strategies that augment the MBA structure. In strategy 1, the feed preparation processes were treated as one unit process accounting area (UPAA), called UPAA1, and the chemical separations processes were treated as another UPAA (UPAA 2/3). In strategy 2, the chemical separations UPAA was further subdivided into two UPAA's: UPAA 2 (codecontamination/partitioning processes) and UPAA 3 (uranium-plutonium copurification processes).

Measurement points for the NRTA strategies were identified, and applicable measurement types and errors representative of current technology were chosen, based on materials and process descriptions. The reference

measurements are used for process control and materials accounting.

## B. Materials Accounting System Optimization

Optimization techniques were used to calculate measurement uncertainties so that performance goals for detecting materials loss are achieved while the total development cost of the instruments is minimized. The cost of improving each measurement uncertainty component is determined by a hyperbolic cost function. Therefore, where the calculated measurement uncertainty is less than what is currently achievable, a development cost is imposed. Because the cost function is nonlinear, we used a nonlinear optimization technique to calculate measurement uncertainties that minimize the instrument development cost.

Measurement uncertainties that meet each of the four accountability performance goals were calculated for each UPAA and for several cases of instrument recalibration. For each UPAA, values for measurement uncertainty components were constrained by specific ranges and by the materials balance standard deviation equations for abrupt and protracted losses.

In this study, we calculated materials measurement uncertainties for instruments used by accounting systems that meet four different performance goals. The performance goals were chosen to represent a range of measurement capabilities and domestic and international safeguards goals. The four levels of NRTA performance goals are listed in Table I. The first two levels correspond to a likely range of measurement capabilities. The third and fourth levels correspond to desired international and domestic goals. The first performance goal is based on what should be possible if present state-of-the-art instrumentation is used. The second performance goal represents reasonable extrapolations of current technology. The third performance goal is based on the International Atomic Energy Agency (IAEA) proposed criteria, and the fourth is based on Nuclear Regulatory Commission (NRC) goals presently being considered.

Each performance goal includes detection of an abrupt (short-term) and a protracted (long-term) diversion with given detection and false-alarm probabilities. These quantities are used to calculate the maximum value of the materials balance standard deviation that will meet the performance goal. When examining these performance goals, remember that the plan-

**TABLE I. NRTA Performance Goals**

Goal		Amount @			Materials Balance	
		Detection (kg Pu)	Detection Time	Detection Probability	False-Alarm Probability	Standard Deviation Upper Limit (kg Pu)
1. Current technology	Abrupt	16	1 day	0.5	0.025	8
	Protracted	150	6 months	0.5	0.025	75
2. Improved technology	Abrupt	8	1 day	0.5	0.025	4
	Protracted	40	6 months	0.5	0.025	20
3. IAEA	Abrupt	8	7-10 days	0.95	0.05	2.4
	Protracted	8	1 year	0.95	0.05	2.4
4. NRC	Abrupt	2	1 day	0.5	0.025	1
	Protracted	2	6 months	0.5	0.025	1

throughput is about 113 kg of plutonium per day and that the chemical separation and feed preparation portions of the process can have an inventory of about 750 kg of plutonium.

Table II lists materials balance standard deviations for each of the UPAA's for both 1-day and 6-month materials balances using current measurement technology. The feed preparation UPAA (UPAA 1) has larger materials balance standard deviations than the other UPAA's have because it has more in-process inventory and the input transfer measurements (spent-fuel NDA) are not well characterized.

Table III lists the relative costs of developing the instrument systems that meet each of the performance goals. One cost unit is the "relative cost" of attaining a measurement uncertainty that is one-half that of current measurement technology. UPAA 2-3 with weekly recalibration of the plutonium concentration measuring instruments for the accountability and product sample tanks will meet goal 1. Hence, the total development cost of the system is zero. If periodic recalibration of key transfer measurements is performed, the relative cost of the system can be reduced by 30% or more. The relative cost of achieving goal 3 or 4 is between 20 and 50 times more than the cost of achieving goal 2.

A dynamic computer model of the HEF chemical separations process was developed. Modeling and simulation of the HEF chemical separations process allow the prediction of the dynamic behavior of materials flows and inventories and materials measurements over a wide range of operating parameters and allow the rapid accumulation of data representative of relatively long

operating periods. Because the optimization calculations used nominal values for process variables and did not include waste streams, a dynamic model of the measurement system, based on the calculated optimal measurement uncertainties, was applied to the simulated process data. Materials balance standard deviations from these simulations agreed with the optimization results.

**TABLE II. Materials Balance Standard Deviations with Current Measurement Technology**

	1 Day (kg Pu)	6 Months (kg Pu)
<b>UPAA 1</b> (feed preparation)		
No recalibration	11.6	636
Weekly calibration	11.6	373
<b>UPAA 2-3</b> (chemical separations)		
No recalibration	7.6	93
Weekly recalibration	7.6	72
<b>UPAA 2</b> (codecontamination/partitioning)		
No recalibration	5.9	289
Weekly recalibration	5.9	109
<b>UPAA 3</b> (copurification)		
No recalibration	5.5	284
Weekly recalibration	5.5	114

**TABLE III. Relative Costs of Achieving the Performance Goals**

	Goal 1	Goal 2	Goal 3	Goal 4
<b>UPAA 1 (feed preparation)</b>				
No recalibration	20	111	1947	2546
Weekly recalibration	9	64	---	---
Daily recalibration	---	53	942	1350
<b>UPAA 2 3 (chemical separations)</b>				
No recalibration	0.8	32	727	1544
Weekly recalibration	0	22	---	---
Daily recalibration	---	19	495	738
<b>UPAA 2 (codecontamination/partitioning)</b>				
No recalibration	11	74	1404	2023
Weekly recalibration	2	36	---	---
Daily recalibration	---	28	595	853
<b>UPAA 3 (copurification)</b>				
No recalibration	9	65	1263	1666
Weekly recalibration	1.7	29	---	---
Daily recalibration	---	21	518	735

For the feed preparation processes, the performance goals cannot be met by current measurement technology. Materials accounting is complicated by the in-process inventories and difficult-to-measure spent-fuel assembly transfers. To alleviate some of these problems, inventories should be reduced wherever possible, spent-fuel NDA techniques should be refined and standards should be developed, and frequent flushouts of the feed preparation process should be considered.

For the chemical separations area (UPAA 2 3), an abrupt loss-detection sensitivity of 15 kg of plutonium and a protracted loss-detection sensitivity of 150 kg of plutonium are attainable with current measurement technology. These loss-detection sensitivities have a 50% detection probability and a 2.5% false-alarm probability. If it is desirable to subdivide the chemical separations process and maintain loss-detection sensitivity, then a flow meter with measurement uncertainties comparable to those for the accountability tank must be developed, or buffer accountability tanks must be added at the 2A feed tank location.

Achieving goal 2 performance levels (8 kg of plutonium abrupt and 40 kg of plutonium protracted) seems reasonable for the HEF chemical separations area. This requires improving in-process inventory measurement

uncertainty to about 1% precision for process tank volume and concentration measurements. It also would require improving accountability and plutonium sample tank transfer measurement uncertainties to about 0.04% relative standard deviation (RSD) volume calibration, about 0.04% RSD volume standards, about 0.1% RSD concentration calibration, and about 0.05% RSD concentration standards.

Attaining goals 3 and 4 requires inventory measurement or estimate errors less than 0.2% RSD, transfer random errors less than 0.03% RSD, and transfer correlated errors less than 0.002% RSD. For comparison, today's primary standards have errors of about 0.04%. Clearly, the proposed international and domestic safeguards goals cannot be achieved without major breakthroughs in measurement technology and standards preparation.

The optimization methodology developed for this study is useful for identifying measurement uncertainty components that dominate materials balance standard deviations and that require development to meet specific performance goals. Further study should be undertaken to determine the sensitivity of the results to the choice of cost functions and constraint ranges for the measurement uncertainties.

#### IV. SAVANNAH RIVER PLANT (SRP)

##### A. JB-Line and New Special Recovery Facility (*H. A. Dayem, K. C. Greenough, and D. P. Martinez, Q-4*)

Support is being provided to SRP in two areas, the existing JB-line process and the New Special Recovery Facility. The existing JB-line effort evaluates materials accounting strategies and, if possible, identifies ID sources and calculates an ID variance. The special recovery effort will develop instrument conceptual design plans and integrated accounting and process control models.

Los Alamos developed a JB-line model (JBMOD) and simulated operation of the JB-line as the first step in evaluating nuclear materials measurement, control, and accounting in the existing JB-line. This model also will be useful in the design and evaluation of proposed JB-line upgrades.

A description of the JB-line process computer model was sent to OSS and SRP for review and comment. SRP is compiling measurement data to evaluate the materials accounting systems performance and formulating a response to the proposed tasks for the special recovery process.

##### B. Sludge Monitor (*N. Ensslin, R. Walton, and R. Siebelist, Q-1*)

Special plutonium recovery operations at the SRP generate filtrate sludge that is stored in a cylindrical

beaker until enough has accumulated to warrant recycling. It is important to monitor the liquid level and plutonium content of this beaker to guarantee that it remains critically safe. SRP has asked Los Alamos to provide such a monitor, which is to be installed below the glovebox containing the beaker.

To provide data for selecting the best measurement technique, we used three assay techniques: total neutron counting, coincident neutron counting, and gamma-ray counting. Total neutron counting is proportional to the  $^{240}\text{Pu}$  content of the sludge and is relatively insensitive to attenuation by intervening materials. However, total neutron counting is affected by room background, neutron poisons in the sludge, and  $(\alpha, n)$  reactions in beryllium, fluorine, or other impurities. Coincident neutron counting is less subject to these effects but is more sensitive to self-multiplication within the sludge. Also, the net coincidence counting rate is very low. Counting of 350- to 414-keV gamma rays is proportional to the  $^{239}\text{Pu}$  content of the sludge and is unaffected by its chemical composition. However, gamma-ray attenuation must be taken into account by careful calculation.

Table IV summarizes measurements carried out by SRP and Los Alamos personnel. The neutron measurements were made with a portable SNAP detector held beneath the glovebox. The gamma-ray measurements were made from below with a heavily shielded and

TABLE IV. Sludge Beaker Measurements through January 12, 1982

Date	Time	Estimated Liquid Level ( <i>l</i> )	Total Neutron Assay (g)	Coincident Neutron Assay (g)	Gamma-Ray Assay (g)
8/18/81	1600	N/A	360	N/A	15
11/13/81	0700	1	---	---	5
11/13/81	1955	1.2	---	---	11
11/15/81	2310	1.2	---	---	17
11/16/81	0840	1.2	---	---	12
11/16/81	2005	1.5	---	---	18
11/17/81	2340	1.8	---	---	19
11/18/81	1200	1.0	---	---	13
11/19/81	2210	1.5	---	---	13
1/11/82	2030	0.5	---	---	57
1/12/82	0845	1	---	---	59
1/12/82	1200	1	---	---	69
1/12/82	2320	1	1775	802 ± 237	76

tightly collimated NaI detector. The neutron measurements, where available, show substantially higher assays than do the gamma-ray measurements. This probably is caused by high ( $\alpha, n$ ) reaction rates and self-multiplication within the sludge. The gamma results probably are more reliable, but this must be confirmed by measuring known solutions now being prepared at SRP.

If assay of the known solutions confirms that gamma-ray measurements can determine the plutonium content

of the beaker, the final shield and collimator will be designed and installed. Present plans call for the use of a light-emitting-diode (LED)-stabilized sodium-iodide detector and electronics package from Ludlum Instruments, Inc. An HP-41CV calculator-printer is available to provide remote control of the assay, perform the necessary calculations, and provide a hard copy of the results to the plant operator.

## V. LOS ALAMOS PLUTONIUM FACILITY

### A. Technical Area (TA)-55 Implementation Support

The Safeguards Systems Group provides technical support to the Los Alamos Plutonium Facility (TA-55) manager and the operational safeguards staff in materials measurement and accounting. This assistance is based on TA-55 operational objectives and the applicability of technical results to other DOE facilities and projects. Current activities include consultation on measurement techniques and measurement control; NDA instrument test, evaluation, and calibration; and in-plant evaluation of a solution mass-measurement system.

An informal report entitled "TA-55 Implementation Support Project Report" describes the support activities during FY 81 and defines the FY 82 activities as of October 1, 1981.

Primary emphasis in this subtask has been on solution mass measurement and low-level plutonium assay.

**1. Solution Mass Measurement (*W. Ford, Q-4; C. Osborn, Q-1; and R. Picard, S-1*).** Determining the plutonium content of a solution in a tank requires a knowledge of the solution plutonium concentration and the total quantity of the solution. These two parameters can be measured in volumetric or gravimetric units. At Los Alamos, we use a gravimetric approach and have developed a solution mass-measurement system based on the pneumatic bubbler-tube method to determine the solution mass in vertical process tanks.

The portable tank calibration system fabrication and testing and evaluation were completed. Temperature calibrations were made using water; density calibrations used ethanol and ethylene glycol. The calibration system has an accuracy of about 5 g using room-temperature

water as the calibration fluid. The change in density of about 0.2 density units carries a change in calibration of about 2%. The results of these studies are presented in a draft report entitled "Solution Mass Measurement."

A system developed to read remotely the output of a pressure transducer or an electronic balance was incorporated in the solution mass-measurement system. All software is EPROM-based and runs on an LSI-11 computer with control through a hand-held terminal. Hard-copy output can be obtained from a small printer.

The problem of pressure transducer damage from over- or under-pressure transients during solution transfer was solved by a pressure transducer having provisions to limit transducer diaphragm movement. Use of a solenoid-operated valve placed between the bubbler tube and the transducer pressure port to prevent solution backflow into the transducer was investigated. Closing the valve allows the full air supply pressure to be applied to the transducer. The valve was cycled 5000 times at full pressure on the transducer without effect.

A device to subject automatically the measurement system to alternate cycles of pressure and vacuum was fabricated and installed on the test-bed tank. This will provide an indication of the long-term effect of over- and under-pressure on the system. To date, about 3500 cycles have been completed with no apparent effect. The test will continue until the system has been cycled 5000 times.

Accuracy of the calibration system is limited, in part, by the inherent accuracy of the Ruska electromanometer used in the system. A lower-range Ruska was ordered to investigate the possibility of improving accuracy.

The measurement system was demonstrated to plant operations, accountability, and safeguards research and development (R&D) personnel. Because of interest expressed by operations personnel, the project was revised

to include an in-plant evaluation. A second system will be fabricated and installed on a tank at TA-55 for calibration, testing, and evaluation. Currently, plant installation is scheduled for late March.

**2. Low-Level Plutonium Assay (S.-T. Hsue, Q-1).** The nondestructive determination of low-level plutonium (1 to 1000 mg/l) in the presence of varying amounts of americium is a problem encountered in plutonium recovery operations. Typically, effluent solutions from the anion-exchange columns used in the recovery of plutonium at TA-55 present this type of problem. These effluents nominally have a low plutonium content (less than 100 mg/l) and a high americium content relative to the plutonium (Am/Pu about 1 to 20). An NDA instrument for assay of such solutions is under test and evaluation at TA-55. The NDA system is based on measuring the L x rays following alpha decay and has a detection limit of about 1 mg/l.

During this report period the data collected previously on the low-filtrate solutions have been analyzed by the GRPANL program developed by R. Gunnink of Lawrence Livermore National Laboratory (LLNL), rather than by the subtraction technique used previously. The results indicate that with the peak-fitting technique the  $L_{\beta}$  and  $L_{\gamma}$  x rays give results in good agreement with each other for the plutonium assay and that the 26- and 59-keV peaks give consistent results for the americium assay. For calibration solutions, the plutonium can be determined to an average deviation of 5% for concentrations ranging from 10 to 200 mg/l of plutonium and Am/Pu ratios to 0.15 (plutonium and americium concentrations known to about 1%). For actual process solutions, the average deviation is about 30%; however, the destructive analytical technique used for comparison is not highly accurate. For solutions with an Am/Pu ratio greater than 0.1, the estimated accuracy of the destructive method is 20%. The peak-fitting technique was evaluated for solutions with Am/Pu ratios up to 4.

The determination of low levels of plutonium in solutions with Am/Pu ratios greater than 4 will require separation of the americium from plutonium. Radiochemical separation in a sample chamber using liquid-liquid extraction techniques is being investigated. A commercially available container, which would be suitable for use as a sample cell, has shown adequate resistance to attack by the organic solutions used in extraction with reagents such as TOPO and TTA. Future efforts will be directed towards combining the radiochemical separation with the NDA measurement of plutonium.

**3. Solution Assay Instrument for TA-55 (T. Marks, T. K. Li, and J. L. Parker, Q-1).** A new plutonium solution assay instrument (SAI), built and installed at TA-55, replaces a similar 3-yr-old system in which the germanium detector performance had deteriorated badly and the electronic hardware had become so unreliable that the whole system required constant maintenance. The new system is an upgrade as well as a replacement for the older system, though the fundamental measurement principles and procedures remain the same.

The new SAI system has two measurement stations attached to a single MCA/computer system. This allows twice the sample throughput and greater convenience, yet requires little more than the space and capital expense of a single unit system plus one additional germanium detector and associated nuclear instrument module (NIM) electronics. In principle, each measurement station can be optimized for a particular type of solution, for example, high americium-to-plutonium ratio or high plutonium concentration. The assay setup and printout of results for both stations occur on the same hard-copy terminal.

To ensure the quality of assay results, the SAI program requires daily and weekly check runs and monitors key aspects of all assay runs. Results will not be printed unless all checks have passed established limits; however, if one station fails its measurement control sequence, operation of the other station is not affected. Limits are established for the resolution and position of key spectral peaks, the ratio of key peak areas, the maximum background counting rate, a standard foil assay, and precision runs.

Because most calibration and measurement control parameters are stored in a disk file, they can be changed conveniently, minimizing the need for reprogramming. To initiate the dialogue necessary for parameter changes, an operator needs to know a password, thus preventing an unauthorized user from tampering with the parameters. However, easy modification makes these parameters subject to the skill and integrity of the privileged operator(s).

Should reprogramming be necessary, the new system is programmed in FORTRAN, allowing straightforward updates and software modifications by an experienced programmer. The system had used a version of BASIC that was probably too easy to modify for an in-plant facility because it is subject to inadvertent changes by untrained operators. The two other existing SAI systems are constrained by programs written in assembly language, which are difficult to modify, thus inhibiting desired changes and updates.

The dialogue called up by an operator to set up an assay is kept as simple as possible. Whenever possible, answers to computer prompts are checked to assure correctness. If an error is detected, the original prompt is repeated and sometimes supplemented by additional instructions. Because this is effective in preventing computer crashes and nonsensical results, it minimizes operator frustration.

It is especially important that the resolution remain good on a system intended to measure americium and plutonium in the same sample. Past experience has indicated that the resolution of the P-type germanium detectors deteriorates noticeably with time, apparently as a result of neutron damage. We believe that newly available (at additional cost) N-type germanium detectors will deteriorate less rapidly (only 5 to 10% as fast) with neutron damage. On the present system, we have installed one of the usual P-type detectors and one of the N-type detectors, thus allowing a comparison under similar conditions.

Currently, both measurement stations are configured to measure any type of solution generated from the plutonium purification and americium recovery processes. For solutions generated from the plutonium purification process, the total (Am + Pu) concentrations range from 0.1 to 500 g/l and the Am/Pu ratios vary from a few parts per million to 10; for solutions generated from the americium recovery process, the total concentrations range from 0.1 to 50 g/l and the Am/Pu ratios vary from 0.5 to 100. After gaining more operating experience, we may decide that expecting each station to assay accurately the full range of solutions is impractical. In that case each station can be optimized to measure different, more limited ranges of solutions by additional gamma-ray filtering and computational changes.

**B. Applied Systems Integration** (R. C. Bearse, D. G. Shirk, W. Ford, R. S. Marshall, J. Sapir, R. M. Tisinger, A. L. Baker, C. C. Thomas, Jr., Q-4; C. E. Nordeen, CMB-11; and F. Kelso, OS-2)

We are defining, testing, and evaluating an integrated safeguards system, comprising materials control and accounting (MC&A), physical protection, plant operations, and process control subsystems, that is both effective and acceptable to facility operators. This integrated safeguards system will be demonstrated with a target date of June 1985. Current effort involves perfect-

ing rapid-inventory capabilities using data from the TA-55 Plutonium Facility/Los Alamos Safeguards System (PF/LASS) to make inventory and related data available in near-real-time in a format usable in an integrated system while assuring the integrity of the accountability data. Activities include development of on-line, near-real-time NDA instrument networks, holdup measurement systems, and a rapid physical inventory and materials balance capability.

**1. On-Line NDA Instrumentation Network.** During this period, we designed an on-line NDA instrumentation network with several functional capabilities: data collection, reduction, and analysis; accountability and decision analysis; on-line process assistance; and on-line graphics.

To function efficiently, the network must be able to initiate transactions from a primitive device. We have designed a smart communications interface unit that can be placed between the preprocessor and the device of interest. This unit will allow data entry by a compact ASCII keyboard, data output by a small thermoprinter, and both on- and off-line operation of the device. Arbitration for the various options will be handled by a software control microprocessor resident in the interface. This interface will allow convenient data entry for both process monitoring and accountability purposes while minimizing operator effort for data entry. The compact printer and terminal will allow placement of the data entry devices at the process work station and will avoid the cost and size of a full-screen CRT or hard-copy terminal.

An electronic balance will be used for demonstration and test and evaluation of the smart communications interface. All components of the system, except the balance, have been received.

**2. Instrument History Activity.** Instrument history data in the PF/LASS data base are generated by a measurement control program. Analysis and evaluation of these data can provide a better understanding of instrument performance and a basis for critical review of the measurement control program.

We completed a preliminary analysis and evaluation of the instrument history data for the electronic balances used in the FFTF process. This preliminary effort was intended to indicate what could and, more importantly, could not be extracted from the data. The effort involved translating and editing the original instrument history

data tapes (from March 1980 through March 1981) obtained from the Los Alamos Nuclear Material Management Accountability and Control Group OS-2. These tapes contain the results of the measurement control tests, such as the daily accuracy checks. Programs were written for the PDP 11/34 for the translation, editing, plotting, and analysis of the data. Although these programs were suitable for the limited preliminary effort, they are not appropriate for use as routine production codes. Plots of the results of the accuracy test (t-statistic) were made and analyzed to correlate the various behavioral patterns and trends with written records. Because of lack of recalibration information in the data base, estimates of the total, systematic, and random error variances were calculated for each of the FFTF-process electronic balances for six recalibration cycles. These calculations confirmed that the single "historical" sigma value used for all balances and test weights in the accuracy test was not rigorous because of variance in the true sigma from balance to balance and its proportionality to the mass of the individual test weights.

Information from the preliminary study led to further effort. Arrangements have been made with the Plutonium Chemistry and Metallurgy Group (CMB-11) personnel to compile the information in the present instrument logs and to provide such information as it is generated. This information will augment the data stored in the instrument history data base. We also are receiving monthly instrument history data tapes from OS-2. The data base, including NDA instruments as well as balance data, was transferred to the Prime computer, and monthly tapes are used to update it. A program to provide hard copy and to enter the data into core memory for later plotting and analysis was written. Programs for data editing, plotting, and analysis are being written. We will evaluate these data to help understand and predict the reliability and error structure of these instruments. We are continuing to supplement the information in the PF/LASS instrument history data through collection of information from instrument logs and from discussions with instrument operators. In addition to providing a better understanding of instrument performance and measurement uncertainties, these efforts will provide a basis for evaluation, and possible upgrading, of the measurement control program at TA-55.

A study of the performance under laboratory conditions of two 5.5-kg electronic balances, used in conjunction with the test bed for the solution mass-measurement project, has been initiated. The study will permit us to

evaluate a measurement control program using three standard weights versus one using two standards (current practice at TA-55), as well as to obtain information on balance reliability and error structure under controlled conditions.

**3. Holdup Measurements.** Data from the plutonium exhaust air filter monitor installed on a glovebox in which  $\text{PuO}_2$  is screened, blended, and packaged were analyzed. There is evidence of nonlinearity in the plutonium buildup on the filter with throughput. The holdup-throughput relationship, based on measured holdup and associated throughputs as of mid-1981, is approximated by the quadratic function

$$\text{Pu}(g) = 0.1930(x) + 0.0008443(x)^2 \quad ,$$

where  $x$  = throughput in kilograms of plutonium.

