LA-9110-PR
Progress Report

UC-15
Issued: April 1982

LA--9110-PR

FORM 615092

Program Status Report Safeguards and Security Research and Development

February—July 1981

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This work was supported by the US Department of Energy, Office of Safeguards and Security.

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PROGRAM STATUS REPORT SAFEGUARDS AND SECURITY RESEARCH AND DEVELOPMENT

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ABSTRACT

This report, one of a series of biannual progress reports, describes the status of research and development in the Safeguards and Security Program at Los Alamos from February—July 1981. Most work covered here is sponsored by the Office of Safeguards and Security of the Department of Energy; however, project activities that are technically closely related to nuclear safeguards and security also are included where appropriate for conveying information useful to the nuclear community. The report comprises four major subject areas: Security Development and Support; Nuclear Materials Measurement and Engineering; Nuclear Facility Safeguards Support; and International Safeguards, Technology Transfer, and Training. Some technical topics included in the subject areas are computer and informational security, chemical and nondestructive analysis of nuclear materials, process modeling and analysis, nuclear materials accounting systems, evaluation of prototype measurement instrumentation and procedures in nuclear facilities, design and consultation for facilities, technical exchange, training courses, and international safeguards.

PART I SECURITY DEVELOPMENT AND SUPPORT

I. INFORMATIONAL SECURITY

Weapons Life-Cycle Study (E. K. Tucker)

The Pre-Phase III Weapons Life-Cycle Vulnerability Study is nearly complete and results will be presented to key Department of Energy (DOE) personnel in August

1981. The final report will be combined with a counterpart Lawrence Livermore National Laboratory (LLNL) study in a single classified document to be issued jointly in early 1982. Data collection for the Post-Phase III Study has begun.

II. COMPUTER SECURITY

A. DOE Computer Security Technical Center (E. A. Springer and D. J. Bailey)

We have begun to develop the Computer Security Technical Center functions that support the DOE computer facilities. We are providing technical support to the Office of Security and Safeguards (OSS) Headquarters, as requested, and are monitoring computer security R & D projects in the Department of Defense and other agencies, specifying integrated computer security systems, conducting computer security R & D, and providing computer security education. The goal of the computer security R & D program is to develop computer security measures to preclude compromise of classified information.

The Computer Security Technical Center Long-Range Plan and Management Plan have been completed, as well as a computer security R & D task review for FY 80, FY 81, and FY 82. A TEMPEST (emanations security) effort has begun to prepare specifications for retrofitting the VT 100 and Data Media terminals. We have contracted with SYTEK, Inc. to develop a security policy model and with PE Systems, Inc. for further development of the key notarization system (KNS).

The Computer Security Group meeting in Idaho Falls, June 18-19, 1981, was successful, and many people expressed interest in participating in the Group's activities and in selecting R & D test sites. The first Computer Security Newsletter was published and distributed at the meeting.

B. Computer Network KNS (D. Brenner)

The KNS implementation at Los Alamos has been delayed because of problems in obtaining the intelligent terminals that will load the firmware on the erasable read-only memory (EROM).

We provided new KNS modules to PE Systems, Inc. to test at the National Bureau of Standards (NBS). The KNS modules were modified to