The holdups predicted by the above equation, which was derived for a total throughput of about 120 kg of  $\text{PuO}_2$ , agree with the measured values within a few percent in the best cases and within 15% in the worst cases.

**4. Computer Activities.** To minimize interference with processing at the reference facility (TA-55) and maximize our ability to investigate the effects of various accounting strategies, process modifications, and alternative integrated systems components, we elected to use modeling and simulation techniques and a computer that is not part of the reference facility system. Our initial work with the PF/LASS data base used a Prime computer. Because of the limited availability of data-base systems for the Prime computer, we have chosen a VAX 11/780 for our integrated systems development. We have transferred all the data prepared for the Prime computer to the VAX. We now have 156 808 transactions on the VAX computer, representing all activity at TA-55 between July 1, 1979, and March 31, 1981. The data occupy about 100 000 blocks of disk space, about 15% of total disk capacity, and about 30% of the space available to VAX users.

We are investigating data-base management packages for the VAX from the standpoint of suitability for use with data bases such as the PF/LASS. Packages being evaluated include DRS, ADABAS-M, DATATRIEVE, and FRAMIS.

The DRS package was available on the Statistics Group's (S-1) computer for testing. Initial tests indicate that DRS is capable of handling our task but at

considerable cost in understanding the nuances of the software.

We attempted to use the VAX FORTRAN keyed read/write option. After several months of data were entered, attempting to add additional records to the file took more than 10 s/record, a rate that negates the use of this technique. Because the pre-1982 version of the DEC-produced DATATRIEVE is designed for analyzing and retrieving data filed with keyed read/write, we have not investigated DATATRIEVE in any detail.

We have arranged to obtain ADABAS-M for evaluation; it is a minicomputer version of the long-standing ADABAS, which runs on IBM computers. ADABAS, in the IBM version, is a world standard and is used by the IAEA for general data-base needs and nuclear accountability. We have prepared two test programs to allow us to begin our analysis of ADABAS when it arrives for a 1-month test (late January 1982).

We obtained from LLNL a copy of their newly developed relational data base, FRAMIS, which was used to rearrange the order of data in a form more suitable for ADABAS and to output these data into files that load into ADABAS. FRAMIS was used to generate an inventory and to generate MIP accounts based on the available data. Although slow, it did work successfully. FRAMIS is a flexible system, but it is too slow for nuclear accountability work where rapid update and interaction are required.

We have investigated a precompiler called Easy Structured Programming (ESP) developed at Los Alamos. This program helps develop FORTRAN code so it is easily changed, transported, and immediately documented. We have adopted ESP as the standard language to prepare further computer programs. Its output is a FORTRAN code that can be understood by any FORTRAN user.

Group OS-2 has developed a complete new set of software, based on the concept of generic transactions, to handle their nuclear accountability system at TA-55. Although this software was developed for the Data General computer, it is highly structured, and examination indicates that the underlying philosophy is well thought out. We will attempt to adapt the OS-2 package for our use.

**5. Voice Systems.** During this report period most effort with voice systems has been directed toward voice verification, although a simple demonstration was designed to illustrate the use of voice to enter commands. Further effort in voice actuation is planned.

Experimental evaluation of talker identification/verification strategies is an involved, time-consuming process. We developed a model for computer simulation of talker verification using many strategies. The inputs to the model are the strategy to be used and a table of phrase-recognition probabilities. Monte Carlo techniques are used to generate the probability of the talker's acceptance. Phrase-recognition probabilities are generated from experimental data using a cross section of actual speakers as users and impostors.

This simulation method allows us to vary the required number of recognition successes, number of recognition failures allowed, phrase-recognition probability, number of vocabulary items, and number of trials. Data are generated for different strategies and different phrase-recognition probabilities showing tradeoffs between impostor success and valid-user rejection. Thus, both strategies and vocabulary selection can be evaluated. Experimental trials were conducted to verify the validity of the simulation. For an arbitrary requirement of three recognitions before four nonrecognitions, experimental results yielded an impostor success rate of 2.8%, whereas the model predicted 2.6%.

A satellite version of the voice-verification system was developed that fits a standard 3.5-in. rack space. The software was converted from disk-based to EPROM-based code. The vocabulary templates reside in a host computer and are down-loaded to the satellite.

The speaker-identify/verification system performed well enough in limited laboratory tests to warrant more extensive testing. An agreement was reached with Allied-General Nuclear Services (AGNS) to allow operation and evaluation of the system in their plant for comparison with their present system. The system implementation will occur in three stages: (1) a stand-alone system, (2) a system controlled by the AGNS Access Control System, and (3) a fully integrated, minimal hardware system.

## VI. LIQUID METAL FAST BREEDER REACTOR (LMFBR) LARGE DEVELOPMENT PLANT

(H. A. Dayem, Q-4)

A final report, "Conceptual Design for Safeguarding the LDP Liquid Metal Fast Breeder Reactor," was issued by Los Alamos National Laboratory and Sandia National Laboratories<sup>6</sup> and included in DOE report CDS200-18. The safeguards systems design combines materials measurement and accounting, containment/surveillance, and physical protection to address both domestic and international concerns. Los Alamos developed the materials measurement and accounting system, and Sandia developed the containment/surveillance and physical protection systems.

The materials measurement and accounting system determines the location and quantity of all nuclear materials in the facility. The entire facility is treated as a single materials balance area with two transfer measurement points (receipts and shipments) and three inventory measurement points (fresh driver fuel storage, ex-vessel storage tank, and reactor core). Fuel assembly transfers are measured using item identification and NDA. Fuel assembly inventories are maintained by item identification of transfers between inventory measurement points and by NDA measurements. The reactor core inventory can be verified by monitoring assembly movements to and from the core and by using reactivity and

control-rod position measurements. NDA and item identification measurements, which are important from both the safeguards and plant operations viewpoints, provide timely information on the location and fissile content of each assembly.

Accounting data from the MMAS will be available to the IAEA. The inspectors will make additional measurements, draw materials balances, analyze the data for consistency, and correlate the fresh fuel data with the fabricator's data and the spent-fuel data with the reprocessors's data. As a result of this analysis and other verification activities, the inspector must reach a technical conclusion, i.e., "a statement, in respect of each materials balance area, of the amount of material unaccounted for over a specific period, giving the limits of accuracy of the amounts stated."<sup>7</sup>

Information from the MMAS combined with the containment/surveillance subsystem provides independently verifiable information to the IAEA while optimizing IAEA resources. Elements of the containment/surveillance subsystem include fuel movement monitors, fuel assembly identification device readers, radiation monitors, pool acoustic monitors, and camera surveillance.

## VII. EXXON NUCLEAR FAST FACILITY

(G. W. Eccleston, S. Johnson, and T. Van Lyssel, Q-1)

A computer-controlled delayed-neutron interrogation (DNI) assay system was designed and fabricated for the Exxon Nuclear Idaho Company (ENICO) Fluorinel and Storage (FAST) Facility. The DNI will nondestructively determine the <sup>235</sup>U content in highly enriched spent fuels and waste solids resulting from fuel dissolution. The instrument is assembled at Los Alamos for measurement tests and completion of software diagnostic, data collection, and analysis programs. Figure 5 shows assembly of the shield components into the interrogator. The assembled DNI and electronic control racks are shown in Fig. 6.

Figure 7 shows a diagram of the DNI control system. The facility design required dual electronics to maintain an operable system following the failure of unrelated components. Operation and control of the interrogator

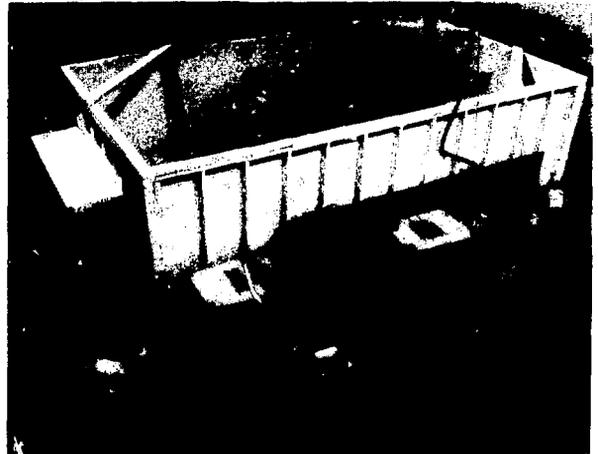
are accomplished with a dual computer system composed of Digital Equipment Corporation (DEC) LSI-11 microcomputers connected to mass storage disks. All signals to and from the computers pass through a computer select chassis. All input signals are routed directly to both computers. Only one computer system is required for operation, and the backup is normally turned off. Output signal lines from both computers are connected to a manual switch. Selection of a computer to control and operate the system is accomplished by setting the manual switch to the appropriate system to connect the computer output lines to the interrogator.

The dual computer system is approximately 100 m from the interrogator cubicle in the operations room adjacent to the main facility computer. Signals between the interrogator and operational computer pass through



Fig. 5. Assembly of shield components into the FAST interrogator.

Fig. 6. Assembled interrogator and electronics equipment racks.



a signal routing chassis in a rack next to the interrogator cubicle. This chassis contains the  $^{252}\text{Cf}$  source control and display hardware, multiplexers to select waste tube or fuel tube signals, diagnostic control hardware, and signal conditioning electronics.

Movement of the  $^{252}\text{Cf}$  source is accomplished with a closed-loop feedback control system (Fig. 8). The source is attached to a Teleflex cable and connected to a stepping motor through a Teleflex gear wheel. Pulses

generated under software control step the motor. An optical encoder attached to the motor provides the feedback signal to a software program, resulting in the fastest possible motor speeds. Source transfer times of 0.5 s over a transfer distance of 180 cm are reliably obtained.

A key-operated safety interlock and cable position sensor logic were incorporated in the source transfer design. The sensors are connected to motor drive safety

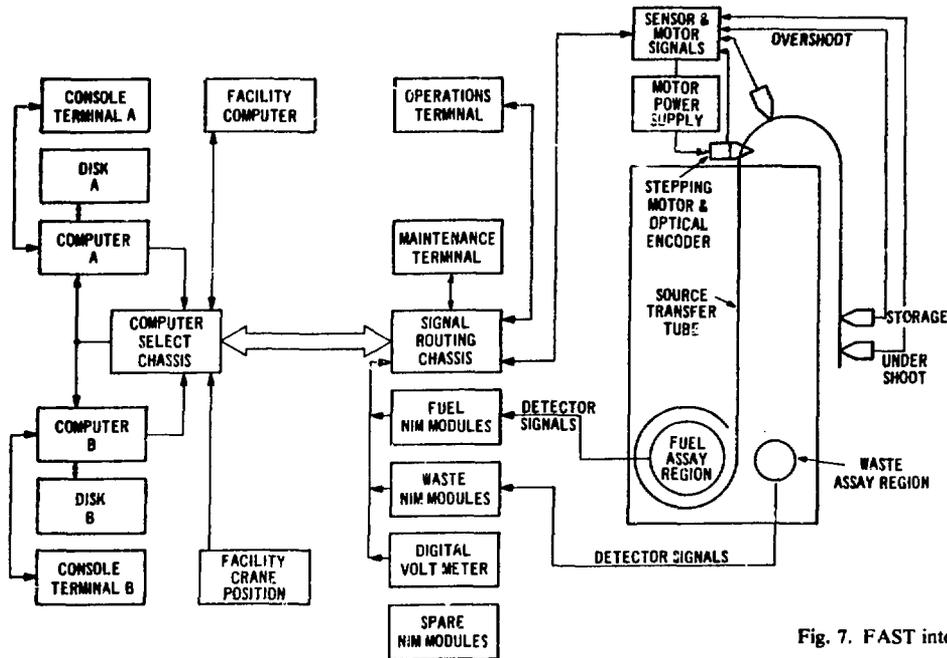


Fig. 7. FAST interrogator control block diagram.

logic, which prevents pulses being fed to the stepping motor if the cable is driven too far in either direction. A zero sensor is also used to reposition the source accurately in its storage location and provide a reference for transfers. When the safety interlock switch is thrown, an interrupt signal to the computer is generated and the software moves the source to storage where it is held in position by a hardware interlock. Further movement of the source is disabled until the interlock switch is moved manually to the enable position.

Neutron events from the waste or fuel regions are processed through six NIM analog channels, converted

to digital TTL-level pulses, routed to a multiplexer, and then passed to six-channel scaler interfaces connected to the backplane of each computer. Fuel and waste region neutron signals are processed in separate NIM units, and a multiplexer, under software control, selects either fuel or waste signals for measurement.

Diagnostic equipment is an integral part of the FAST electronics system. Power supply and detector high-voltage levels can be read with a digital voltmeter. In addition, power supply levels are continuously monitored by electronics in the signal routing chassis. Significant drift or failure of a supply will generate a computer

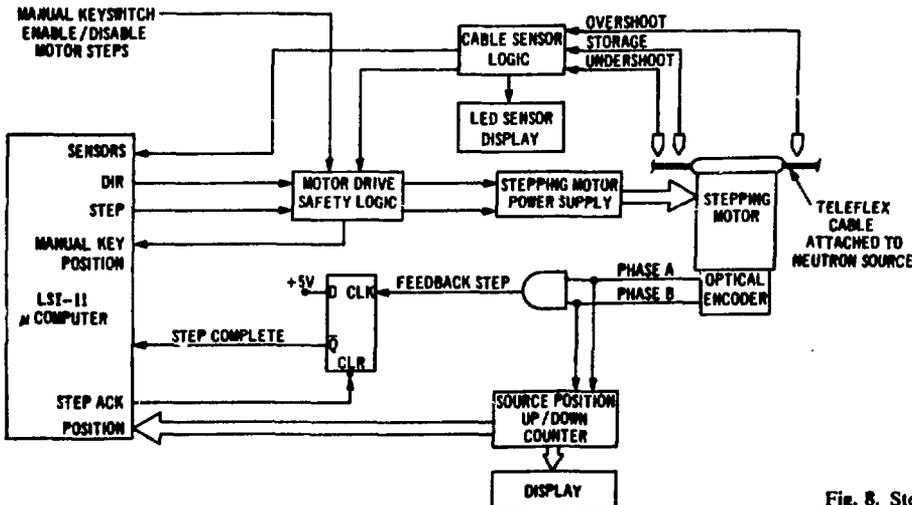


Fig. 8. Stepping motor control block diagram.

interrupt, alerting personnel to the problem. A computer-controlled programmable pulser connected to all detector preamplifier test inputs permits monitoring all detector channels for proper operation. Diagnostic programs were written to test system components, such as the source drive and sensor operation, and locate problem areas when reference measurements do not agree.

Operation of the DNI is achieved by a hard-copy operations terminal in the crane corridor above the

interrogator cubicle. This location allows the operator visual access for positioning and moving samples to and from the interrogator tubes.

Two californium sources from Savannah River Laboratory (SRL) in the FAST shield cask were calibrated by the NBS. The neutron emission rates were  $1.76 \times 10^9 \pm 1.8\%$  n/s and  $2.79 \times 10^7 \pm 1.5\%$  n/s for the large and small sources. These sources will be used for the measurement tests on the DNI.

### VIII. UNITED NUCLEAR CORPORATION (UNC)

(J. K. Sprinkle, Jr., J. E. Stewart, K. E. Kroncke, M. Hykel, J. Leavitt, and C. O. Shonrock, Q-1)

The extrusion process used by UNC to fabricate fuel rods produces nonuniform uranium loading at the ends of the extrusion. Sections containing the nonuniformities are cropped and collected for return shipment to the uranium supplier. At present, the special nuclear material (SNM) accounting values for this material are estimated with a by-difference method. We designed an instrument to assay the uranium contained in these end crops by measuring passive neutrons emitted from  $^{238}\text{U}$  spontaneous fission. The large throughput requirements were a primary design constraint. Consequently, the feasibility of assaying full shipping crates was evaluated and determined to be adequate. This feature increases confidence in the assay because the crates can be secured before the assay to prevent material removal. The counter design and placement within the facility have been strongly dependent on the material-handling aspects of the large, heavy shipping crates. Provision of an exclusion area around the counter will help minimize the background in the counter. Preliminary measurements with a box of defective fuel have indicated that the design objectives of 10% accuracy and 2% precision can be achieved for loadings larger than 45.4 kg of uranium in a 1000-s assay.

Several measurements were made at Los Alamos with a modified  $4\pi$  barrel counter<sup>9</sup> that was modified and used as a test bed for investigating fuel loading effects with and without a cadmium liner. Figure 9 shows the net neutron emission per rod as a function of the number of

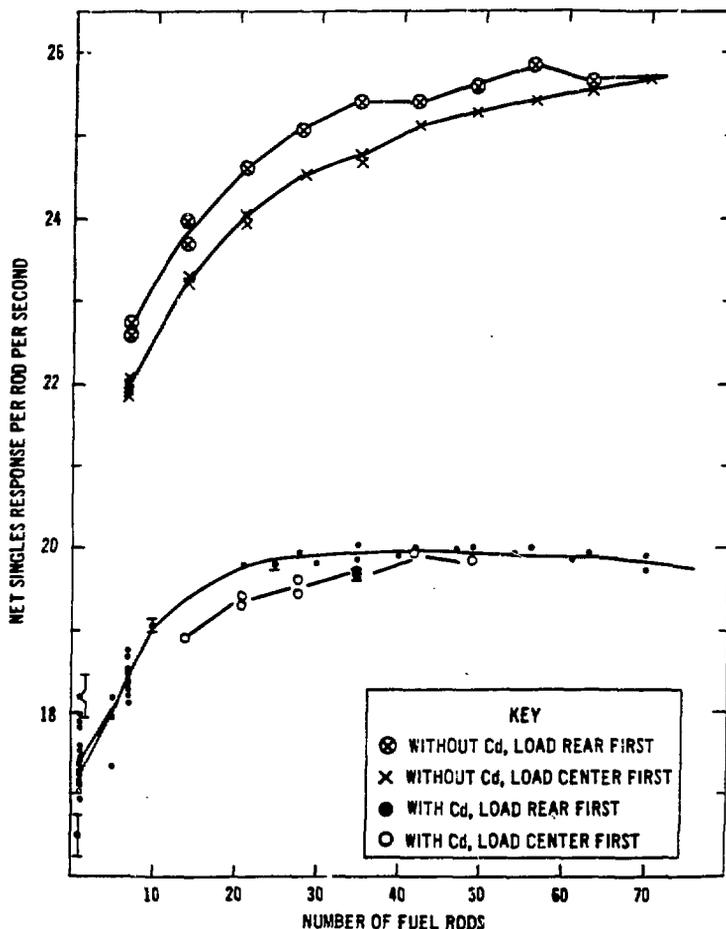
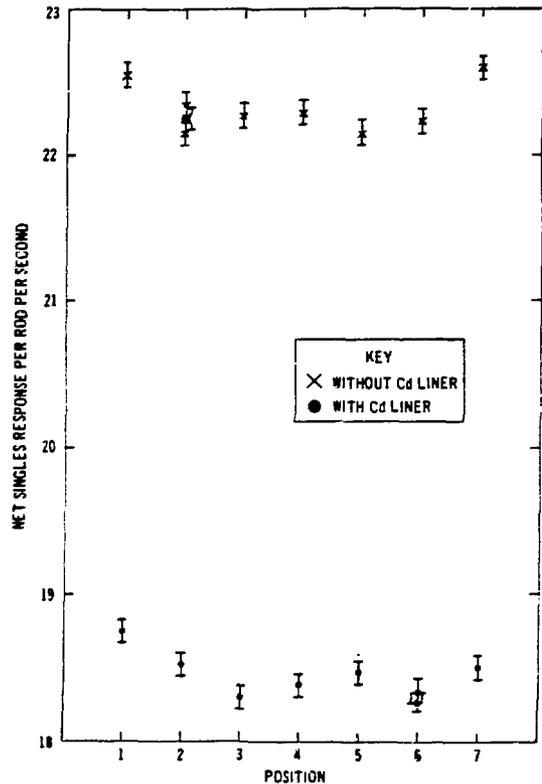


Fig. 9. The  $4\pi$  counter response as a function of increasing mass. The upper (lower) lines depict the response without (with) an inner cadmium liner. This figure also shows the variation in response caused by loading the center or the edge of the counter first.

rods in the barrel counter. Each rod contains 7.5 kg of uranium metal, enriched to 0.95%. The addition of a cadmium liner on the inside of the counter decreased the count rate but flattened the response. This figure also demonstrates a slight variation in the counter response, depending on the uranium's position. The maximum variation was 10% for 1 rod; however, the variation was 2% for 14 or more rods (more than 105.2 kg).

Figure 10 shows the variation in the counter response as a row of seven rods was moved from the front to the rear of the counter. The maximum variation was 2%. Cited results using the barrel counter have been used for design of the actual end-crop box counter. The design has been completed and construction will begin soon. According to our schedule, the counter will be delivered to UNC by August 1982.

Fig. 10. The  $4\pi$  counter response as a function of position. A row of seven fuel rods was moved from the front to the rear of the counter.



## IX. SAFEGUARDS TECHNOLOGY TRAINING PROGRAM

(H. A. Smith, N. Ensslin, and T. D. Reilly, Q-1)

The Safeguards Technology Training School, "Fundamentals of Nondestructive Assay of Fissionable Material with Portable Instrumentation," was held October 5-9. A total of 32 students attended, including 6 foreign visitors. NDA techniques using gamma-ray and neutron-based instruments were covered in four separate laboratory sessions with eight instructors. Lectures also were given on basic neutron detection methods and on the fundamentals of gamma-ray physics.

During December 7-11, 26 students (including 7 foreign visitors) attended the course, "Gamma-Ray Spectroscopy for Nuclear Material Accountability." Details of computer-based NDA techniques and instrumentation using high-resolution gamma-ray spectroscopy were covered in three laboratory experiments, with instruction from six Los Alamos safeguards staff members. In addition, lectures were given on general gamma-

ray spectroscopy and on three in-plant applications of the NDA techniques studied in the laboratory exercises.

The Basic NDA Inspector Training Course was presented twice during this period to IAEA safeguards inspectors. The course was presented at Los Alamos, August 26-September 3, 1981, and January 18-28, 1982, with 12 IAEA participants attending each session. They spent 7 days in the NDA training laboratories at Los Alamos; several inspectors came early to the later session for additional introductory training. The participants, from 15 countries and 6 continents, represented each of the 6 inspection sections of the IAEA. This course provides instruction in the use of NDA instruments, many developed at Los Alamos, available at the IAEA. The participants, working in pairs with a Los Alamos instructor, study instruments such as the high-level neutron coincidence counter, the SAM-2, and the

Silena MCA. They also use the new portable MCA developed at Los Alamos for the IAEA and measure the many nuclear materials available at Los Alamos. New experiments were added to the sessions involving the measurement of plutonium isotopic composition and the

measurement of irradiated MTR fuel elements at the Omega West research reactor.

This course is offered four times a year; future sessions will be scheduled at the request of the IAEA.

## PART 2. SECURITY DEVELOPMENT AND SUPPORT

### I. INFORMATION SECURITY

#### A. Weapons Life-Cycle Study (*R. J. Dietz, NSP/DST; E. K. Tucker and J. L. Merson, IT-3*)

Results of the Pre-Phase-III Weapons Life-Cycle Vulnerability Study were presented to key DOE personnel. Consequently, a sensitivity analysis of the results of that study and the counterpart study at LLNL were performed. The results of the two Pre-Phase-III studies and the sensitivity analyses of them have been compiled in a single classified report that is now in final draft.

#### B. Freedom of Information Act (*A. T. Peaslee, S-4; E. K. Tucker and J. L. Merson, IT-3*)

A study of the Freedom of Information Act was well under way when it had to be terminated at OSS direction

because of a reprogramming of funds. No further activity in this area is foreseen.

#### C. Information Security Assessment (*E. K. Tucker and J. L. Merson, IT-3*)

We are fortunate that an unusually well qualified technician will join the project during February. A first draft of the Information Security Model that is near completion will undergo continued refinement during the year.

### II. COMPUTER SECURITY

#### A. Computer Security Center (*E. A. Springer, OS-4*)

The purpose of the DOE Center for Computer Security at Los Alamos is to evaluate, develop, implement, and demonstrate improved computer security techniques and hardware throughout the DOE. The Center formulates the overall program plan based on policy and guidance provided by the Division of Security, OSS. The Division of Security retains overall program management responsibility and authority for the budgeting and distribution of resources.

From August 1981 through January 1982, the Center completed its organization and staffing, obtained necessary equipment and facilities to conduct research

and development, and contacted appropriate personnel within the DOE community and other Federal agencies.

The second issue of the Center's quarterly newsletter was published and mailed, and a third issue will be distributed to approximately 150 DOE and DOE-contractor personnel at the fourth conference of the Computer Security Group, February 9-11 in Albuquerque. This conference is sponsored by the DOE Center for Computer Security at the Los Alamos National Laboratory. A summary of the conference minutes, in addition to the third newsletter, will be mailed to more than 300 people who have asked to be placed on the Center's mailing list. Center personnel responded to various technical questions concerning computer security, some

of which require research by Center personnel, who will respond as quickly as possible.

#### **B. Key Notarization System (KNS) (D. P. Brenner, OS-4)**

The objective of KNS is to provide a secure means to protect sensitive computer files and to provide for user authentication and secure communications. Demonstration systems of the KNS have been installed at the DOE/OSS headquarters and in the Computer and Telecommunications Group office at Los Alamos. KNS programs for the RSX-11 operating system are nearing completion, and a demonstration system will be installed at Oak Ridge in the near future. The KNS will be demonstrated at the fourth conference of the Computer Security Group. The Center will continue to develop stand-alone, transportable KNS devices for application at DOE facilities.

#### **C. Network Security Controller (D. Nessett, LLNL)**

A security controller for distributed processing is being developed to provide security for computer network resource sharing. A long-term goal is to develop computer security technology that will allow the interconnection of DOE computer equipment at several sites. This technology eventually may be applied to a DOE-wide computer and communications network. A capability-based management system is being designed and tested on the OCTOPUS system at LLNL, and a test bed for the high-speed DES chipset has been completed. The M-68000 system, a prototype of a hardware cryptographic controller, is operational. A cross-assembler for the M-68000, which originally ran on an IBM 370, was modified to run on a VAX. We are investigating new problems in the security of distributed operating systems, specifically how various threats against the security of interprocess communication can be thwarted. Research has been conducted on how network physical security characteristics admit various threats against interprocess communication. This work, which will be used in solving

the major question of interprocess communications security, will be reported at the fourth conference of the Computer Security Group.

#### **D. Management Guidelines for Risk Analysis (G. Corynen, LLNL)**

The principal objective of the Risk Analysis Project is to develop a tool with which the security risks associated with DOE automatic data processing (ADP) sites can be assessed and managed in a cost-effective manner. Risk analysis has been divided into two phases for ease of handling: risk assessment and risk management, with risk assessment being addressed in the past 6 months. Because a further breakdown seemed necessary, risk assessment was divided into deterministic and probabilistic phases. The overall logical structure for assessing the deterministic risk aspects of a facility is well under way. The probabilistic overlay will be completed during the next phase. This approach breaks a security problem into clearly identified parts that can be assessed separately then recombined by precisely defined rules, using such tools as graphs and logic structures. In the deterministic phase, risk assessment is based on a highly structured questionnaire to be completed by ADP personnel untrained in risk analysis. Their answers then are translated into a relational data base in the computer as a graph model of the facility. As a manual for field use, the questionnaire typically will consist of 25 forms for large facilities. Each form allows the detailed assessment of a specific part of any facility. We also have discovered that this risk analysis methodology is directly applicable to the analysis of computer security problems.

#### **E. Secure Operating Systems (H. W. Egdorf, OS-4)**

The Center is in the process of obtaining a Honeywell Level 6 Secure Communications Processor (SCOMP) for testing and evaluation. SCOMP is designed to serve as a front-end machine for multilevel operating systems that concurrently process various levels of classified information.

## PART 3. SAFEGUARDS TECHNOLOGY DEVELOPMENT

### I. CHEMICAL AND ISOTOPIC ANALYSIS (*J. E. Rein and Staff, CMB-1*)

Safeguarding uranium, plutonium, and thorium requires measuring the amounts and isotopic distributions of these elements to provide inventory control throughout the nuclear fuel cycle. Rapid chemical assay and isotopic methods that provide accurate and precise measurements are needed for a variety of nuclear materials, including pure products, reactor fuels having complex chemical compositions, and many types of scrap and waste materials.

Overall program objectives are to (1) develop fast dissolution techniques and analytical methods for determining plutonium and uranium, with emphasis on scrap-type and difficult-to-dissolve materials; (2) design and construct automated apparatus for determining plutonium and uranium; (3) prepare well-characterized, plutonium-containing reference materials for use in the Safeguards Analytical Laboratory Evaluation (SALE) program, for distribution by the NBS and the New Brunswick Laboratory (NBL), and for calibration of NDA equipment at Los Alamos; (4) participate in an inter-DOE laboratory program for measuring half-lives of the longer lived plutonium isotopes; (5) evaluate mass-spectrometric techniques for IAEA verification of plutonium and uranium contents of reprocessing plant dissolver solutions; and (6) characterize chemically special lots of nuclear materials as required by DOE.

#### A. Development of Assay Methods

1. **Microgram-Sensitive Spectrophotometric Determination of Plutonium** (*N. M. Saponara and S. F. Marsh, CMB-1*). We completed development of the extraction-spectrophotometric method for determining 2.5 to 17.5  $\mu\text{g}$  of plutonium with a RSD of 1.5%. This method involves extraction of the Pu(IV)-chlorophosphonazo III complex from 1.5M HCl into n-pentanol and spectrophotometric measurement of this intensely colored complex. Many metals and nonmetals in fuel cycle materials do not interfere, and a preceding anion-exchange-resin-column separation increases the specificity even more. A Los Alamos report has been prepared.

2. **Controlled-Potential Coulometric Determination of Uranium** (*N. M. Saponara and D. D. Jackson, CMB-1*). Development has been initiated of a controlled-potential coulometric method for determining uranium. Objectives are a precision of less than 0.2% RSD, measurement of low-milligram amounts of uranium, high specificity to tolerate impurities in nuclear fuel cycle materials, and application to the Los Alamos automated controlled-potential coulometer developed previously for determining plutonium.<sup>9</sup>

The working electrode used almost exclusively for the controlled-potential coulometric determination of uranium is a pool of mercury because it provides adequate hydrogen overvoltage for measuring the U(VI)-U(IV) couple without decomposition of water. Such an electrode is difficult to use in an automated analyzer. A review of the literature indicates that a solid electrode system characterized by operational simplicity is not available. Systems that have been used include (1) prerduction of both uranium and plutonium with Ti(III) to U(IV) and Pu(III), then oxidation of U(IV) to U(VI) by generated Pu(IV) at a gold electrode;<sup>10</sup> (2) reduction of U(VI) to U(IV) by generated Ti(III) at a platinum electrode with a reduction catalyst present;<sup>11</sup> (3) reduction of U(VI) to U(IV) at a silver electrode, which was not quantitative;<sup>12</sup> and (4) carbonaceous electrodes of glassy carbon,<sup>13</sup> boron carbide,<sup>14</sup> and ke-F graphite.<sup>15</sup> These carbonaceous electrodes have not provided the reproducible, large surface areas necessary for precise controlled-potential coulometric determination of milligram amounts of uranium.

Selected materials as electrodes in combination with complexants selective for U(IV), to enable use of a more positive potential for the reduction of U(VI) to U(IV), are being investigated.

3. **Complexometric Titration of Uranium** (*D. D. Jackson and R. M. Hollen, CMB-1*). The previously developed method<sup>16</sup> for determining microgram amounts of uranium, based on titration of the U(VI)-arsenazo complex with pyridine-2, 6-dicarboxylic acid to colorimetric end point, has been improved for use with the automated analyzer. In the original method, a combined

buffer and interference-element-masking solution was added to the sample, and pH was adjusted to 4.9 by adding acid or base before titration. By omitting nitric acid from the buffer-masking solution and increasing its hexamethylenetetramine content, a pH adjustment no longer is necessary. This modification increases acidity tolerance to 16 milliequivalents, which, for a 1-ml maximum sample volume, provides wide applicability to nuclear fuel cycle materials.

## B. Development of Automated Analyzers

1. Automated Controlled-Potential-Coulometric Plutonium Analyzer for SRP (*D. D. Jackson, R. M. Hollen, and F. H. Kelley, CMB-1*). Construction of this instrument and its testing are expected to be complete on schedule by the end of February 1982. All mechanical portions, including the sample transport mechanism, reagent dispensers, and the cell and electrode cleaning system, have tested satisfactorily. The control and read-out system, consisting of a Hewlett-Packard HP-85 programmable calculator with a built-in printer-plotter and various interfaces, is assembled and testing has started. The last major task of developing instrument control programs is under way.

2. Automated Complexometric Uranium Titrator (*D. D. Jackson and R. M. Hollen, CMB-1*). General design of this instrument, to use the highly selective method described previously for determining microgram amounts of uranium, has been established. The mechanical portion of the instrument is being constructed, and the control system, centered on a Hewlett-Packard HP-85 programmable calculator, is being designed.

Basic features are a probe colorimeter to measure changing absorbance during titration, a turntable transport system with a capacity of 36 sample cells, a positive-displacement dispenser to deliver the combined buffer and impurity-element-masking solution, an automated buret to deliver the titrant, and the system to control all mechanical operations, process titration information, and provide a printout of results.

Laboratory experiments have established the cell dimensions; the means of solution stirring; the means of cleaning the cell, stirrer, and colorimeter probe; and the dimensions of the tip of the titrant delivery buret. The combination of cell size and solution volume is an

experimentally established compromise that provides complete immersion of the colorimeter probe, effective stirring, small-volume change during titration, adequate absorbance, and low total volume for waste recovery. Solution stirring is attained with a glass paddle at 1800 rpm, and its location relative to the colorimeter probe is critical. The established position provides rapid exchange of the bulk solution with the cavity in the colorimeter probe without gas-bubble formation. The rinse-cleaning of cell, stirrer, and colorimeter probe is attained by an aspiration system similar to that used in the automated controlled-potential coulometer.<sup>9</sup> To attain reproducible titrant delivery, the buret tip is immersed in the solution. Use of a small-diameter tip gives no significant carryover between samples and no significant diffusion of bulk solution into the tip during a titration.

## C. Development of Dissolution Technology (*S. F. Marsh, CMB-1*)

Nuclear fuel cycle materials, especially incinerated scrap materials, that have been subjected to acid leachings are highly resistant to dissolution. Their dissolution is necessary before chemical analysis to attain accurate uranium and plutonium contents. New dissolution techniques are being explored using calcined PuO<sub>2</sub> as the test refractory material.

The Los Alamos sealed-reflux dissolution apparatus<sup>17</sup> is used for many dissolution-resistant materials. A usual dissolvent is an acid mixture of 12M HCl, 0.25M HNO<sub>3</sub>, HF at an upper temperature limit of 150°C, restricted by pressure buildup. Use of acid mixtures with HBr, which is a more effective dissolvent for PuO<sub>2</sub> than HCl and has a lower vapor pressure, is being investigated. A mixture of 7.2M HBr, 2.4M HCl, and 0.06M HF contained at 190°C gave improved dissolution of PuO<sub>2</sub>.

Hydriodic acid is an even more effective dissolvent than is HBr for PuO<sub>2</sub>. Concentrated HI is only 7.4M. An attempt to prepare a higher concentration acid dissolvent by bubbling hydrogen iodide gas into 9M HBr was not successful because of the low solubility of HI.

Fusions with the iodide salts LiI, KI, and NaI having melting points of 446, 582, and 651°C will be evaluated. Unsuccessfully tried were fusions with high-melting-point organic acids of oxalic, tartaric, malonic, dichloroacetic, and trichloroacetic acids.

## II. STANDARDS DEVELOPMENT

### A. Reference Materials (*J. E. Rein and Staff, CMB-1*)

1. **Plutonium and Americium Reference Solutions** (*J. W. Dahlby and T. R. Hahn, CMB-1*). Four sets of reference solutions were prepared for use in calibrating various nondestructive analyzers. These sets included six solutions having 160 to 375 g/l of plutonium, six solutions having 98 to 200 g/l of plutonium, one solution with 124 mg/l of plutonium, and one 5-mg/l americium solution. Each reference solution was characterized for actinide content, isotopic composition, and volume as requested, and the solutions were placed or sealed in special containers when furnished.

2. **Uranium-Graphite Reference Powder Mixture** (*J. J. Miglio, CMB-1*). A reference material consisting of 59 g of  $U_3O_8$  powder and 845 g of graphite was prepared, characterized, and packaged for use in calibrating a nondestructive analyzer.

3. **Plutonium Spectrographic Matrix Reference** (*J. W. Dahlby and L. Duval, CMB-1*). Fourteen units of the NBS SRM 945 were packaged and shipped to NBL at the request of NBS. These 5-g sealed reference samples have been kept in storage at the request of the NBS.

### B. Review of NDA Regulatory Guides (*H. A. Smith, Q-1*)

Final reviews and revisions of the eight NRC Regulatory Guides on NDA<sup>18</sup> were completed in September 1981. These reviews have gone out for NRC comment,

and all are expected to require some refinement. Comments have been received on more than half of the guides, and second revisions have been made. The remaining comments should be received over the next few months. No new work in this area is anticipated.

### C. Preparation of HEU Standards for NRC Inspectors (*H. Smith, J. Parker, J. Sprinkle, and T. D. Reilly, Q-1*)

For a newly initiated contract with the NRC for FY 82, a set of NDA standards has been prepared for use by NRC inspectors to confirm the calibration and accuracy of licensee NDA measurements. The standards consist of six 2-l polypropylene bottles of HEU oxide mixed with graphite flour. The bottles were produced by the Los Alamos Materials Technology Group (CMB-6). The standard material was analyzed, and the standards were assembled and weighed by the Los Alamos Analytical and Instrumental Chemistry Group (CMB-1). The six bottles contain 0, 10, 20, 50, 100, and 200 g of HEU, and preliminary measurements will begin soon on the 50-g sample. Complete measurements on all six standards are contemplated to document fully their character, behavior, and measurability. Additional sets of standards are required by the contract over the 12-month period following the arrival of contract funds. The nature of the additional sets of standards will depend on the needs of the NRC inspectorate, as expressed by the response of the regional inspection offices to a survey now under way. Formal NUREG reports will be generated, which detail the fabrication procedures and testing results on all standards produced under this contract.

## III. NDA DEVELOPMENT AND SUPPORT

### A. Vehicle SNM Monitor (*P. E. Fehlau, J. M. Bieri, J. D. Atencio, and K. V. Nixon, Q-2*)

Past reports<sup>19-25</sup> have described an investigation of vehicle SNM monitoring that included designs with radiation detectors placed in the roadbed beneath a vehicle and designs with radiation detectors placed beside a vehicle to form a vehicle portal. The roadbed monitor has its detectors well distributed under the most

common size vehicles; because little change occurs in the detector background when the monitor is occupied, long monitoring periods, a minute or so, can be used to achieve good detection sensitivity for small quantities of SNM. On the other hand, tall or long vehicles have reduced sensitivity at remote points far from the roadbed detectors. Where sensitivity for small quantities of SNM is not required (for instance, in safeguarding entire nuclear weapons or entire weapons subassemblies),



**Fig. 11.** Vehicle portals have gamma-ray detectors in columns that form the portal. This Ford van is one of the vehicles used in studying vehicle portal performance.

small, long, or tall vehicles can be adequately monitored by slowly driving the vehicle through a vehicle portal.

Vehicle portals like the one in Fig. 11 are much different from a roadbed monitor because the gamma-radiation detectors face the ground rather than the sky. Because the ground is the origin of most gamma-radiation background, a vehicle in the monitor shields the detectors from some of the background and reduces the detector background count rate. This is important because monitoring uses a comparison between the number of detected gamma rays during passage and the number of gamma rays detected in an equal time interval of no occupation. An alarm takes place only when the

occupied count exceeds the unoccupied by a significant amount; thus, the amount of SNM that can be detected includes some quantity that simply supplies the background deficit caused by occupancy. A simple compensation for the deficit is not possible because the amount of background reduction depends on the size and construction of the vehicle. Table V has examples of the observed background reduction for several vehicles in the Fig. 11 portal with the detector columns spaced at two distances. For comparison, most vehicles reduce the roadbed monitor background to about 0.970 of the unoccupied background. One part of applying vehicle SNM portals to meet specific implementation goals requires studying how the vehicle background attenuation varies with different types of gamma-ray detectors and with different detector spacing.

The different detectors shown in Fig. 12 are the most promising candidates from a preliminary evaluation of solid organic (plastic), NaI(Tl), and bismuth germanate gamma-ray scintillators. The vehicle portals shown include the wide plastic scintillator portal that appeared in Fig. 11, a taller portal using narrow plastic scintillators, and a tall portal with two large (12-cm-diam by 3.8-cm-thick) NaI(Tl) scintillators on each side. These portals have 4.9 m between sides; when the occupied background reduction and the detected SNM signal, which decrease with separation, are combined, this 4.9-m separation gives the best SNM detection performance. The portals are used with different vehicles and different forms of SNM to make both stationary and stepped scanning measurements. Figure 13 is a plot of scanning measurements made with the secure SNM transport vehicle shown in Fig. 12 when it was empty and when it contained a large surface area sample of enriched uranium. Arrows indicate the unoccupied background count rate in the tall plastic scintillator portal. The lower curve demonstrates background reduction when the

**TABLE V. Vehicle SNM Portal Monitor Background Reduction at Two Detector Spacings**

Motor Vehicle Type	Occupied Background Fraction of Unoccupied at 3.7-m Spacing	Occupied Background Fraction of Unoccupied at 4.9-m Spacing
Datsun pickup truck	0.936	---
Chevrolet pickup truck	0.900	---
International Travelall	0.861	0.904
Ford van (in Fig. 11)	0.840	0.900
Step van	---	0.896
Secure SNM transport vehicle	---	0.783



Fig. 12. Three vehicle portals with different scintillators are positioned together for simultaneous performance measurements. The secure SNM transport vehicle is typical of the long, tall vehicles that require vehicle portal columns to be about 3.3 m high.

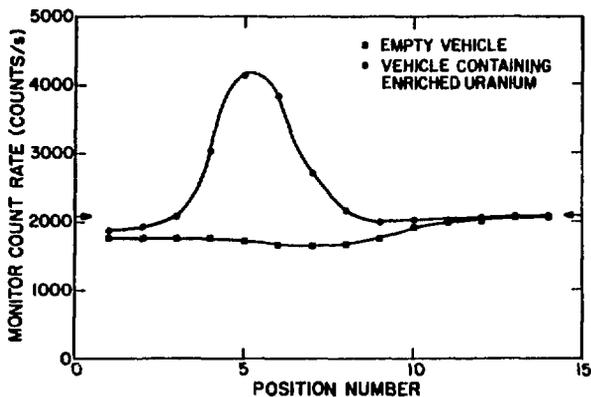


Fig. 13. An automated data-collection system records monitor count rates at numbered positions 91 cm apart. The empty secure transport vehicle suppresses the background, making it harder to detect SNM.

vehicle moves through that monitor, stopping at numbered positions 91 cm apart. The upper curve represents the same measurement with the SNM inside the vehicle. These data are simultaneously accumulated for each portal in several gamma-ray-energy intervals for later analysis. A computer-controlled multiple scaler records the data, which are keyed manually to the numbered position of the vehicle in the portal array.

Analysis of the measurements determines the relative performance of the different portals and the best way to use them. For example, the data plotted in Fig. 13 show a background deficit of about 21% when occupied compared to about 3% for a roadbed monitor. With a 1-

s counting interval and a 4-standard-deviation alarm criterion, an additional count rate of 9% of background for Fig. 13 and an additional 13% of background for the roadbed monitor need to be provided to cause an alarm. By extending the count time to 1 min, the relative size of the statistical contribution is decreased to less than 2%, reducing the needed signal from 30 to 23% for this vehicle portal and from 16 to 5% for the roadbed monitor. Thus, compared to the large improvement in the roadbed result where one-third as much SNM signal is needed to alarm, there is only a fractional improvement in the vehicle portal. In fact, long counting times add little sensitivity compared to sensitivity gained from slow motion through the portal that brings the SNM closer to the detectors.

The scanning results in Fig. 13 also allow comparison of different count intervals and scanning speeds. Table VI illustrates two scanning speeds that might be practical and gives the detectable amount of material for the optimum time interval. There is a slight improvement at the lower speed. Such analyses can provide information on practical parameters for alarm logic used in the monitor control module and can establish the speed intervals over which the desired sensitivity is obtained.

At this time, we have a goal to provide suitable vehicle monitors for the Nevada Test Site (NTS). In addition to the measurements that will be needed to verify adequate performance for a chosen vehicle portal design, we must also establish such basic information as the background radiation intensity, where the monitor will be used, and the material being safeguarded. The last question is particularly important because the SNM at NTS is fabricated by and usually in the custody of NTS users rather than host personnel. Thus, detailed information on size, shape, and material type that may pass through a vehicle portal requires a survey of engineers responsible for past nuclear test shots and designers of future test shots.

TABLE VI. Detectable Amount of SNM at Two Passage Speeds

Passage Speed (km/h)	Counting		Detection Threshold Fraction of Test Source
	Time Interval (s)	Distance Covered (m)	
3.2	1	0.91	0.25
8	2	4.6	0.29

Preliminary surveys of background intensity at NTS and past test nuclear material lead us to believe that the vehicle SNM portal will satisfy the NTS security requirement. Now we must more completely characterize the operating environment and establish good models for future protected devices and components as part of our monitor development program.

**B. Compact K-Edge Densitometer (L. Cowder, P. Russo, and C. Hatcher, Q-1)**

Los Alamos development of K-edge densitometry for measuring plutonium in solution has been along the lines of automated systems. These systems function either in-line, such as the SRP process line system<sup>26</sup> currently being installed, or at-line, such as the Tokai-mura, Japan, reprocessing plant system<sup>27</sup> that was installed as an appendage to a glovebox. Recent development<sup>28</sup> at Los Alamos of a portable battery-powered MCA opens possibilities for a compact densitometer that need not be permanent. Responding to the expressed need for a more compact, less expensive instrument, we have begun development of a K-edge densitometer that will use the portable MCA and a hand-held high-purity germanium detector.

Design objectives include measurement precision and accuracy that are comparable to the larger systems.

As shown in Fig. 14, the compact densitometer can be applied to authentication of liquid samples in a glovebox.

using the identical sample vials used in the larger systems without removing the plutonium sample from the glovebox. This is accomplished by inserting the compact densitometer into one of the glove ports. A disposable clamp, kept in the glovebox, provides alignment of the sample to assure the accuracy required for reproducibility of the sample dimension.

Following measurement procedures established by the automated systems, the compact densitometer will use a software program stored in the MCA that prompts the user through the measurement. Movement of sources is accomplished by manual operation of a three-position lever. The sources are to be size-compatible with previous Los Alamos systems to provide interchangeability should the need arise.

The compact densitometer is in the design stage. Experiments with plutonium samples using the prototype, complete with MCA and software, are expected during the summer of 1982.

**C. Plutonium Isotopic Measurements for Small Product Samples (T. K. Li and T. E. Sampson, Q-1)**

We have initiated a study for measuring plutonium isotopic ratios in product materials using a nondestructive gamma-ray technique. The product materials, impacted on a foil, are expected to be in the range of 100 to 1000  $\mu\text{g}$  of plutonium.

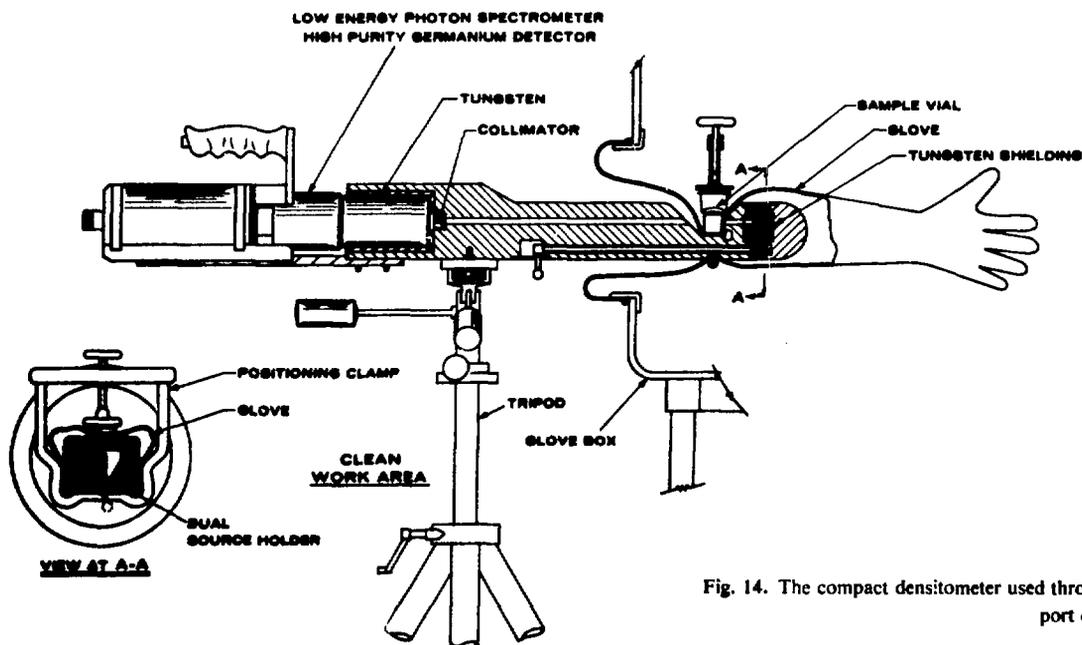


Fig. 14. The compact densitometer used through the glove port of a glovebox.

Recently the nondestructive gamma-ray technique has been used to determine plutonium isotopic compositions in bulk samples.<sup>29-31</sup> The technique uses either a small, planar, high-purity germanium detector (1 or 2 cm<sup>3</sup>) to analyze gamma-ray spectra in the 100- to 400-keV range or a large germanium or Ge(Li) photon detector (about 70 cm<sup>3</sup>) to analyze gamma-ray spectra in the 120- to 640-keV range. In general, the method successfully determines the isotopic compositions for <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>241</sup>Pu within a few hours count time. However, the precision for <sup>240</sup>Pu measurements<sup>31</sup> in both the 160- and 642-keV regions was 1 to 3% within about a 14-h count time for a plutonium sample mass less than 0.25 g. In this case, a counting time over 200 h would be required to assay a small sample with a plutonium mass less than 1000 µg to obtain a precision better than 3% for <sup>240</sup>Pu. Because a 200-h count time is not practical for monitoring product materials, the only useful gamma rays for analysis of small samples will be the high-intensity, low-energy gamma rays at 43.48, 45.23, and 51.63 keV from <sup>238</sup>Pu, <sup>240</sup>Pu, and <sup>239</sup>Pu, respectively.

Although these low-energy gamma rays have been used by Gunnink et al.<sup>32</sup> and Cowder et al.<sup>27</sup> for NDA of freshly separated solutions, no one has used this energy region for analyzing moderately aged samples in which the very intensive 59.54-keV line from <sup>241</sup>Am/<sup>237</sup>U dominates the spectra. The low-energy gamma rays become difficult to analyze because they are strongly interfered with by the Compton continuum of the 59.54-keV gamma ray. However, this difficulty can be lessened by carefully selecting a detector with the proper combination of resolution, efficiency, and peak-to-Compton ratio at energies below 60 keV. We tested five types of detector with the active volume varying from 1 to 70 cm<sup>3</sup> to determine which will meet our requirements. Figure 15 shows the low-energy (38- to 60-keV) gamma-ray spectra of sample SIS, 512, with a 20-ks count time. Although the IGP detector shows the best resolution and peak-to-Compton ratio, its efficiency is too low. On the other hand, the N-type coaxial (NTC) detector has the highest efficiency but the lowest resolution. The LGC and IGC detectors have poor resolutions, efficiencies, and peak-to-Compton ratios. The GLP detector has the best combination of resolution, efficiency, and peak-to-Compton ratio. Table VII summarizes the detector types, sizes, and resolutions and their precision of isotopic ratios. With a 20-ks count time, the highest estimated precision (1σ) of 0.4 and 0.56% for <sup>238</sup>Pu/<sup>239</sup>Pu and <sup>240</sup>Pu/<sup>239</sup>Pu, respectively, was obtained by the GLP detector. Based on these results, the GLP detector

appears the best choice for measuring small samples with plutonium mass less than 1000 µg.

We have analyzed two aged product samples (about 600 µg of plutonium) by using the GLP detector. Relative efficiency (RE) variations from sample self-absorption, detector efficiency, and external absorbers are calculated by linear interpolation between known efficiency points from <sup>239</sup>Pu gamma rays at 38.66 and 51.63 keV. A ln RE vs ln(ln E) interpolation is used, where E is the gamma-ray energy. Ratios are measured absolutely without standards using known half-lives and branching ratios. Small biases can be eliminated by additional calibration. The preliminary results compared with mass-spectrometer results are reported in Table VIII. Agreements between the gamma-spectroscopy and mass-spectrometer results are very good. The precision (1σ), estimated from counting statistics without including uncertainty from RE, is better than 0.6% for a 20-ks count time, which is compatible with throughputs of a few samples per day. The counting precision should improve proportionally with the square root of the counting time and sample mass.

As mentioned above, a simple linear ln RE vs ln(ln E) interpolation between two RE points at 38.66 and 51.63 keV has been used to calculate the relative efficiencies at 43.48 and 45.23 keV for the data in Table VIII. To optimize the best results, we have examined two other possible combinations of linear interpolations: ln RE vs ln E and RE vs E. The results of three linear interpolation methods for the same spectra (20-ks count time) on two samples taken by the GLP and NTC detectors are compared with those measured by mass spectrometry in Table IX. In general, the results from three linear interpolations are very close. The linear interpolation of ln RE vs ln E appears to have the best overall result. Careful calibration for the selected detector will be initiated to determine which linear interpolation should be used in the algorithms to extract the isotopic ratios from the spectra.

#### D. Element Identification by Thermal-Neutron-Capture Gamma Rays (*T. W. Crane, Q-1*)

Because most elements capture thermal neutrons and emit characteristic gamma rays,<sup>33-35</sup> neutron irradiation and gamma-ray counting can identify and measure elements. This technique can determine the practical limits of measuring the elemental content of 108-ℓ drums.

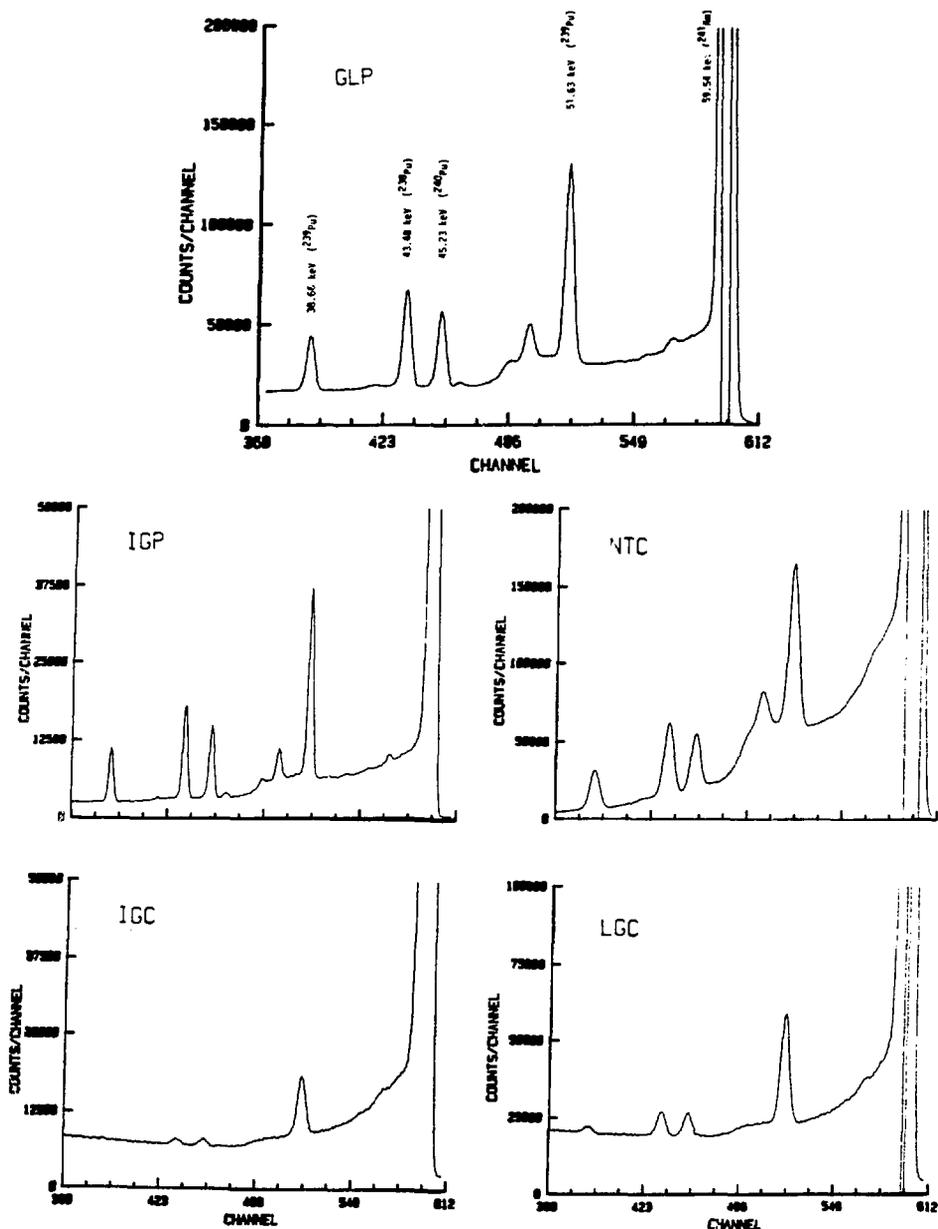


Fig. 15. Low-energy gamma-ray spectra from various types of detectors.

Table X shows the detectability limit for various elements in 208-l drums. The detectability limits (3 standard deviations above background, 100-s counting time) are based on experimental measurements using a 208-l drum test bed. These measurements used a 50- $\mu\text{g}$ - $^{252}\text{Cf}$  neutron source and a highly collimated, 16%-efficiency Ge(Li) detector. Samples were located in the central region of the drum. The sensitivity measured with

the test bed is approximately the same as would be achieved for a system with less collimation viewing the entire drum. With either a larger neutron source or larger gamma-ray detector, the sensitivity could be lowered.

As seen from Table X, the thermal-neutron-capture gamma-ray method is particularly sensitive to neutron poisons. A detectability limit below 1 g is achieved for all neutron poisons except lithium. The poor sensitivity for

**TABLE VII. Comparison of Detector Resolutions and Precisions for Isotopic Ratios from Various Types of Detectors**

Model	Type	Size	Volume (cm <sup>3</sup> )	Resolution (FWHM) at 51.63 keV (eV)	Estimated Precision (1σ) <sup>a</sup>	
					<sup>238</sup> Pu/ <sup>239</sup> Pu	<sup>240</sup> Pu/ <sup>239</sup> Pu
IGP	Planar	100 mm <sup>2</sup> by 10 mm	1	390	0.80	1.02
GLP	Planar	1000 mm <sup>2</sup> by 10 mm	10	510	0.40	0.56
IGC	Coaxial	37.5-mm diam by 29 mm	43	640	21	18
NTC	N-type coaxial	42.3-mm diam by 35 mm	49	730	0.48	0.58
LGC	Coaxial	49-mm diam by 39 mm	75	670	4.03	4.22

<sup>a</sup>Precisions (1σ) estimated from measurements of sample SIS 512 (about 600 μg Pu) with a 20-ks count time.

**TABLE VIII. Comparison of Isotopic Ratios by Gamma-Ray Spectroscopy (Gamma) with Mass Spectroscopy (Mass)**

Sample	Pu Mass (μg)	Count Time (ks)	<sup>238</sup> Pu/ <sup>239</sup> Pu			<sup>240</sup> Pu/ <sup>239</sup> Pu		
			(× 10 <sup>-3</sup> )			(× 10 <sup>-2</sup> )		
			Mass	Gamma	Mass/Gamma	Mass	Gamma	Mass/Gamma
SIS 512	~600	20	1.483	1.483 ± 0.006 <sup>a</sup>	1.000 ± 0.004	7.19	7.13 ± 0.04	1.008 ± 0.006
		80		1.491 ± 0.003	0.995 ± 0.002		7.14 ± 0.02	1.007 ± 0.003
SIS 513	~650	20	1.483	1.463 ± 0.007	1.014 ± 0.005	7.17	7.18 ± 0.04	0.999 ± 0.006
		80		1.475 ± 0.004	1.005 ± 0.003		7.19 ± 0.02	0.997 ± 0.006

<sup>a</sup>Precisions (1σ) are estimated from counting statistics. Uncertainties from estimated efficiencies were not included.

**TABLE IX. Comparison of Isotopic Ratios Among Three Linear Interpolation Methods**

Sample	Detector	Linear Interpolation	Mass/Gamma		
			<sup>238</sup> Pu/ <sup>239</sup> Pu	<sup>240</sup> Pu/ <sup>239</sup> Pu	<sup>238</sup> Pu/ <sup>240</sup> Pu
SIS 512	GLP	ln RE vs ln(ln E)	1.000	1.008	0.990
		ln RE vs ln E	0.997	1.004	0.990
		RE vs E	1.000	1.007	0.990
	NTC	ln RE vs ln(ln E)	1.010	0.976	1.035
		ln RE vs ln E	1.005	0.972	1.035
		RE vs E	1.017	0.982	1.035
SIS 513	GLP	ln RE vs ln(ln E)	1.014	1.000	1.015
		ln RE vs ln E	1.011	0.997	1.015
		RE vs E	1.012	0.997	1.015
	NTC	ln RE vs ln(ln E)	1.003	1.003	1.000
		ln RE vs ln E	0.999	0.999	1.000
		RE vs E	1.008	1.007	1.000

TABLE X. Elemental Thermal Neutron Capture Gamma-Ray Sensitivities for 208-l-Drum Assays

Element	Number of Gamma-Ray Lines <sup>a</sup>	Detectability Limit <sup>b</sup>
Hydrogen <sup>c</sup>	1	14.2 g
Helium	0	
Lithium <sup>d</sup>	7	3.2 kg
Beryllium	7	8.6 kg
Boron <sup>d</sup>	7	150 mg
Carbon	3	40.3 kg
Nitrogen <sup>c</sup>	43	1.7 kg
Oxygen	0	
Fluorine	11	5.6 kg
Sodium	51	176 g
Magnesium	18	286 g
Aluminum <sup>c</sup>	51	605 g
Silicon	27	970 g
Phosphorus	60	2.0 kg
Sulfur	33	400 g
Chlorine	41	15.3 g
Potassium	88	280 g
Calcium	46	792 g
Scandium	87	21.8 g
Titanium	39	45.2 g
Vanadium	62	68.2 g
Chromium	56	202 g
Manganese	76	48.2 g
Iron <sup>c</sup>	42	508 g
Cobalt	59	29 g
Nickel	49	122 g
Copper	66	95 g
Zinc	71	1.2 kg
Cadmium <sup>d</sup>	38	420 mg
Gadolinium <sup>d</sup>	17	879 mg
Mercury	41	3.2 g

<sup>a</sup>When usable, escape peaks are included.

<sup>b</sup>Count time of 1000 s, 3 standard deviations above background.

<sup>c</sup>Possible interference with measurement system components.

<sup>d</sup>Neutron poison.

lithium is because the primary neutron-absorbing isotope of lithium, <sup>6</sup>Li, captures neutrons without emitting gamma rays.

For nuclear waste management, the goal is to measure the fissile and transuranic (TRU) content. Methods for very low level detection, much less than 1 g, or methods

for high-density materials use active neutron interrogation to determine the quantity of fissile material. In some cases, the matrix materials can cause large errors. Neutron poisons such as boron or cadmium are particularly bad offenders. The thermal-neutron-capture gamma-ray technique can be used to detect these poisons and thereby avoid underestimating material. The elemental composition information can be used to establish that the waste has been correctly categorized.

Nuclear safeguards measurements also have matrix interference problems when fissile material is measured in large containers. The thermal-neutron-capture method also will help to identify poisons and assure that the correct material calibration is used. In addition, some defense facilities handle classified materials that often are neither fissile nor radioactive. However, because classification sometimes is based on material composition, the capture-neutron method might be used to screen for these items.

The results in Table X also can be applied to environmental monitoring of chemical wastes. Mercury can be readily identified (a 3-g detectability limit is about a 30-ppm sensitivity) and chlorine also is easily detected. Because chlorinated hydrocarbons often are hazardous (polychlorinated biphenyl, perchloroethane, etc.), chlorine in substances where chlorine is not normally found might be used as a signature to help identify hazards or possible illegal waste disposal practices.

E. Automated On-Line L-Edge Measurements of SNM Concentration for the AGNS Miniruns (*P. A. Russo, T. Marks, and M. M. Stephens, Q-1; A. L. Baker and D. D. Cobb, Q-4*)

Techniques for computerized accounting and control of nuclear materials at reprocessing facilities in near-real-time have been developed, tested, and evaluated by Los Alamos at the AGNS facility in Barnwell, South Carolina. AGNS had devised a series of week-long miniruns during which uranium solutions were recycled (in a closed loop) through the plutonium purification process. During these runs, data used by the facility for process monitoring and materials control were available for the materials accounting demonstrations. The details of these miniruns and the early results of efforts for NRTA have been presented in Ref. 36. Among the areas cited for improvement was the capability for accurate, on-line NRT assay of SNM concentration. Los Alamos provided an L-edge instrument,<sup>37,38</sup> modified for operation on flowing solution streams, for use in the final minirun of FY 81.

The on-line demonstration at AGNS required shipping a calibrated instrument, and the empirical calibration procedure required sealing the flow-through sample cell that was filled with a well-characterized reference solution of SNM. The calibration efforts at Los Alamos used solutions of depleted uranium at concentrations of 30, 45, and 60 g/l.

Use of a transmission measurement like that used in the L-edge assay of SNM concentrations in flowing solution streams requires<sup>39</sup> the capability for automatic control of the x-ray-generator (transmission-source) current to maintain a constant count rate in the Si(Li) detector as the sample concentration (and hence the transmission) changes. The design and testing of the required electronics for this capability composed one of two development efforts (the second being software) in modifying the existing instrument design for on-line operation.

The L-edge instrument in Figs. 16 and 17 was shipped to AGNS during the week of August 3 and was installed and tested during the week of August 10. The testing consisted of repeated L-edge assays of a well-characterized (50-mg/cm<sup>2</sup>) uranium foil. This measurement control procedure demonstrated consistency between the (precision and absolute result) foil assays at AGNS and the foil results obtained at Los Alamos during the calibration.

The L-edge instrument operated on-line, continuously and automatically, for the 1-wk minirun beginning on



Fig. 16. The L-edge equipment before on-line installation at AGNS. The electronics rack (far left) contains (bottom to top) the minicomputer, the MCA and display, pulse-processing electronics, oscilloscope, and dual floppy disk drive. The center station contains the x-ray generator power supply and controls and the measurement station on top (refer to Fig. 2). The hard-copy terminal is at the right.

August 18, 1981. The instrument automatically transmitted uranium concentration results, precise to about 0.7%, to the materials accounting and control system (MACS) computer once every 5 min throughout the run. These results were accessed by the programs designed for demonstration of NRTA.

The L-edge flow-through cell was plumbed into the line that samples the product of the 2B solvent-extraction column (that is, the 2BP stream). The flow diagram in Fig. 18 shows this stream within the plutonium purification cycle. In series with the L-edge cell in the 2BP sampler was an instrument for determining solution density and conductivity. This "densimeter," developed by AGNS, is used for process control purposes to evaluate uranium concentration in near-real-time using the measured density and an acidity deduced from the measured conductivity. Destructive analyses of solutions withdrawn from the 2BP sampling line were also performed three or four times daily.

The L-edge results were in reasonable agreement with the results of destructive analysis of samples withdrawn from the 2BP line for the duration of the minirun. Figure 19 shows the L-edge results (circles) and the results of the off-line destructive analysis (squares) plotted vs time. The L-edge results also showed reasonable agreement with the process control (densimeter) results for uranium concentration. A detailed presentation of the on-line L-edge results appears in Ref. 40.

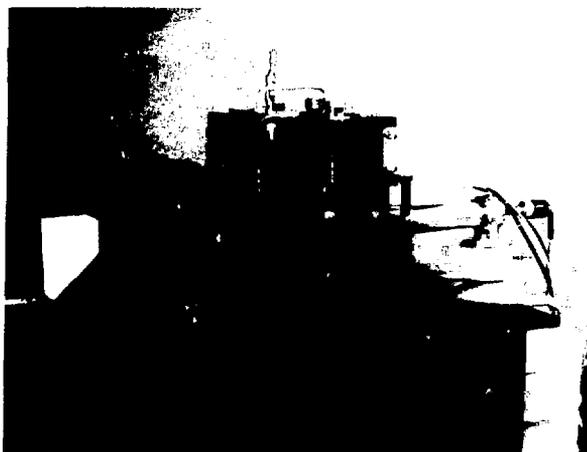


Fig. 17. The measurement station for the L-edge densimeter shows (left to right) the x-ray head, the plastic secondary containment box, and the Si(Li) detector. The inlet and outlet tubes for the flow-through sample cell are shown extending from the side of the secondary containment box. These tubes were plumbed into the 2BP sampling line for the on-line measurements of the flowing process stream.

energy  $E_0$  before escape at final energy  $E_p$  (Ref. 51). For a given initial energy  $E_0$ , the escape energy  $E_f$  can be directly related to the total path length  $L(E_0 \rightarrow E_f)$  and projected range  $R(E_0 \rightarrow E_f)$  traversed by the particle from birth to escape. Hence, by using a Monte Carlo code for ray tracing, the effective alpha-induced neutron production from the distribution of  $UF_6$  gas in an operating centrifuge may be computed.

The probability that an alpha particle of initial energy  $E_0$  will produce a neutron by an  $(\alpha, n)$  reaction within a material with macroscopic  $(\alpha, n)$  cross section  $\Sigma(e)$  and stopping power  $dE/dx(E)$  before escaping at energy  $E_f$  is given by

$$P(E_0 \rightarrow E_f) = \int_{E_0}^{E_f} \frac{\Sigma(E)}{dE/dx(E)} dE \quad (1)$$

A plot showing computed values of  $P(E_0 \rightarrow E_f)$  for the three initial alpha-particle energies of  $^{234}U$  is given in Ref. 51 and reproduced here as Fig. 21. The total path length traversed by the alpha particle in slowing from initial energy  $E_0$  to final energy  $E_f$  is given by

$$L(E_0 \rightarrow E_f) = \int_{E_0}^{E_f} \frac{dE}{dE/dx(E)} \quad (2)$$

Energetic alpha particles dissipate energy in inelastic collisions that result in ionization and excitation of the

surrounding atoms. The alpha-particle trajectory is affected only slightly in small momentum transfers with electrons; that is, the paths of energetic alpha particles tend to be straight.<sup>52,53</sup> Figure 22 shows  $L(E_0 \rightarrow E_f)$  vs  $E_f$  computed by numerically evaluating the integral in Eq. (2) for an initial alpha-particle energy of 4.773 MeV and for  $UF_6$  as the slowing down material. Along with the total path length, the projected range  $[R(E_0 \rightarrow E_f)]$  is also shown in Fig. 22. The relationships between total path length and projected range for alpha particles slowing in uranium and fluorine were taken from Ref. 54.

To use the "thin-target" neutron production data efficiently in Monte Carlo ray-tracing calculations, a data set was formed with projected range as the independent variable. These data are plotted in Fig. 23, showing neutron production by a 4.773-MeV alpha particle vs projected range in  $UF_6$ . This plot was fitted (with a maximum deviation of 22%) using the formula

$$P(r) = 0.95 + 0.53 \ln R \quad (3)$$

A calculational model of the distribution of  $UF_6$  density within the spinning rotor of an operating gas centrifuge was constructed for use in Monte Carlo simulations of the alpha-particle transport process, including neutron production. Figure 24 shows the centrifuge rotor and indicates the operational  $UF_6$  distribution. Integrating the radial component of the momentum equation, using the ideal gas law,<sup>55</sup> and rearranging yields an approximate expression for the radial  $UF_6$  density distribution. The  $UF_6$  mass density  $\rho$

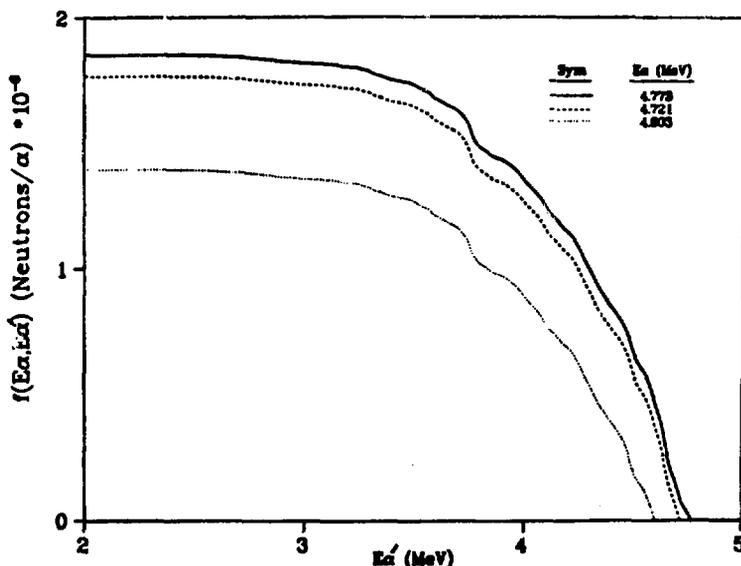


Fig. 21. Neutron production probability for the 4.603-, 4.721-, and 4.773-MeV alpha particles of  $^{234}U$  before escape at energy  $E_a$ .

Fig. 22. Total path length and projected range of a 4.77-MeV alpha particle in UF<sub>6</sub> vs terminal energy.

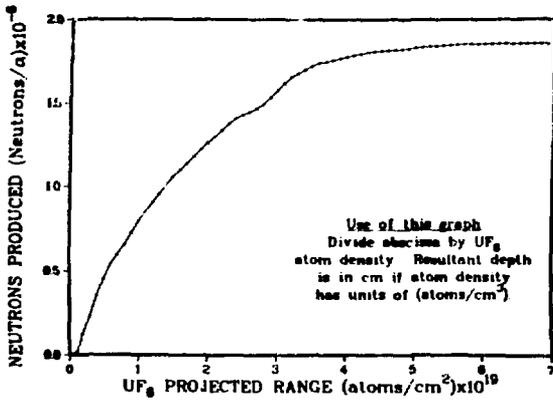
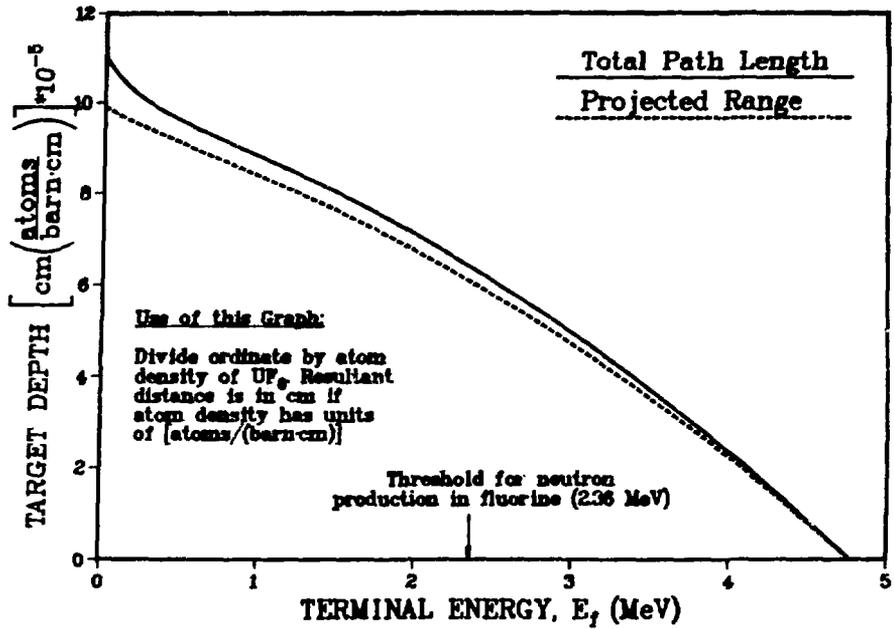


Fig. 23. Neutron production by a 4.77-MeV alpha particle vs projected range in UF<sub>6</sub>.

M is the UF<sub>6</sub> molar mass, Ωa is the rotational velocity of the rotor wall, R is the ideal gas constant, and T is temperature. The differential mass element for UF<sub>6</sub> gas between the rotor center post ( $r = r_0$ ) and the rotor wall ( $r = a$ ) for a rotor of height h is given by

$$dm = 2\pi h \rho(r) r dr \quad (6)$$

Integrating the mass element between r and a and normalizing yields an expression for the fraction of UF<sub>6</sub> lying between r and a. The fraction F is given by

$$F = \frac{1 - e^{-y}}{1 - e^{-y_0}} \quad (7)$$

where

$$y = -A^2 \left( \frac{1 - r^2}{a^2} \right) \quad (8)$$

and

$$y_0 = -A^2 \left( \frac{1 - r_0^2}{a^2} \right) \quad (9)$$

at radius r is given as a function of the wall mass density ρ(a) by the expression

$$\rho(r) = \rho(a) \exp \left[ -A^2 \left( \frac{1 - r^2}{a^2} \right) \right] \quad (4)$$

where

$$A^2 = \frac{M \Omega^2 a^2}{2 R T_0} \quad (5)$$

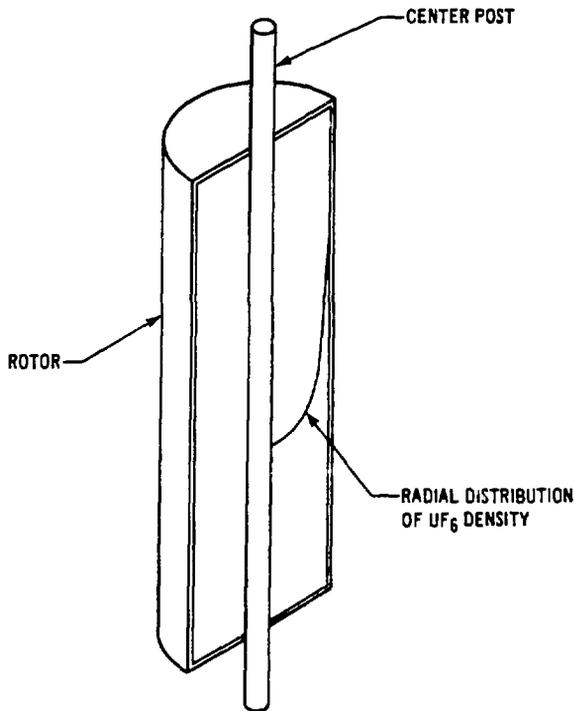


Fig. 24. The centrifuge rotor, indicating operational UF<sub>6</sub> distribution.

Manipulation of Eq. (7) yields the formula

$$\frac{r}{a} = \left[ 1 + \frac{\ln(1 - F')^{1/2}}{A^2} \right], \quad (10)$$

where

$$F' = F(1 - e^{-y_0}). \quad (11)$$

For Monte Carlo transport simulations, a special source subroutine was prepared for the MCNP code<sup>56</sup> to specify the initial conditions of alpha particles. Equation (10) was used in the source subroutine to sample randomly for the starting position, and particles were transported in a geometry similar to that of Fig. 24. The UF<sub>6</sub> gas volume inside the centrifuge rotor was divided into 10 radial zones. For each case considered, zone boundaries were adjusted so that 10% of the UF<sub>6</sub> gas was contained in each zone. In the transport simulation, straight-line alpha-particle paths were tracked through the zones of varying UF<sub>6</sub> density. As particles crossed zone boundaries, target depth was accumulated. At the point of escape from the geometry, the neutron produc-

tion associated with the accumulated target depth was recorded using Eq. (3) and a special tally subroutine. Enough particle histories were simulated to satisfy a preset statistical precision.

The data and methods described above were used to calculate neutron production for a US gas centrifuge rotor model. Neutron production relative to the thick-target value was computed over a range of operating parameters. Figure 25 shows relative thin-target neutron production vs average UF<sub>6</sub> density for a fixed rotor speed. The chosen dynamic range of UF<sub>6</sub> densities in Fig. 25 spans those currently planned for the Portsmouth Gas Centrifuge Enrichment Plant (GCEP). Figure 25 shows relative neutron production to be a fairly strong function of average UF<sub>6</sub> density over a dynamic range of densities typical of centrifuge operating conditions. Figure 26 shows, for fixed average UF<sub>6</sub> density, the relative neutron production vs rotor speed. Over a large dynamic

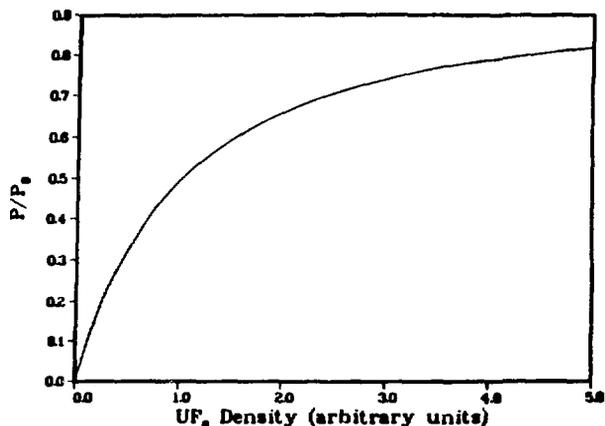


Fig. 25. Relative thin-target neutron production in an operating centrifuge vs average UF<sub>6</sub> density.

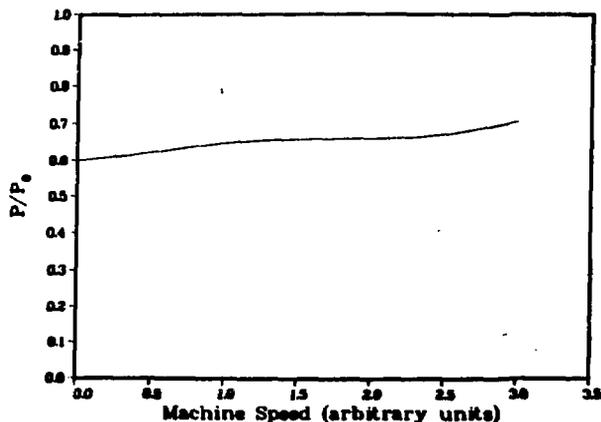


Fig. 26. Relative thin-target neutron production in an operating centrifuge vs machine speed.

range, Fig. 26 indicates that relative neutron production is weakly dependent on the speed of the rotor and thus on the internal  $UF_6$  density distribution.

Source-term data, like those in Figs. 25 and 26, are being used to predict effectiveness of neutron monitor arrays for detecting HEU production in GCEP centrifuge cascades as well as those characteristic of foreign technology.

**2. Gas-Phase  $UF_6$  Enrichment Monitor (R. B. Strittmatter, S. C. Bourret, S. S. Johnson, J. N. Leavitt, and R. W. Slice, Q-1).** The gas-phase  $UF_6$  enrichment monitor was installed in the Paducah-product feed station of the Oak Ridge Gaseous Diffusion Plant for field test and evaluation. The first process  $UF_6$  feed material was introduced into the monitor on January 15. Calibration data are being collected and the initial calibration should be completed in February. After this calibration period the monitor will be in continuous operation, performing a near 100% sampling of  $UF_6$  from the Paducah-product feed station into the diffusion plant.

The monitor design, which was engineered for in-plant use, is based on the measurement experience and the performance data of a laboratory prototype described previously.<sup>57,58</sup> The NaI(Tl)-based instrument was designed to be rugged and unobtrusive to plant operations and to require minimal operator intervention, while providing the sophisticated design to perform measurement control and verification functions. (See Figs. 27, 28, and 29.) Major components of the monitor include the measurement chamber, detector, measurement verification device, evacuation and sampling system, by-pass plumbing, electronics, and software.

**a. Measurement Chamber.** The cylindrical measurement chamber is of monel construction with thin windows for the transmission of 60-keV gamma rays from an external  $^{241}Am$  source. The chamber is 11.4 cm in diameter and 9.8 cm tall. The height of the chamber was optimized for an operating pressure of 15 psia. The top and bottom windows are 0.075 and 0.152 cm thick, respectively. The chamber is heated by the facility steam that flows through channels in the monel block.

**b. NaI(Tl) Detector.** The NaI(Tl) detector whose preamplifier/base can be seen extending from the lead shield in Fig. 29 has a 12.7-cm-diam, 1.27-cm-thick NaI(Tl) crystal coupled to a 12.7-cm-diam pure NaI light pipe. The resolution of this detector is 7.2% at 662 keV. A thermal barrier between the detector and the heated measurement chamber is provided by a 0.16-cm-thick sheet of phenolic located below the measurement chamber. The two copper tubes on either side of the detector shielding in Fig. 29 are for forced-air cooling of the detector face, if necessary.

**c. Measurement Verification Device.** This device, used to provide system verification and secondary-calibration data, is attached to the top of the measurement chamber. It consists of a Geneva-mechanism that drives a tungsten wheel with four positions and a stationary 1-mCi- $^{241}Am$  source. The  $^{241}Am$  source used for the transmission measurement is located directly above the tungsten wheel. In the normal operating mode a hole in the wheel provides an unobstructed path for the  $^{241}Am$  60-keV gamma rays. The three other positions provide verification and calibration data. The second position contains a 0.007-cm-thick, 93%-enriched uranium foil to provide a

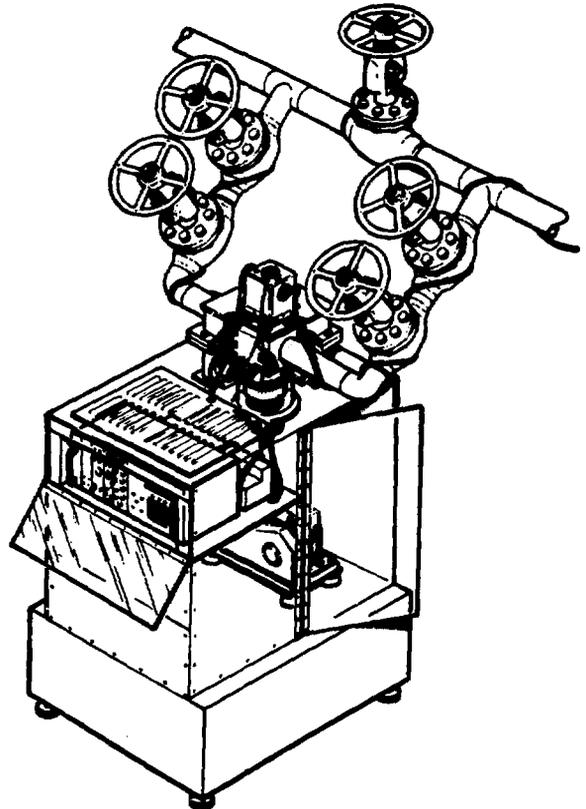


Fig. 27. The enrichment monitor, showing the major components and the associated by-passing plumbing.

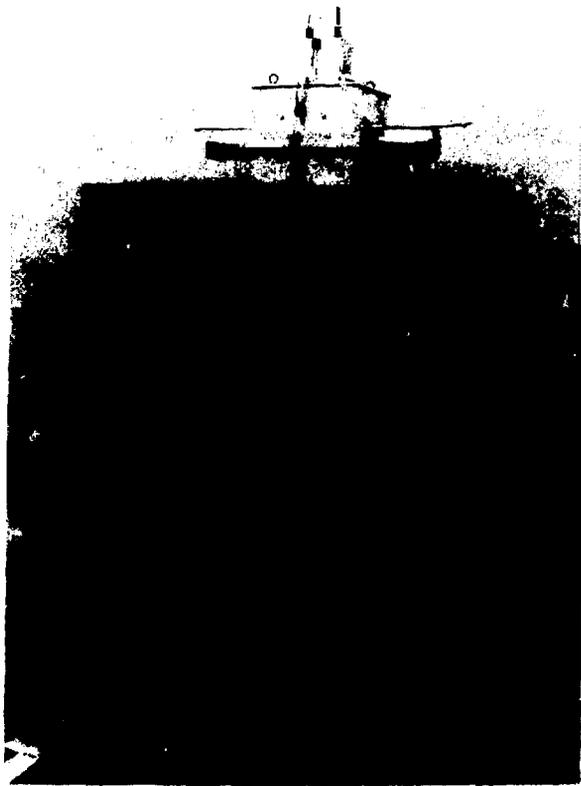


Fig. 28. Front view of the enrichment monitor before installation, showing system electronics, shielding enclosure for measurement chamber, and 3-in. feed lines.

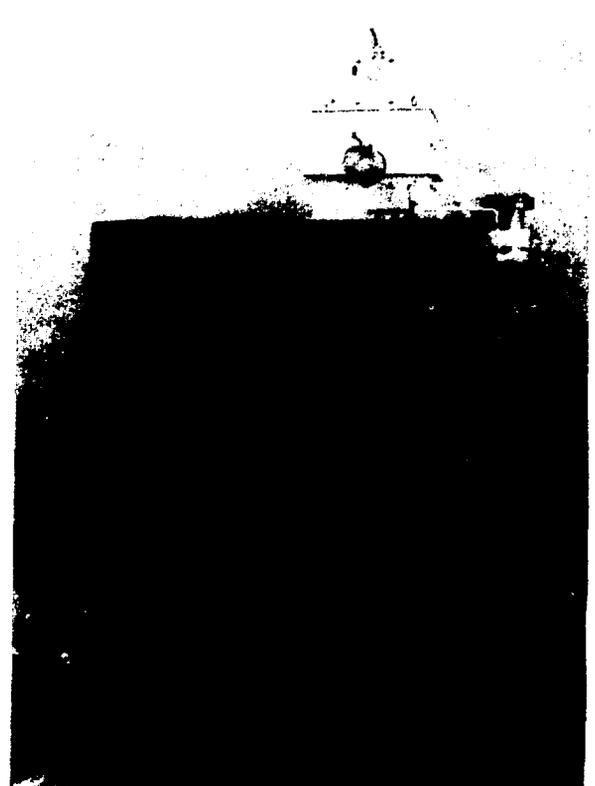


Fig. 29. Side view of enrichment monitor before installation, showing detector tube base, vacuum pump, chemical trap, and sampling station.

measurement of the 186-keV gamma-ray intensity from a standard source. Because the transmission of 60-keV gamma rays through this disk is similar to that through the  $UF_6$  for a normal operating pressure, it also provides data to check the  $UF_6$  density measurement. The third position contains a 0.075-cm-thick stainless steel foil that provides additional data to check the  $UF_6$  density measurement. In the fourth position the 0.48-cm-thick tungsten wheel shields the  $^{241}Am$  source from the detector and allows the measurement of the background under the 60-keV peak.

*d. Evacuation and Sampling System.* The evacuation and sampling station provides the flexibility to perform calibration and background measurements necessary for a thorough test and evaluation. Experience gained during the test and evaluation may indicate that the evacuation and/or sampling stations are not necessary for future instrument designs. The evacuation station is based on a

mechanical vacuum pump and chemical trap. The chemical trap, sampling connection, vacuum pump, and associated plumbing can be partially viewed in Fig. 29. The chemical trap is loaded with soda lime and should provide several years service before the trap material must be changed. If a pump fails, pump oil cannot enter the chemical trap because of a pressure-sensor switch, solenoid valve, and control electronics.

*e. By-Pass Plumbing.* The by-pass plumbing provides a means by which a normal  $UF_6$  flow can be delivered to the diffusion plant, if for any reason flow through the measurement chamber is not possible. When flow through the chamber is shut off for calibration measurements, the chamber is evacuated and a  $UF_6$  standard can be introduced into the chamber. The double valves shown in Fig. 27 are required for safety reasons if the feed station is operated while the sample chamber is not installed.

*f. Electronics.* The electronics system for detector operation, signal processing, data acquisition, data manipulation, and system control is based on the NIM standard using a combination of commercial and custom-designed components. The programmable data-acquisition and control instrument, the PDAC-100, developed by Los Alamos, is a microprocessor-based MCA packaged in a six-wide NIM module. It is capable of computation, control, and input/output functions. The 1024-channel, 20-bit MCA memory stores data from an external commercial ADC. The data are manipulated by a set of algorithms preprogrammed in read-only memory (ROM). EAROM, an electrically alterable, nonvolatile memory, is used for storage of calibration factors and parameters that are entered through a 20-key pad located on the front panel.

Output is provided through a front-panel LED display and through two serial and two parallel ports. One serial port is now used for a remote printer and a modem for data transmission over a telephone line. Communication to the device is normally implemented through the front panel keypad, although input also can be received through the input/output ports.

Because the need for spectral display is usually limited to system verification and calibration, the spectral display is through an external x-y-z oscilloscope.

*g. Software.* Operation of the enrichment monitor is controlled by a FORTRAN and assembly language program stored in ROM. Normal operation of the program consists of repeated assay cycles that start automatically at power-up and require no operator interaction. During a cycle the program starts and stops the ADC, retrieves channel counts, performs the density and enrichment analyses, and then displays the enrichment percentage on the LED display. The printer is an optional device that is used to log the assay cycles and to indicate any errors during operation. The CRT, also optional, displays the gamma-ray spectrum and can be manipulated by the operator at the keypad to expand regions of interest and movement of the cursor.

The operator can stop acquisition to use one of the other available functions. If a calibration is selected, assay cycles are performed with the wheel in any position, and all intermediate results, such as peak areas, are logged on the printer. The operator can also choose to view or change system parameters, such as peak windows or calibration constants. After any changes the new parameters are stored in EAROM and are read into

random access memory (RAM) at the next program restart.

**I. High-Temperature Gas-Cooled Reactor (HTGR) Scrap Measurements (*J. Sprinkle and N. Ensslin, Q-1; H. R. Baxman, CMB-8*)**

The Physical Chemistry and Metallurgy Group (CMB-8) at Los Alamos acquired 100 buckets of HTGR scrap for evaluating a recovery procedure. They desired an upgrade of their NDA system<sup>59</sup> to handle this material. A temporary solution to this request was mutually agreed on, with further work depending on whether they acquire more material of this type. Four containers of HTGR scrap were milled and blended by CMB-8, then placed in 250-ml graduated cylinders for assay on the Q-1 segmented gamma scanner (see Table XI). Six samples were taken from each can for assay, and the listed uncertainties are the standard deviations of the six results. These uncertainties are less than or equivalent to the estimated precision for single assays. These assay values confirmed the success of the blending and provided standards for calibrating the random driver neutron counter used to assay the scrap buckets (see Table XII). Ninety percent of the cans have from 50 to 67 g of uranium and can be assayed with a linear calibration. Cans with heavier loadings will be estimated from these data. A more precise calibration will be deferred until more HTGR material is received by CMB-8.

Because of the high ratio of thorium to uranium in this material, future assays of process solutions by the uranium solution assay system will require considerably larger transmission corrections than are normally used in this instrument. The Type I solutions (10 to 500 ppm) will be assayed as Type II (normally 0.5 to 50 g/l), with longer count times to obtain a transmission correction. The Type II solutions will be restricted to a transmission

TABLE XI. Results for HTGR Samples

ID	SGS (g)	TAG (g)	SGS/TAG
CAN 03	2.84 ± 0.08	2.78	1.02
CAN -3	3.26 ± 0.14	3.32	0.97
CAN 98	3.18 ± 0.04	5.91	0.54
CAN 99	19.01 ± 0.30	12.44	1.53

TABLE XII. Results for Cans of HTGR Grounds

ID	TAG (g)	SGS (g)	Net Coincidence	Net Coincidence per g	Correction Coincidence	Correction Coincidence per g
CAN 03	58	59	19.78	0.335	2.54	0.0431
CAN -3	51	50	18.05	0.361	2.23	0.0446
CAN 98	125	67	23.33	0.348	3.10	0.0463
CAN 99	256	391	36.55	0.093	9.50	0.0243

above 75%, the region where the transmission correction is valid. The Type III product solutions should not contain thorium, a fact to be verified by a visual inspection of the passive gamma-ray spectrum.

J. Spent-Fuel Assay

1. Self-Interrogation of Spent Fuel (*D. M. Lee, Q-1; L. O. Lindquist, Q-2*). Whereas active assay is the one technique that measures directly the fissile content of spent fuel, the requirement for a high-intensity neutron source can be a serious disadvantage. A technique has recently been developed that eliminates the need for external isotopic sources, yet retains the advantages of an active assay system.<sup>60</sup>

A self-interrogation system is shown in Fig. 30. The reflector is a material with a high neutron-absorption cross section. In this method, the assay is accomplished by changing the reflector from water to a material that absorbs neutrons. By absorbing neutrons, the induced fissions in the fissile material resulting from those neutrons that normally would have reflected back into the fuel assembly are eliminated.

Mathematically, the neutron count rate  $N_R$ , with the absorber in place, can be written as

$$N_R = N_p - N_j \quad (12)$$

where  $N_p$  is the passive neutron count rate with a water reflector and  $N_j$  is the count rate reduction when the absorber is in place.  $N_j$  depends on the original passive rate  $N_p$ , the fissile content, and geometrical factors.  $N_j$  can be expressed as

$$N_j = f(x, y, z, U, Pu, \dots) N_p \quad (13)$$

where the dependence on the passive neutron rate is explicitly shown. The unspecified function  $f$  contains all

other variables. Combining and rearranging Eqs. (12) and (13), we can express the rate  $N_j$  as

$$-N_j = N_R - N_p = -f(x, y, z, Pu, U \dots) N_p \quad (14)$$

The unspecified function  $f$ , which is very complex, contains all parameters describing the fuel assembly, including the geometry, reflector, moderator, and fissile content. Because, in general, the fissile content is the only variable, a correlation to  $f$  provides a means of assaying the fuel assembly.

The undefined function  $f$  can be approximately determined by treating the fuel assembly as a multiplying system.<sup>6</sup> In this approximation the passive neutron rate  $N_p$  with a water reflector can be written as

$$N_p = M_1 S_0 \quad ; \quad M_1 \approx \frac{1}{1 - k_1} \quad (15)$$

where  $S_0$  is the unmultiplied source strength of the fuel assembly,  $M_1$  is the multiplication of the fuel assembly,

\*The validity of Eq. (15) is probably good for values of  $k \geq 0.9$  but becomes only approximate for  $k \leq 0.9$ . However, the simple mathematical treatment provided by Eq. (15) helps to explain the basic principles of this measurement technique.

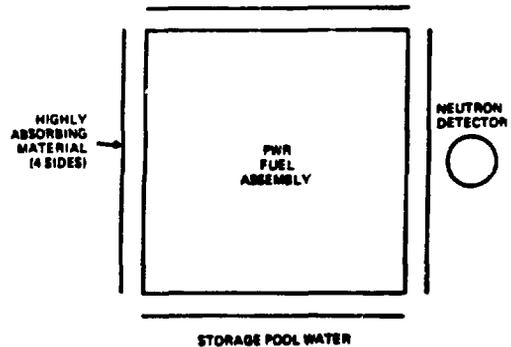


Fig. 30. A generic self-interrogation system.

and  $k_1$  is the multiplication constant. A similar expression can be written for the neutron rate  $N_R$  when the absorbing reflector is in place,

$$N_R = M_2 S_0 ; \quad M_2 \approx \frac{1}{1 - k_2} \quad (16)$$

Combining Eqs. (15) and (16) and rearranging,

$$\begin{aligned} N_R - N_p &= M_1 M_2 \Delta k S_0 \\ &= N_p M_2 \Delta k \\ &= \frac{\Delta k}{1 - k_1 + \Delta k} N_p \end{aligned} \quad (17)$$

where

$$\Delta k = k_1 - k_2 \quad (18)$$

If Eqs. (14) and (17) are compared, the function  $f$  can be written as

$$f = \frac{-\Delta k}{1 - k_1 + \Delta k} = -M_2 \Delta k \quad (19)$$

From Eq. (19) the function  $f$ , which is the fractional count-rate change when the absorbing material is in place, depends on the change in reactivity of the fuel assembly.

The results of preliminary calculations and measurements on the correlation of the function  $f$  to burnup are shown in Fig. 31. Based on these results, the estimated sensitivity at about 35 000 MWd/tU for about a 2.4% precision on  $f$  is a 1000-MWd/tU precision on burnup. The advantages of this method are that no extra neutron sources are required and any spent-fuel assembly, no matter how intense the passive neutron rate, can be assayed.

**2. Calculated Gamma-Ray Responses to Sources Placed in PWR Fuel Assemblies (J. R. Phillips and G. E. Bosler, Q-1).** The relative responses of a hypothetical detector were computed for gamma-ray sources placed in specified fuel pin locations in a 15-by-15 pressurized-water-reactor (PWR) fuel assembly. Gamma-ray energies of the sources ranged from 511 to 2186 keV,

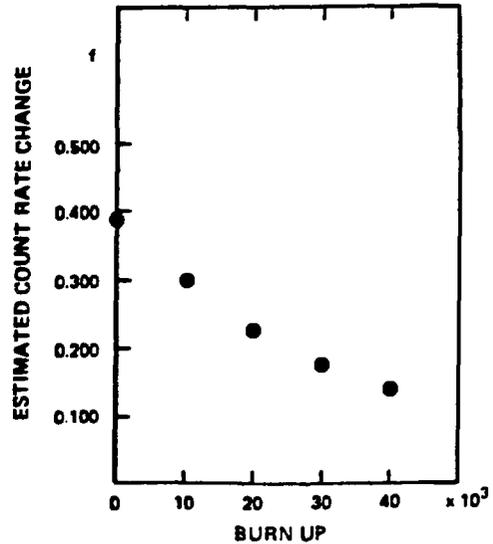


Fig. 31. Estimated fractional count-rate change vs burnup (MWd/tU).

thus covering the energy range in which most fission and activation products emit gamma rays. The paths of individual gamma rays were traced using a Monte Carlo program<sup>56</sup> that takes into account the incoherent and coherent scattering as well as the photoelectric effect and pair production. The code was modified to allow use of a general lattice\* for describing the fuel assembly. Each fuel pin and guide tube are modeled, and the gamma rays are transported through the lattice in exactly the same manner as they would be in the actual fuel assembly. The response to placing the simulated sources in various positions was calculated to determine quantitatively where the gamma rays that are emitted by a fuel assembly really originate.

The isotopes associated with the seven gamma rays investigated are listed in Table XIII. Of these isotopes,  $^{137}\text{Cs}$  is most often used to estimate the relative or absolute burnup of fuel material. The isotope ratios,  $^{134}\text{Cs}/^{137}\text{Cs}$  and  $^{154}\text{Eu}/^{137}\text{Cs}$ , can also be used to estimate the exposure of irradiated fuel materials.<sup>61</sup> The  $^{106}\text{Rh}$  isotopes can be used to estimate the fraction of fissions from  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , and the  $^{144}\text{Pr}$  isotope can be used to estimate the cooling time since discharge from the reactor.

Several scanning geometries were simulated, but only one will be described. Figure 32 shows the scanning

\*Modifications were furnished by G. W. Eccleston, G. E. Bosler, and R. G. Schrandt, Los Alamos National Laboratory, 1981.

**TABLE XIII. Fission Product Gamma Rays from Irradiated PWR Fuel Material**

Isotope	Gamma-Ray		Half-Life
	Energy (keV)	Branching Ratio	
<sup>106</sup> Rh	512 <sup>a</sup>	0.21	29.8 s (parent is <sup>106</sup> Ru with a 366.4-day half-life)
<sup>134</sup> Cs	604.6	0.976	2.062 yr
	795.8	0.854	
<sup>137</sup> Cs	661.6	0.851	30.17 yr
<sup>154</sup> Eu	1274.4	0.355	8.5 yr
<sup>144</sup> Pr	1489.2	0.0026	17.3 min (parent is <sup>144</sup> Ce with a 284.5-day half-life)
	2185.6	0.0066	

<sup>a</sup>Can have interference from annihilation gamma rays with energies of 511 keV.

geometry in which the sources were placed in sequential rows in the 15-by-15 PWR fuel assembly. Gamma-ray sources were uniformly distributed within the seven counter fuel pins in each row, and the intensities were calculated at the end of the 1-m collimator. Table XIV gives the percentage of gamma rays from each row that contributes to the total calculated response. For example, 74.6% of the total gamma flux of the 511-keV gamma ray originates in the first row, and 17.2% originates in the second row.

Over 90% of the <sup>137</sup>Cs signal originates in the outer three rows of the fuel assembly; therefore, its signal is not really representative of the entire cross section of the fuel assembly. To obtain the 90% limit, over five rows must be considered when measuring the 2186-keV gamma ray of <sup>144</sup>Pr. The results of these calculations can be used to interpret more accurately the results obtained from the gamma-ray spectrometry measurements of spent-fuel assemblies.

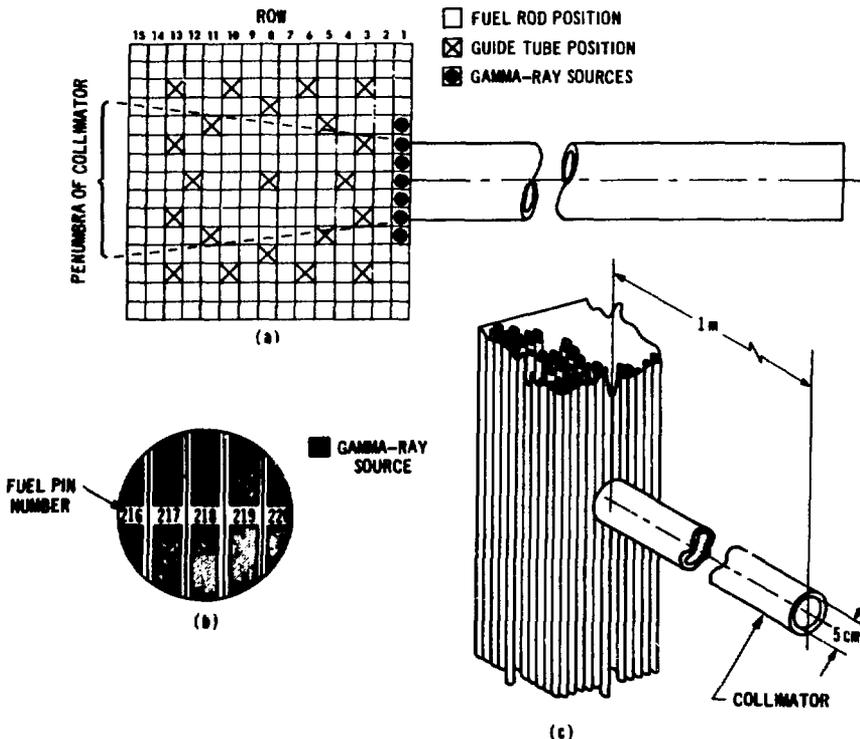


Fig. 32. Model used for Monte Carlo calculations.

TABLE XIV. Percentage of Gamma Rays from Each Row Based on the Total Values

Gamma-Ray Energy (keV)	Row Number							
	1	2	3	4	5	6	7	8
511.0	74.6 ± 3.0	17.2 ± 1.0	6.1 ± 0.5	2.1 ± 0.2	---	---	---	---
604.6	67.6 ± 2.6	20.6 ± 1.2	8.6 ± 0.6	3.2 ± 0.3	---	---	---	---
661.6	61.3 ± 2.5	20.7 ± 1.2	9.5 ± 0.6	3.5 ± 0.3	3.4 ± 0.3	1.0 ± 0.1	0.6 ± 0.1	---
795.8	54.0 ± 2.1	22.4 ± 1.2	11.9 ± 0.7	5.2 ± 0.4	4.6 ± 0.3	1.9 ± 0.1	---	---
1275.0	40.2 ± 1.4	22.4 ± 1.0	15.2 ± 0.7	7.7 ± 0.4	8.3 ± 0.4	4.0 ± 0.2	2.2 ± 0.1	---
1489.0	36.6 ± 1.2	21.9 ± 0.9	15.9 ± 0.6	8.8 ± 0.4	9.4 ± 0.4	4.6 ± 0.2	2.8 ± 0.2	---
2186.0	32.8 ± 1.1	20.7 ± 0.8	15.9 ± 0.6	9.1 ± 0.4	9.9 ± 0.4	5.4 ± 0.2	3.7 ± 0.2	2.5 ± 0.1

3. *Calculated and Measured Fuel Assembly Responses (G. W. Eccleston, Q-1).* Experiments and calculations are in progress to determine the effects of fissile distribution and loading in a fuel assembly on a steady-state neutron measurement. Calculations provide the external-source-induced fission neutron density ( $\nu_j F_{ij}$ ) by isotope  $j$  and fuel pin  $i$  throughout the assembly. Detector response  $R_i$  to neutrons emitted from fuel pin  $i$  is determined experimentally by inserting a  $^{252}\text{Cf}$  neutron source into a fuel pin. The total detector response  $R_T$  to fissions induced in the fuel assembly then can be determined according to

$$R_T = R_s + \sum_i \sum_j \nu_j F_{ij} R_i$$

where

$R_s$  = detector response from the external source,

$F_{ij}$  = fission rate of isotope  $j$  in pin  $i$ ,

$\nu_j$  = neutrons produced from a fission of isotope  $j$ , and

$R_i$  = detector response resulting from a neutron emitted in pin  $i$ .

Initial work has focused on an unirradiated 3.2%-enriched  $\text{UO}_2$  15-by-15 PWR fuel assembly immersed in water. In Fig. 33, the induced fission neutrons ( $\nu_j F_{ij}$ ) are the dotted surface, and the detector response for neutrons started in pins are the solid surface. A plan view of the experimental geometry is shown in Fig. 34. A single  $^{235}\text{U}$  fission chamber was used to measure neutrons emitted from the pins; therefore, the response distribu-

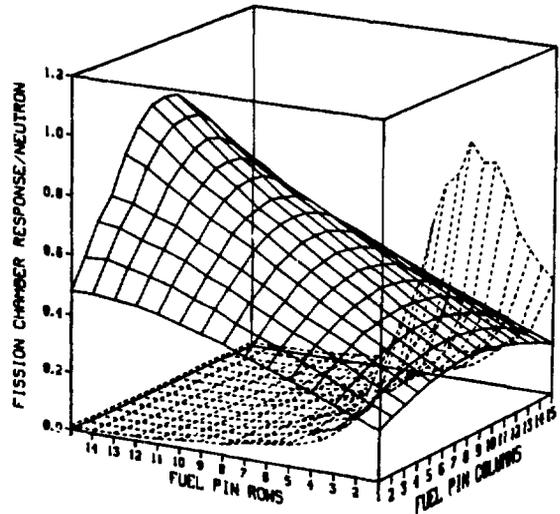


Fig. 33. Fuel array source-induced fission distribution and fuel pin detector response distribution.

tion decreases near the edges of the assembly. Fission events were induced in the fuel pins by a single  $^{252}\text{Cf}$  source separated by 2.5 cm from the assembly edge.

Based on initial calculations and experiments, a measurement geometry is being designed to equalize the detector sensitivity to fuel pins throughout the fuel array. This design will incorporate a source-tailoring region to flatten the fission rate along the fuel assembly rows. In addition, several detectors will be used along the opposite side of the array to produce a more uniform response to pins at the assembly edge.

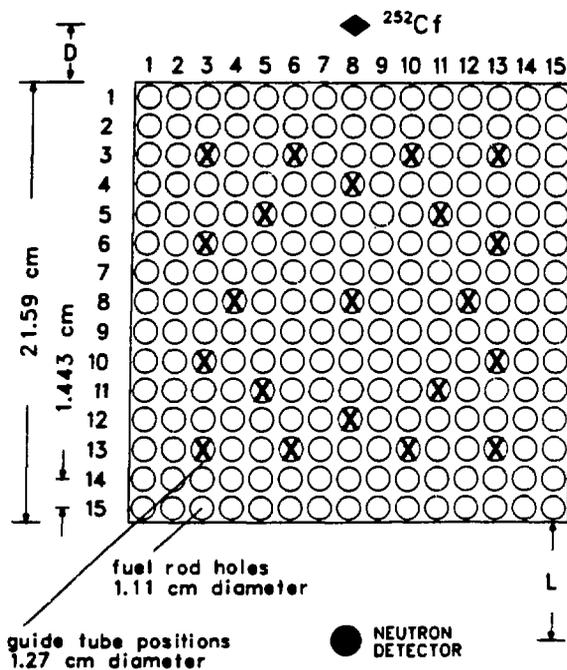


Fig. 34. Plan view of the PWR fuel assembly geometry.

#### IV. SYSTEMS DESIGN AND INTEGRATION

##### A. Design of Integrated Materials Accounting and Physical Security Systems (*J. T. Markin, E. A. Hakkila, A. L. Baker, T. R. Canada, R. G. Gutmacher, W. J. Whitty, and J. P. Shipley, Q-4*)

The first phase of the joint Sandia National Laboratories (SNL)/Los Alamos study was completed with publication of a two-volume report outlining a methodology for designing and evaluating an integrated safeguards system.<sup>62</sup> This methodology is applicable to the safeguarding of all DOE nuclear facilities with respect to sabotage and materials loss. Design and evaluation of an integrated safeguards system are divided into three steps: definition of safeguards objectives within the facility, synthesis of candidate designs that accomplish these objectives, and evaluation of these designs using performance measures that indicate the degree of accomplishment.

Facilities within the nuclear fuel cycle are categorized according to the type of material used and the safeguards concerns associated with operations on that material. Therefore, broad safeguards objectives for each facility type should be stated. Synthesis of safeguards designs that meet these objectives consists of identifying safe-

guards subsystems relevant to the objectives, identifying design constraints such as noninterference with process or safety, and identifying interfaces and relationships between facility and safeguards activities where interference or mutual reinforcement is possible. The last activity can highlight the integration potential between safeguards elements and facility elements. Evaluating candidate system performance addresses both abrupt and protracted materials-loss scenarios with the performance measures probability of detection, expected time to detection, and probability of interrupting the adversary. The final process of selecting a safeguards system is facilitated by an optimization scheme for selecting subsystems that maximize overall systems performance within a cost constraint.

##### B. Systems Concepts for DOE Facilities: Analyses of PF/LASS Data Base (*R. C. Bearse and R. M. Tisinger, Q-4; C. E. Nordeen, CMB-11*)

Our objective is continuing development of techniques for improved nuclear materials accounting that provide the technical foundation for specific facility support

efforts and the basis for MC&A systems design criteria. Activities include examining existing near-real-time MC&A systems, developing a methodology for handling and analyzing data generated by such systems, and constructing techniques for structuring safeguards systems to take maximum advantage of facility and process design features.

Primary efforts during this report period were directed towards completion of the analysis of the PF/LASS data base, documentation of decision analysis techniques for NRTA, and application of decision analysis techniques to both actual and simulated FFTF process data.

The PF/LASS data analysis effort was designed to provide an understanding of the data base, with emphasis on automated MIP analysis and MIP and CUSUM (cumulative MIP sums) charting. This effort involved analysis of measurement code assignments, measurement uncertainty evaluation and assignments, data-base editing techniques, error propagation, and generation of MIPs and their associated errors.<sup>63</sup>

We have transferred the PF/LASS data base to the VAX computer so that actual FFTF process data can be analyzed using DECANAL. However, to evaluate the effects of process changes, alternative measurement techniques, and accounting strategies on materials accounting and control in the FFTF process without undue interference with the operation, we will use modeling and simulation techniques. Presently we are upgrading the FFTF model code to permit simulations that are more representative of the actual operation.

Work in the planning stage includes (1) converting the simulation from the GASP-IV-based MODEL code to the SLAM-based MODEL code, (2) adding a 4-h overtime shift on Tuesday and Thursday every other week to the process models, and (3) adding Monday morning startup and Friday afternoon shutdown delay effects to the process model.

#### C. Development of Decision Analysis Techniques (*J. T. Markin, A. L. Baker and J. P. Shipley, Q-4*)

Materials accounting for SNM in fuel-cycle facilities is implemented more efficiently by applying decision-analysis methods, based on estimation and detection theory, to analyze process data for materials loss. To apply decision-analysis techniques to NRTA data, Los Alamos

has developed the computer code DECANAL. A draft report on the application and theory of the program will be issued as a DECANAL User's Manual.<sup>64</sup> The manual begins with an overview of the program, including a brief discussion of statistical tests such as the Shewhart chart and CUSUM that are used in the program. The manual describes the data input requirements, which are the data needed to close a materials balance and calculate the variance of the balance. Data input procedures are illustrated with several examples.

DECANAL output consists of the results of several statistical tests for materials loss and estimates of the amount of material lost. Example problems in the manual show how the output is used in making decisions about materials losses that have a specified detection probability and a controlled false-alarm rate.

The DECANAL User's Manual provides an easily applied materials accounting tool that does not require any advanced knowledge of statistics or decision theory.

#### D. Simulation Upgrades (*E. A. Kern and D. P. Martinez, Q-4*)

A user's manual for the measurement simulation code MEASIM was written during this reporting period.<sup>65</sup> The MEASIM code was developed over 5 yr, primarily for modeling the process measurements in nuclear materials processing facilities. It has been used exclusively in the performance assessment of materials accounting systems. The User's Manual provides the necessary information to use the code in these applications, and several examples that demonstrate most of the code's capabilities are provided.

The process modeling code MODEL has been upgraded by replacing the GASP-IV simulation language with the new and more powerful simulation language SLAM. In addition to the capabilities for modeling continuous systems found in GASP-IV, SLAM provides a versatile "network" modeling capability. Network modeling makes it possible to model efficiently large, complex processes having many queues, bottlenecks, and breakdowns. We used this new capability extensively to model the pellet grinding and inspection line associated with the SAF facility being built at the Hanford Engineering Development Laboratory.

## PART 4. INTERNATIONAL SUPPORT

### I. ENRICHMENT PLANT SAFEGUARDS

(J. C. Pratt and H. H. Hsu, Q-2)

The enrichment plant safeguards project met a major milestone when HEU production was simulated in operating centrifuges. Los Alamos had primary responsibility for gamma-ray measurements with a high-resolution germanium detector. This enrichment plant safeguards strategy is the simple, straightforward concept of having the inspector monitor individual centrifuges with a high-resolution gamma-ray detector. Two weeks before the start of simulated HEU production, we agreed to assist a Union Carbide Corporation/Nuclear Division (UCC-ND) measurement using fixed sodium-iodide area monitors. Los Alamos provided the detectors, electronics, and data-acquisition system for that project. The equipment acquired four spectra simultaneously for 2 h, after which it recorded the accumulated spectra on disk, cleared the spectrum memory, and restarted the acquisition. The equipment functioned in a centrifuge plant environment for 2 months and obtained over 2500 spectra, which were analyzed jointly with UCC-ND, using automatic analysis programs written by Los Alamos.

The germanium-monitoring results were presented at an Enrichment Plant Safeguards project meeting in October at Los Alamos. The detector clearly exhibits the photopeak from  $^{235}\text{U}$  gamma rays for enrichment less than 20% with only a 1-min counting time. One such spectrum is shown in Fig. 35. The area-monitoring results were presented at that same meeting by UCC-ND; subsequently, the UCC-ND candidate strategy was changed to the Los Alamos strategy of monitoring the region near the centrifuges with a germanium detector.

The germanium-monitoring results also were presented at an Enrichment Plant Safeguards Review held at DOE Headquarters. The germanium-monitoring technique was recognized as very effective. While it is also the most intrusive of the five candidate strategies selected for further study, it does not appear to be a risk to technology. In addition, this technique was mentioned in both the October and November meetings as the reasonable follow-up measurement to resolve anomalies generated in less intrusive monitoring strategies.

Our next milestone is describing the strategy for the Enrichment Plant Safeguards Part IV Study.

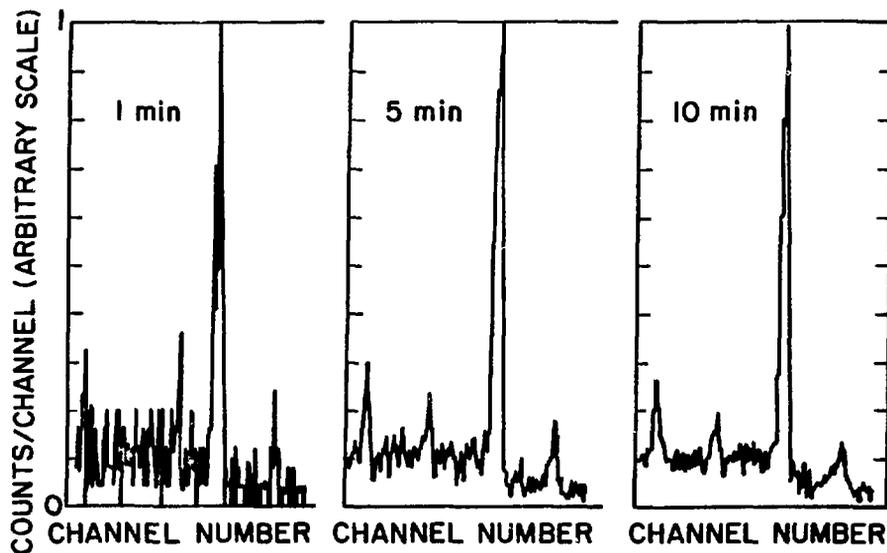


Fig. 35. Germanium spectra, acquired for 1, 5, and 10 min from a centrifuge containing 16%  $^{235}\text{U}$ .

## II. TECHNOLOGY DEVELOPMENT

### A. Cerenkov Spent-Fuel Instrument Upgrade (*N. Nicholson and E. J. Dowdy, Q-2; D. Holt, C. J. Stump, and D. Macy, EG&G*)

An instrument is being tested that will be used to monitor irradiated fuel assemblies stored underwater. The instrument, being developed for the IAEA, will allow an Agency inspector to view the Cerenkov glow emitted from the irradiated fuel and also will permit him to measure quantitatively the intensity of the glow. Earlier instrumentation supplied to the IAEA was capable only of viewing the Cerenkov glow and obtaining qualitative information. A Varo Noctron V Model 9878 Cerenkov viewing device (CVD) was selected as the basis for the new instrument, referred to as the improved Cerenkov viewing device (ICVD). The Varo has a generation-2 image intensifier of 25-mm diameter and is less expensive than the discontinued Javelin Model 222 that the IAEA inspectors now are using. The Varo Noctron V gives a high-resolution image, high gain, and very little barrel distortion. The add-on package that gives this CVD a photometric capability has been designed and fabricated, and the complete assembly is ready for preliminary testing and calibration. The assembly weighs 8 pounds, not including an auxiliary battery that powers the photometric electronics. A back-lighted LCD digital readout of the observed intensity is provided above the large biocular viewer. Figure 36 shows the Varo Noctron V CVD and the modified Varo Noctron V ICVD. In this figure, the objective lens of each instrument is at the center of the photo and is the smaller of the two lenses of each instrument.

The add-on package contains the necessary optical components to transport a fraction of the incoming light through a prismatic mirror onto an aperture, through a field lens, and then again onto the input of a photomultiplier tube (see Fig. 37). The straight-through light passes through the prismatic mirror and onto the photocathode of the 25-mm image intensifier. The electronics needed to provide the power and voltage for the tube and digitizing circuitry are in the add-on package. The external 12-V battery is contained in a shoulder bag with enough cable to allow the battery pack to be placed on the floor while the instrument is being used. The objective lenses are standard SLR-35-mm lenses made by Olympus (Zuiko) and include wide angle, normal, and telephoto.

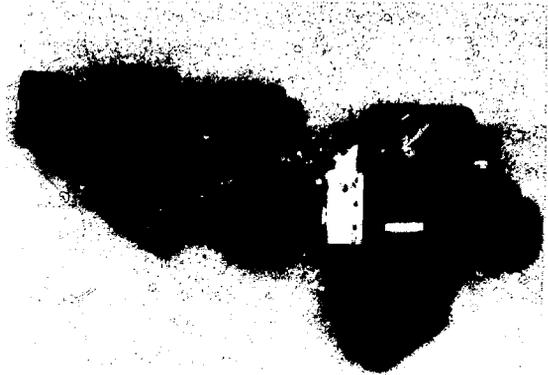


Fig. 36. CVD (left) and ICVD.

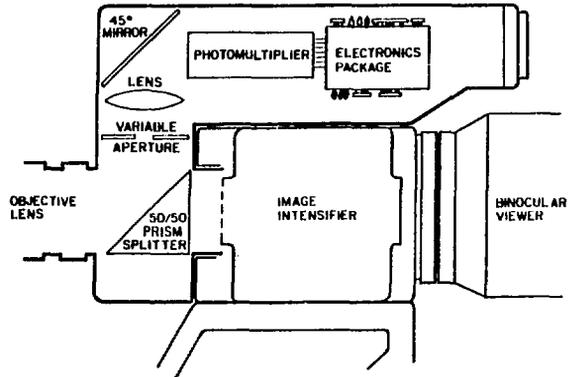


Fig. 37. Major components of the ICVD.

### B. A Cooling-Time Adjustment Procedure for Spent-Fuel Measurements with an Ionization Chamber (*P. M. Rinard, Q-1*)

The IAEA has found it useful to apply a simple cooling-time adjustment procedure on data from spent-fuel assemblies obtained with an ionization chamber.\* The goal of such an adjustment is to eliminate one of the parameters that affect the interpretation of data from irradiated assemblies, thereby improving the check on the operator's declared burnup values.

An ionization chamber's response to an assembly is divided by the declared exposure of that assembly. (The response is only a relative number, so experimental conditions must be reproduced when changing from one

\*This information was supplied by Elmır Dermenjević, IAEA, Vienna, 1981.

assembly to another within a set; different sets can be compared by normalizing the responses from one set to another.) We found that a set of such quotients is correlated with the cooling times of the assemblies by a simple power law,

$$R' = k T^b \quad (20)$$

where  $R'$  is the ionization chamber's response per unit of exposure,  $T$  is the cooling time, and  $k$  and  $b$  are the parameters to be determined from the data. The Los Alamos data shown in Fig. 38 are fitted well with  $b = -1.02$  if  $T$  is given in months. The range of exposures is from 20 000 to 35 000 MWd/tU. The value of  $k$  is not important here: it is the detector's response at a cooling time of one and hence is a relative number like the responses. The shape of the curve is controlled by the parameter  $b$  and is merely shifted up or down by changes in  $k$ .

To investigate this procedure from another starting point, doses in an ionization chamber near an irradiated assembly underwater were calculated using a technique previously reported.<sup>66</sup> Atom densities within the model assemblies were calculated by the Los Alamos Nuclear Data Group.<sup>67</sup> The gamma-ray transport into the ionization chamber a few centimeters from the assembly was then simulated in an approximate manner. The resulting doses were treated in the IAEA's manner. Figure 38 shows the calculated values and the experimental values already mentioned. Values of  $k$  were chosen so the two sets would be superimposed; this is the type of adjustment that would be necessary to compare sets of experimental data taken at different facilities or at the same facility on different occasions when the experimental conditions had changed. These calculations used a mixture of exposures with the same range as those of the fuel assemblies measured experimentally.

These preliminary calculational values form the same general curve as did the experimental data. The parameter  $b$  for the calculational data alone is  $-0.86$ , which is slightly more positive than that obtained for the experimental data. When assemblies with exposures from 20 000 to 40 000 MWd/tU or from 11 000 to 35 000 MWd/tU were included in the calculations, the agreement between the calculational and experimental data degraded.

With such an adjustment curve established for a set of assemblies, an ionization chamber response at cooling times other than at the time of measurement may be determined. All assemblies, for example, could be as-

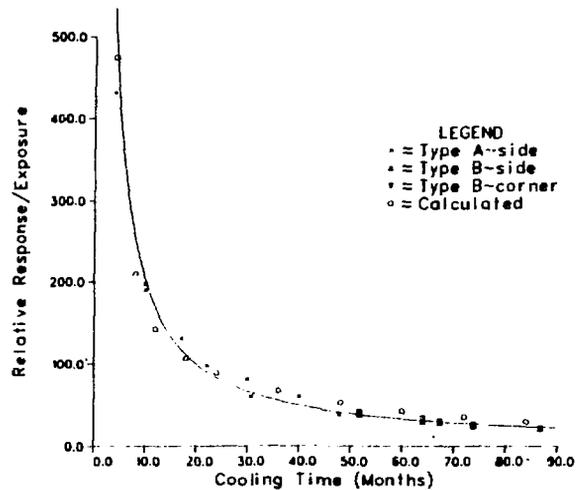


Fig. 38. The experimental data points were obtained from PWR assemblies with exposures between 20 000 and 35 000 MWd/tU. Each ion chamber response was divided by the operator's declared exposure for the assembly causing that response. The calculated values are for PWR fuel assemblies with the same range of exposures and with a detector 5 cm from the side of each assembly. The two sets of points are both relative numbers, so the ordinate scales have been adjusted to cause the two sets to superimpose. The smooth curve is a power law expression of the type given in Eq. (20), fitted to the experimental data  $b = -1.02$ .

signed a response at a common cooling time, and the relative dose rates would give a means of checking the consistency of the operator's declared exposures. A more comprehensive set of calculations of the type described here could indicate the sensitivities of the IAEA's adjustment process to parameters such as exposures, cooling times, initial enrichments, and ionization chamber positions. Eventually, an understanding of the factors that affect this adjustment could lead to more confident use of the process. When the experimental data available, for example, are too few to allow a reliable adjustment curve to be formed from the data alone, a calculated curve could provide some guidance.

### C. Independent Verification of Nuclear Materials Assays Made with In-Plant Instruments (C. R. Hatcher and P. A. Russo, Q-1)

1. Introduction. In verifying declared nuclear materials inventories at facilities under international safeguards, the IAEA has used two types of assays: destructive analysis of samples shipped to the Safeguards Analytical Laboratory (SAL) near Vienna and NDAs at the operating facility using portable equipment. To date,

the IAEA has made little use of data from assay instruments that are permanently installed in operating facilities. One advantage of the IAEA approach is that tampering with the measurements to conceal a diversion of material is extremely unlikely because the measurement methods remain completely under IAEA control.

Several factors have caused the IAEA to consider supplementing the present approach with in-plant instruments for nuclear materials assays. These factors include

- the anticipated need for more measurements and quicker results at large bulk processing facilities, such as reprocessing and enrichment plants;
- the advantages of instruments that are built into a facility, such as in-line instruments and glovebox installations;
- limitations in shipping samples to SAL, such as sample preparation, approval of shipping containers, sample evaporation, delays and costs; and
- limitations of portable NDA instruments for some applications, such as size, set-up time, maintenance difficulties, shipping of radioactive sources for active instruments, etc.

In this study we analyzed ways the IAEA might independently verify\* assays made with in-plant instruments that may be under the control of the facility operator, the IAEA, or a third party such as the state authority responsible for nuclear safeguards. The overall problem of independent verification of nuclear assays can be divided into three parts:

- verification of the sample.
- verification of the assay, and
- protection of the assay data.

In verifying the sample, the inspector must determine that the sample measured is representative of the material at the key measurement point, that no tampering with the material has occurred, and that the sample selected is the one measured.

Verification of the assay involves ascertaining that the assay value and measurement error recorded for each sample are correct. Data reduction to convert raw data to assay results is considered part of the measurement, because in many cases data reduction is performed by the assay instrument.

For small amounts of data, the simplest way to protect the assay data is for the inspector to keep a copy of the

verified assay data. Encoding and transmission of data to a secure memory have been recommended for protecting large amounts of data generated by on-line instruments.

All three aspects of independent verification are important considerations, whether the IAEA uses an in-plant assay instrument or an assay method completely under IAEA control. The IAEA has developed the general framework for the independent verification of nuclear materials inventories<sup>68</sup> and has much experience in the practical application of portable NDA instruments and in destructive analysis of samples shipped to SAL. Other investigators have studied procedures for verifying samples<sup>69,70</sup> as well as methods for protecting the assay data<sup>71</sup>. By comparison, relatively little has been written about independently verifying the actual measurements made with in-plant instruments.\*

**2. General Approach.** There are two fundamentally different methods that can be used to verify independently assays made with in-plant instruments. Method 1 (Fig. 39) uses a protected IAEA measurement technique to compare in-plant instrument results with IAEA results. Method 2 (Fig. 40) uses protected IAEA standards,\*\* known physical constants, and special test procedures to determine the performance characteristics of the in-plant instrument. These two methods can be used separately, or they can be used in combination, thereby providing redundancy.

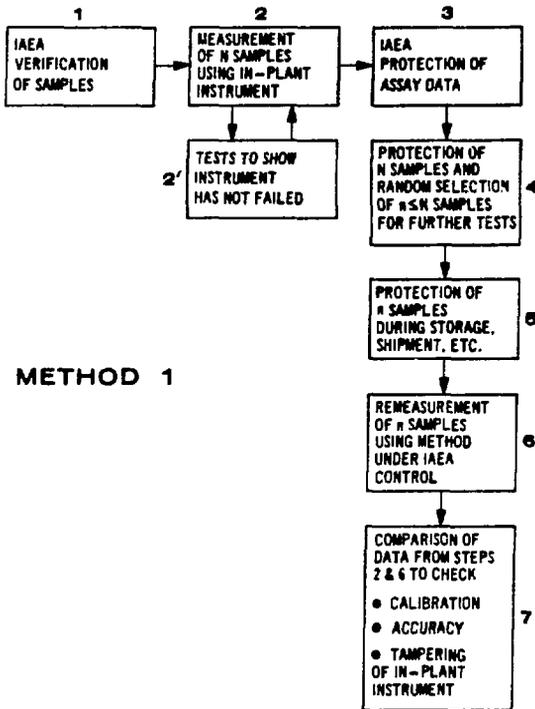
*a. Method 1.* Three basic steps (Fig. 39) are followed by an inspector when using data from an in-plant instrument: verification of the samples, measurement of N samples using the in-plant instrument, and protection of the assay data. To these steps, the inspector must add procedures for independently verifying the measurement.

Most in-plant instruments have associated with them a set of tests to show that the instrument has not failed (see Fig. 39, box 2'). The term generally used to describe these tests is *measurement control*. Some measurement control programs are designed to provide additional information, but the primary purpose is to establish that

\*To distinguish between "independent verification of measurements made with in-plant instruments" and "independent verification of a declared inventory," the IAEA recently has begun to refer to the former as "authentication of measurements made with in-plant instruments."

\*\*Our best source of information has been informal discussions with several technical experts, including Jim De Montmolin (SNL), Frank Houck (Arms Control and Disarmament Agency), Mike Smith (NRC), Don Cobb, Doug Reilly, Tom Canada, G. R. Keepin, and S. T. Hsue (Los Alamos National Laboratory), and Ron Augustson and Erwin Kuhn (IAEA).

\*\*Throughout this report, *standard* is used to refer to both well-characterized reference materials and to physical standards suitable for use in calibration procedures.



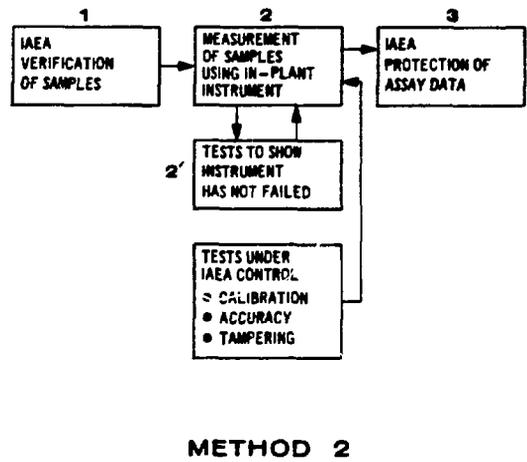
METHOD 1

Fig. 39. Possible procedure for IAEA verification of measurements made with in-plant instruments, based on remeasurement of some samples using a method under IAEA control.

the individual components of the instrument (hardware, software, and operator interface) all are working properly and have not changed characteristics or failed as a result of natural causes.

To verify independently measurements made with the in-plant instrument using Method 1, steps 4 through 7 in Fig. 39 are followed. After the IAEA inspector has received a copy of the assay data (or after the data have entered protected storage),  $n$  samples are randomly selected from the complete set of  $N$  for remeasurement. Samples must be protected by the IAEA during the random selection process and during any subsequent sample preparation, shipment, or storage. Next, the  $n$  samples are reassayed by a measurement technique under IAEA control, such as a portable NDA instrument or facilities at SAL. A comparison of assays made on the  $n$  samples by the two techniques allows the IAEA to establish the calibration and accuracy of the in-plant instrument during the measurement of the  $N$  samples, including any effects that possible tampering with the instrument may have had on calibration or accuracy.\*

\*The term *accuracy* is used here to include all types of measurement errors, such as biases, drifts, and random errors.



METHOD 2

Fig. 40. Possible procedure for IAEA verification of measurements made with in-plant instruments, based on IAEA tests of in-plant instrument characteristics.

Although measurement control (step 2') is redundant with steps 4 through 7, it serves the valuable function of quickly eliminating the cause of most erroneous assays. In an earlier paper,<sup>72</sup> it was shown that the probability of erroneous assays caused by instrument failures usually is much higher than the probability of erroneous assays caused by instrument tampering, and that we can take advantage of the two probability distributions in planning strategies for independent verification. Thus, for efficiency we should plan frequent tests that will detect most instrument failures and less frequent tests that will detect all types of measurement errors, including those caused by tampering.

*b. Method 2.* Figure 40 shows that Method 2 is similar to Method 1 in that the same three basic steps are followed by the inspector and the same kind of measurement control program is used to establish that the instrument has not failed. However, in Method 2, no samples are reassayed using a measurement technique under IAEA control. Instead, in-plant instrument calibration, accuracy, and freedom from tampering are established by working directly with the in-plant instrument.

The greatest problem with Method 2 is in showing that the in-plant instrument is free of sophisticated tampering. Rather than attacking this most difficult part first, it is easier to take the following approach. First, determine calibration and accuracy of the in-plant instrument, assuming that there is no tampering with the instrument; then show that there is no tampering.

Calibration of many types of in-plant instruments can be best accomplished by the use of standards. Because the standards must be protected from possible tampering, they should be kept in IAEA custody or verified by the IAEA when they are used. In some cases, the standards can be transported by the inspectors or kept at the facility under IAEA seal. In other cases, it might be necessary to calibrate the instrument off-site before its installation or to ship an instrument off-site for recalibration. The shipping of radioactive materials (such as enriched uranium and plutonium) under IAEA custody presents logistical problems that depend on the specific countries in question. Therefore, calibration of in-plant instruments on-site using IAEA standards is not always feasible.

A few types of instruments can be calibrated on the basis of known physical constants, using the so-called intrinsic calibration approach. Gamma-ray instruments that measure ratios of gamma-ray intensities typically fall into this category and have the advantage that standards are not needed to establish their calibration independently.

Verifying the accuracy of in-plant instruments is easily accomplished if a range of appropriate standards is available. Even when standards are not available, an estimate of precision can be obtained by repeating measurements on process samples.

Tampering with an instrument can take one of the following forms:

- (1) disabling the instrument, perhaps at a crucial time;
- (2) increasing random error by introducing noise or drifts;
- (3) introducing a fixed change in calibration by tampering with geometry, counting efficiency, etc; and
- (4) varying instrument performance in real time, for example, by using a "button under the table" to change data analysis procedures.

The first three types of tampering will be detected as part of the measurement control program or as part of the tests to establish calibration and accuracy. The fourth type of tampering, when carried out with sophistication, cannot easily be detected without specially designed instruments and procedures. For instance, the in-plant instrument may be made to give correct assays when standards are measured and to give assays that are 10% high when unknown samples are measured.

A number of approaches (other than Method 1) have been evaluated for detecting sophisticated tampering with in-plant instruments, including

- protected standards,
- blind samples,
- blind standards,
- add-a-gram,
- parallel instruments,
- internal consistency of data,
- containment and surveillance,
- inspection, and
- replacement of key components.

Each of these techniques is discussed below.

Protected standards can be used to detect changes in calibration or random error, but they cannot detect tampering if the instrument is made to give correct assays for standards and incorrect assays for process samples.

Blind samples involve concealing the identity of process samples during sample measurement and/or remeasurement. This method is useful for determining measurement precision, but it cannot detect falsified assays that are internally consistent.

Blind standards involve concealing the identity of all items measured, so it is not known whether a standard or a process sample is being measured until after the assay is completed. This approach is appealing in concept, but difficult to implement in most practical situations. The chief problem is ensuring that some covert method is not being used to determine when a standard is being measured.

Add-a-gram first assays an unknown sample, then assays the sum of the unknown sample and a standard. If the instrument is made to read in error by a constant fraction (for example, 10%), this method may detect inconsistencies between the two assays. However, if the instrument is made to read in error by a fixed bias (for example, 10 g), no inconsistency between the two assays will be apparent.

Parallel instruments means that two or more unprotected instruments gather data that can be tested for consistency. One assumes the unlikelihood that all instruments will be tampered with, hence that tampering will produce detectable inconsistencies. Although this method provides a level of assurance that increases with the number of parallel instruments, it can be defeated by tampering with all the instruments.

Internal consistency of data from a single instrument is used in measurement control programs to help establish that an instrument has not failed or changed characteristics. For example, gamma-ray peak ratios or neutron-singles-to-coincidence ratios can be checked and

demanded to fall within prescribed ranges. For instruments that produce many channels of data, this technique is of value in detecting tampering, but the technique can be defeated by tampering with all the data channels.

Containment and surveillance in this application are most likely to take the form of seals on parts or all of the in-plant instrument. For large instrument arrays, surveillance methods may be more appropriate. This approach is effective, but in some instances it may interfere with normal use of the instrument.

Inspection of the instrument by the IAEA inspector is a very effective way to detect tampering, particularly for simpler instruments. Software inspection can be achieved through techniques such as a software bit comparator.

Replacement of key components of the in-plant instrument with equivalent components under IAEA custody will prove simpler and more effective, in some cases, than inspection or containment and surveillance measures. For example, replacing software may be simpler than inspecting it or protecting it with seals.

Several techniques are available for detecting sophisticated tampering using Method 2, but the techniques that seem most practical are the use of seals, inspection, and replacement of key components. Blind standards and parallel instruments may also prove useful in special situations.

**D. Portable MCA (*J. Halbig, S. Klosterbuer, J. Caine, F. A. Duran, O. R. Holbrooks, D. L. Peterson, and M. Stephens, Q-1; J. Torres, Butler Service Group; and R. Whitehill, Missouri Research Labs*)**

The third portable MCA (mini-MCA) unit, which was fabricated and delivered to the IAEA in November 1981, contained improved ADC boards and an enhanced amplifier board. At moderate count rates, the resolution measured was 0.65 keV at 122 keV.

The two mini-MCAs that were delivered to the IAEA in May 1981 were returned from Vienna for exchange of improved ADC and amplifier boards. In addition, improvements were made in the CRT display boards and the power supply boards.

The mini-MCA does not supply  $\pm 24$  V because of power penalties incurred in generating these voltages. New detectors are available commercially that will operate on  $\pm 12$  V. A battery-powered auxiliary box was built that can supply preamplifier and bias power for HRG-type detectors. This allows the detectors that require  $\pm 24$  V of preamplifier power to be operated when used with the mini-MCA.

The software in the mini-MCAs was modified to use the new hardware boards. In addition, minor suggestions from the IAEA, such as setting the serial-port baud rate from the keyboard, have been implemented.

**E. Portable Neutron Coincidence Counter for Small Inventory Samples (*H. O. Menlove, Q-1*)**

The analysis of plutonium inventory samples by inspectors has been made increasingly difficult by transportation regulations. To reduce shipping requirements and to obtain more timely results, independent on-site verification capability is needed, particularly for the international inspection of reprocessing plants and plutonium facilities.

We have developed a high-efficiency, small neutron coincidence system for quantitative verification of the amount of plutonium in product inventory samples. Solutions withdrawn from a reprocessing plant can be assayed in the vials normally used to transfer samples to an analytical laboratory. Pellets and powders also can be assayed.

A diagram of the inventory sample counter is shown in Fig. 41. The detector contains 16  $^3\text{He}$  tubes (2.54 cm in diameter by 30 cm long) with a gas-fill pressure of 6 atm. The absolute detector efficiency is 40%, and the

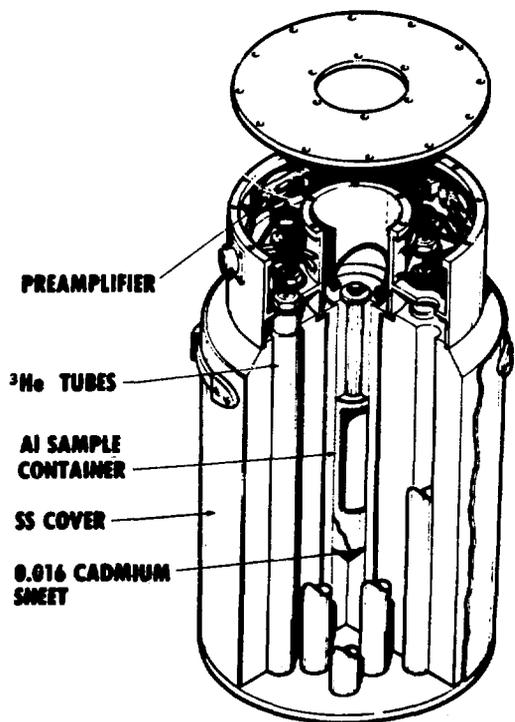


Fig. 41. Inventory sample coincidence counter.

sample cavity will accommodate samples up to 5.0 cm in diameter and 15 cm tall. The detector and moderator configuration has been designed to make the system relatively insensitive to plastic bagging and hydrogen content in the sample solution.

The unit has been designed to be operated with the high-level neutron coincidence counter electronics, and six preamplifiers have been placed inside the high-voltage junction box on top of the detector. The complete system is shown in Fig. 42.

Figure 43 shows the calculated measurement precision (1 standard deviation) for different sample types and measurement times. A measurement precision of 0.5% can be obtained for a 10-ml solution containing 250 g Pu/l (20%  $^{240}\text{Pu}$ ) in 200 s. Similar precisions can be

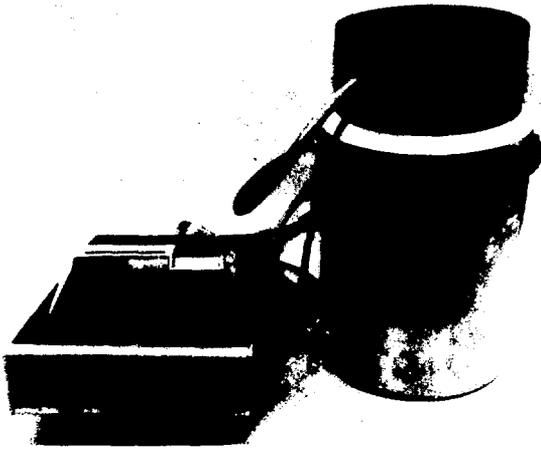


Fig. 42. Inventory sample coincidence counter showing the shift-register electronics and the HP-97 calculator.

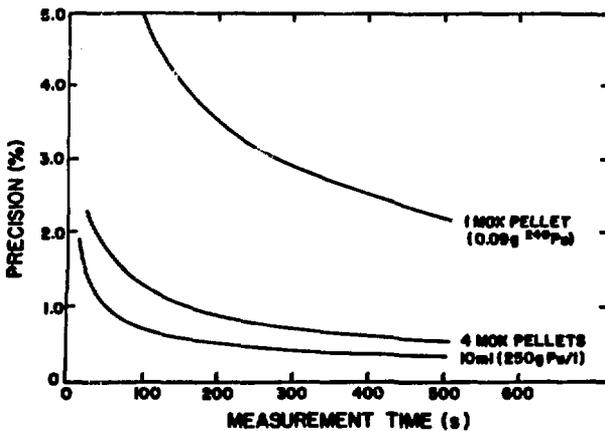


Fig. 43. Calculation of measurement precision (standard deviation) vs time for different types of plutonium samples.

obtained for MOX pellets and powders in the same mass range.

To study the response of the counter for small-mass plutonium samples, measurements were performed on single pellets of light-water-reactor (LWR) MOX fuel and on fast-breeder-reactor (FBR) pellets (see Fig. 44). The designation 4.9% plutonium corresponds to the fraction of Pu/MO<sub>2</sub>. The UO<sub>2</sub> in the pellets was natural enrichment (0.7%), except for the FBR pellets (21.6% Pu/MO<sub>2</sub>), which contained HEU (93%). A straight line fit the data, and the multiplication differences in the pellets were negligible. In addition to these pellet measurements, the counter will be evaluated for use with powders and plutonium-nitrate solutions.

#### F. Passive/Active Coincidence Collar for Total Plutonium Measurement in MOX Fuel Assemblies (H. O. Menlove, Q-1)

The active neutron Coincidence Collar<sup>73</sup> was designed for applications to LWR fuel assemblies that contain no plutonium. For the LWR fuel (UO<sub>2</sub>) the americium/lithium interrogation source is used to induce

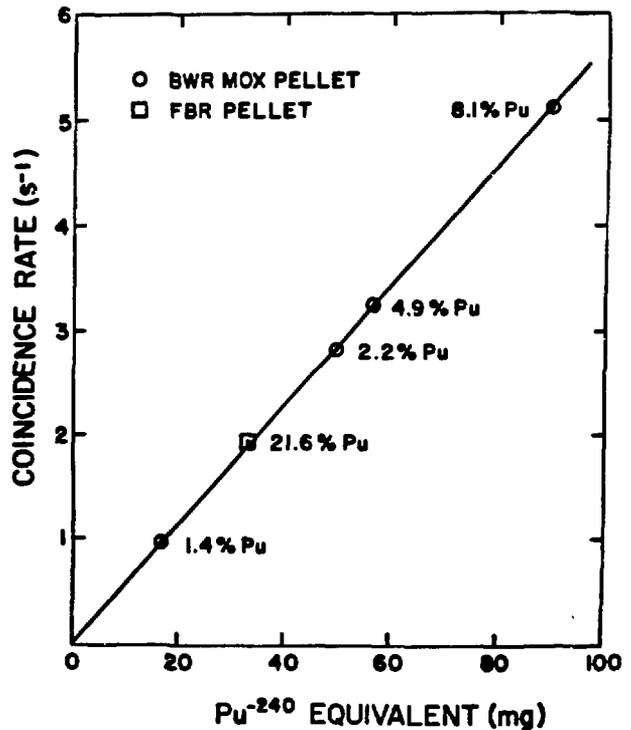


Fig. 44. Measurement results using the inventory sample coincidence counter for FBR and mixed-oxide fuel pellets.

fission reactions in the  $^{235}\text{U}$  in the fuel assembly, and the coincidence rate is proportional to the fissile content.

On the other hand, for MOX fuel assemblies, the plutonium in the fuel gives a passive neutron signal that can be used for the assay. The Coincidence Collar has been modified for FBR and LWR MOX fuel assembly assay by replacing the americium/lithium neutron source with a fourth detector bank to give symmetry to the detection efficiency. Figure 45 shows the modified unit called the Passive Coincidence Collar.

The normal passive-mode measurement gives the  $^{240}\text{Pu}$  (equivalent), but additional information, such as the plutonium isotopic ratios, is needed to determine the total plutonium content. The total plutonium content also may be obtained by combining the passive measurement with an active measurement of fissile content in the assembly.

The large quantity of  $\text{PuO}_2$  in a fuel assembly is a strong source (about  $10^6$  n/s) of fast neutrons (1 to 2 MeV). These neutrons originate from spontaneous fission and  $(\alpha, n)$  reactions, and in addition, there is

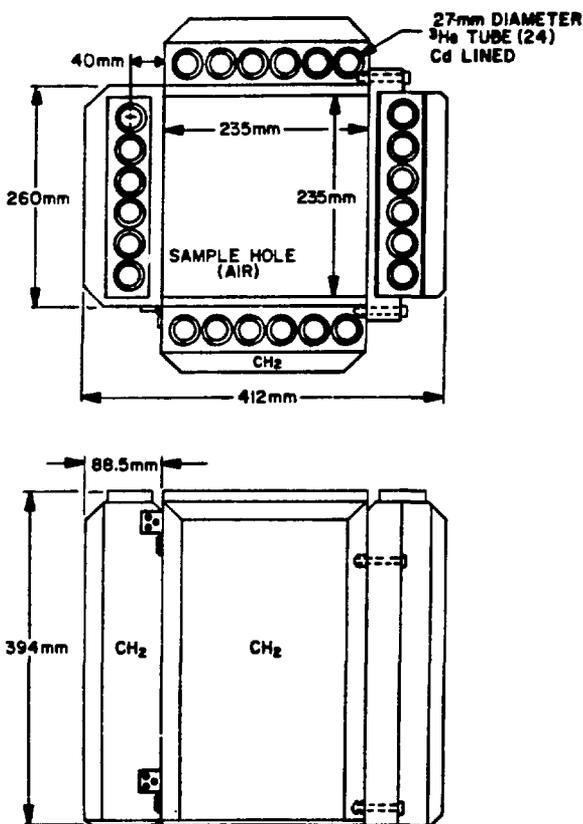


Fig. 45. The Passive Coincidence Collar for application to mixed-oxide fuel assemblies.

significant multiplication of the neutrons from induced fission reactions in the plutonium. These neutrons can be used for self-interrogation of the assembly by reflecting them back into the assembly with the body of the collar. To determine what fraction of the neutrons are from the primary neutron source and what fraction are from the reflection process, we must change the albedo or the reflection property of the boundary surrounding the assembly. By using the difference in the response between the reflective and nonreflective boundary conditions, the portion of the signal from the induced fissions in the fuel may be isolated from the reflected neutrons.

This general approach has been investigated previously by Close and Krick<sup>74</sup> for FBR subassemblies and Lee and Lindquist<sup>75</sup> for spent PWR fuel assemblies underwater. The present work has an important difference in that neutron coincidence counting is used rather than singles counting. This has the advantage of enhancing the induced fission fraction over the gross neutron background because the induced fissions have a higher effective neutron multiplicity. The coincidence counting also helps the response penetrability into the assembly center because the multiplication is higher in the center and is amplified by the coincidence counting. This is the same phenomenon observed<sup>73</sup> with the normal Coincidence Collar.

Figure 46 illustrates the fuel assembly surrounded by the Passive Coincidence Collar, with the reflected neutrons returning from the collar walls. The return of thermal neutrons into the assembly can be prevented by inserting a cadmium sheet (about 0.4 mm thick) between the wall and the assembly (see the dotted line in Fig. 46). This effectively reduces the multiplication or reactivity of the assembly-moderator combination.

Both the coincidence rate ( $R$ ) and totals rate ( $T$ ) are measured with and without the cadmium absorber. The normal passive-mode calibration curve corresponds to  $R$  vs  $^{240}\text{Pu}$  (equivalent), and it generally is necessary to make corrections for the multiplication from the fissile component. Various techniques have been used to make this correction, but the present cadmium subtraction determination gives a more direct measure of the fissile component and multiplication than have past procedures.<sup>76</sup>

The induced fission rate from the reflected neutrons is related to the quantity

$$R(\text{no Cd}) - R(\text{Cd}) \equiv \Delta R \quad ; \quad (21)$$

however, the value of the induced coincidence counts is also proportional to the neutron source strength, which is

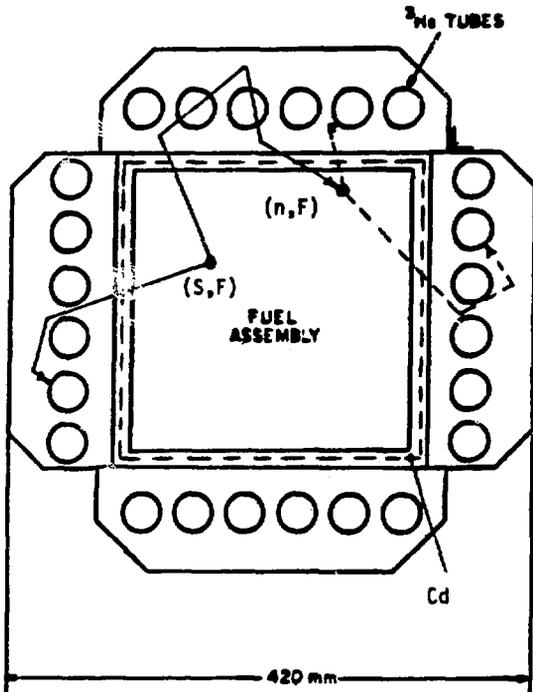


Fig. 46. The neutron reflection method for a mixed-oxide fuel assembly inside the Passive Coincidence Collar.

different for each subassembly. To normalize the source strength out of the response function, divide by T to obtain the quantity

$$\frac{\Delta R}{T} \quad (22)$$

which is proportional to the fissile content independent of the source strength through a calibration curve.

To test this concept, a series of measurements were performed using a mockup PWR fuel assembly with removable rods. The characteristics of the fuel assembly are given in Table XV.

For the experiments, a  $^{252}\text{Cf}$  neutron source ( $2.3 \times 10^4$  n/s) was placed in the center of the 15-by-15-rod PWR mockup assembly. We must introduce the californium neutron source to simulate the  $\text{PuO}_2$  source because the  $\text{UO}_2$  has such a low intrinsic neutron yield. The assembly contained 204 fuel rods and had 21 control-pin channels. Fuel rods were systematically removed from the assembly to change the uranium content by a maximum of 42 rods, as given in Table XVI. Both T and R were measured for each rod-loading configuration, with and without the cadmium liner. The results of the response ratio  $\Delta R/T$  vs uranium content are shown in Fig. 47. Replacing the  $^{252}\text{Cf}$  source by one that was five times more intense did not change the value of the ratio  $\Delta R/T$ , showing the effectiveness of the source-flux normalization procedure.

Fissions from the reflected neutrons are dominated by thermal neutron fission reactions, and normally this results in the problem of shallow penetrability of the interrogation. However, for the high-mass samples, such as LWR or FBR fuel assemblies, there is sufficient multiplication to propagate surface fission reactions into the sample's interior. Neutron coincidence counting is important because it amplifies the multiplication response. For example, a normal spontaneous fission event has a  $\nu$  (average number of neutrons per fission) of about 2.2, whereas the induced fissions from reflected

TABLE XV. Mockup PWR Fuel Rod Characteristics

Array size	15 by 15
Number of rods	204 (21 open channels)
Rod diameter (OD)	10.8 mm
Rod cladding	Zircaloy-2
Uranium enrichment	3.19%
Linear $^{235}\text{U}$ loading (assembly)	38.76 g $^{235}\text{U}/\text{cm}$
$\text{UO}_2$ active length	1.035 m
$\text{UO}_2$ density	10.48 g/cm <sup>3</sup>

TABLE XVI. Results of Passive/Active Measurements Using PWR Mockup Fuel Assembly

Number of Rods Removed	Totals/s T(Cd)	T(no Cd) - T(Cd) ( $\Delta T$ )	R(no Cd) - R(Cd) ( $\Delta R$ )	$ \Delta R/T(\text{Cd})  \times 100$
0	3104	658	161.5	5.20
12	3060	628	149.2	4.88
22	3019	587	135.6	4.49
32	2950	563	117.0	3.99
42	2915	531	109.6	3.76

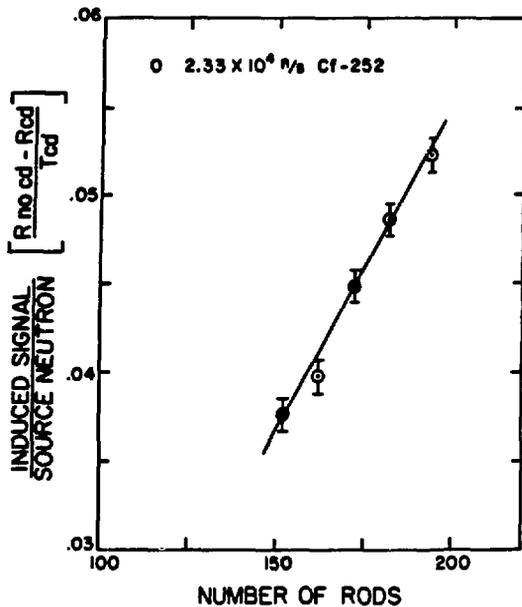


Fig. 47. Measured coincidence response per source neutron  $[R(\text{no Cd}) - R(\text{Cd})]/T(\text{Cd})$  vs fissile content in the PWR mockup assembly using a  $^{252}\text{Cf}$  driver source.

spontaneous fission neutrons have an effective  $\nu$  of about 3.6 ( $2.2 - 1 + 2.4$ ) for the coincidence time gate (32 to 64  $\mu\text{s}$ ). The coincidence counting rate is much more sensitive to the average value of  $\nu$  than is the totals rate.

### III. TECHNICAL EXCHANGES

#### A. International Working Group on Reprocessing Plant Safeguards (IWG-RPS) (E. A. Hakkila and J. P. Shipley, Q-4)

The final planning session of the IWG-RPS was held in Vienna, September 28—October 2, 1981. The IWG-RPS, which has been in existence for 3 yr, has the following objectives:

- to assist the IAEA in conducting a comprehensive study of safeguards systems and techniques for reprocessing facilities,
- to coordinate the R&D related to the comprehensive study in member states and the Agency, and
- to report on the results of the comprehensive study.

The IWG-RPS work was reviewed primarily in two topical reports and four subgroup reports. Los Alamos prepared and distributed one of the topical reports and

To check the penetrability of the technique, a group of 10 rods was removed from the perimeter and  $\Delta R/T$  was measured. These rods then were replaced, and 10 rods were removed from the central section of the assembly. The values of  $\Delta R/T$  were the same within the counting statistics of a few per cent.

In summary, this method gives the reactivity of the fuel assembly, which is primarily a function of the fissile content for a given sample-moderator configuration such as a fuel assembly in the collar. For FBR subassemblies, the combination of the active fissile determination with the passive mode  $^{240}\text{Pu} + ^{242}\text{Pu} + ^{239}\text{Pu}$  measurement gives a more complete verification of the total plutonium content.

This general technique can be applied to any high-mass sample of  $\text{PuO}_2$  and  $\text{UF}_6$  as well as to MOX and spent-fuel assemblies. The primary requirements are that the sample have a self-source of neutrons, a well-defined geometry, and a reasonably large geometric cross section or solid angle for the reflected neutrons to hit the sample. An evaluation of the passive/active technique will be performed during the next year for different types of FBR and LWR-MOX fuel assemblies. This technique is promising for FBR fuel assemblies because the total plutonium content can be verified both before and after irradiation in the reactor.

provided major input to two subgroup reports and to the final comprehensive study report.

#### B. IAEA Expert Consultants' Meeting on Nuclear Facility Design Assisting the Implementation of IAEA Safeguards (E. A. Hakkila, Q-4)

Los Alamos personnel participated in an IAEA Expert Consultants' Meeting on Nuclear Facility Design Assisting the Implementation of IAEA Safeguards held at IAEA headquarters, December 14-18, 1981. Incorporating features during the facility design stage is important to permit improved safeguards activities in conventional accounting and NRTA and containment and surveillance. This meeting addressed general design principles and design features for LWRs and fuel-reprocessing plants. Draft material for two of the seven chapters discussed at the meeting was prepared at Los Alamos.

### C. IAEA Consultants' Working Group on NRTA (*J. P. Shipley, Q-4*)

Los Alamos personnel participated in an IAEA Consultants' Working Group meeting on the Status of Near-Real-Time Accounting held at IAEA headquarters, January 25-29, 1982. Further studies of the principles and status, and additional field tests and demonstrations, of NRTA as it applies to international safeguards were recommended by the IWG-RPS. A major portion of the material discussed at this meeting was prepared at Los Alamos.

### D. Joint US-Federal Republic of Germany (FRG) Program (*E. A. Kern and J. P. Shipley, Q-4*)

As part of the US-FRG Bilateral Safeguards Program, one member of the safeguards staff from KFK-Karlsruhe spent 2 wk at Los Alamos studying modeling and simulation techniques for nuclear facilities. A second member of the KFK safeguards staff arrived for an extended visit to study, with Los Alamos staff members, statistical treatment of safeguards data.

Work performed by Dieter Sellinschegg, a visiting staff member from KFK-Karlsruhe, on the innovations algorithm as a decision analysis tool suggested that instrument recalibration for transfer measurements was undesirable. The innovations algorithm was studied further using Monte Carlo simulation to determine the effect of different diversion and recalibration scenarios.

The innovations technique was evaluated using a simple process model suggested by Sellinschegg. False-alarm and detection probabilities were computed by using Monte Carlo simulations. Each run consisted of 200 materials balances. With no recalibrations of the transfer measurements, the innovations method had much higher probabilities of detection and a much lower false-alarm probability than could be achieved with a standard CUSUM test. However, these higher detection probabilities are attained only when the diversions are delayed until at least the 20th materials balance. The longer the diversion is delayed, the more sensitive the innovations method becomes. Therefore, it is unnecessary to consider all possible subsequences as is done when generating alarm charts.

On the negative side, we found that the innovations method gives unacceptable results when recalibrations are used. With a recalibration interval of 10 materials balances, the CUSUM yields a higher probability of detection than does the innovations algorithm without

recalibration. When the recalibration interval is increased to 20 materials balances, the innovations method without recalibration is more sensitive than is the CUSUM with recalibration.

The innovations method looks for a change in the current state of the system, thus making it impossible to distinguish between a diversion, a recalibration, or any other disturbance to the system. In addition, the innovations method gives no information concerning the amount of diverted material.

In summary, with no recalibrations, the innovations algorithm appears to offer some significant advantages over our current decision analysis tools with regard to detection sensitivity and false-alarm probability. Poor results are obtained when the diversion begins early in the simulation or when the measurements are recalibrated. The innovations algorithm has some advantages over our current alarm-chart analysis with regards to computation requirements and interpretation of results.

### E. Technical Exchange with the European Community (*T. D. Reilly, Q-1*)

The DOE has entered into an Agreement of Cooperation with the Commission of the European Communities regarding a technical exchange program on nuclear safeguards. Negotiations on this agreement have gone on the last 2 yr; however, in late fall of 1981 the final text was agreed upon.

The agreement includes six tasks involving NDA instrumentation, seven tasks involving containment and surveillance equipment, and three tasks each in the areas of safeguards systems analysis and reference materials for nuclear material assay. Los Alamos is involved in many of these tasks with other DOE laboratories. The major European laboratory is the Joint Research Centre (JRC) in Ispra, Italy.

Los Alamos loaned a high-level neutron coincidence counter (HLNCC) to the JRC for testing and evaluation in one of the NDA tasks. This HLNCC was returned to Los Alamos in November 1981 after 1 yr. Two Los Alamos staff members went to Ispra at the start of this period to set up the instrument and instruct JRC personnel in its use. The HLNCC was tested at Ispra, at the EURATOM Safeguards Directorate headquarters in Luxembourg, and at a plutonium fuels fabrication plant near Rome, Italy. Personnel in the electronics division at Ispra studied the instrument from the point of view of integrating it into the safeguards data collection and

processing system that they are developing for the EURATOM safeguards inspectors. A final report on the work done with the HLNCC is being written by Ispra staff. The EURATOM Safeguards Directorate will acquire several of these devices for safeguards inspections conducted with IAEA inspectors.

Another NDA task involves the use of an active-well coincidence counter at a uranium fuels fabrication facility in Germany. A Los Alamos staff member visited Ispra in October 1981 to prepare the work plan for this experiment. Information also has been sent to facilitate the necessary approvals to bring the required neutron sources into the plant. Before these discussions, the traveler attended the semiannual meeting of the European Safeguards Research and Development Association (ESARDA) NDA working group in Luxembourg. This organization coordinates much of the work in safeguards technical development in the European community. Los Alamos has an official observer to this group, which greatly aids the technical exchange program.

Another Los Alamos staff member visited Ispra in November after a meeting at the IAEA in Vienna to discuss possible projects under the task pertaining to cooperation on the design of electronic instrumentation for safeguards measurements. One goal of this task is to achieve compatibility between instrumentation developed at the two laboratories, especially that to be used by the IAEA.

## F. Other Technical Exchanges (Q-4 Staff)

A senior member of the Los Alamos Safeguards staff visited Japan for a 3-wk series of discussions and seminars as the guest of the Japan Atomic Energy Research Institute (JAERI). The state of the art of near-real-time accountability, contactor inventory estimation, and modern multiple-period statistical data treatment and decision analysis were presented to representatives from Power Reactor and Nuclear Fuel Development Corporation, JAERI, Nuclear Materials Control Center, and the Japanese nuclear-fuel and utilities industry.

Two senior members of the Mitsubishi Metal Company were briefed on Los Alamos work in NRTA for reprocessing plants and on safeguards instrument development for reprocessing plants.

Two members of the Safeguards Systems staff organized and chaired invited sessions on the impact of IAEA safeguards on US nuclear facilities. One staff member presented an invited paper on safeguards systems design at this meeting.<sup>77</sup> A contributed paper describing isotopic ratios as a function of fuel burnup also was presented.<sup>78</sup>

A member of the Safeguards Systems Group participated as an invited lecturer in the Ispra course, "Mathematical and Statistical Methods in Nuclear Safeguards," and presented two papers.<sup>79,80</sup> Course attendees were from the European Economic Community and the IAEA.

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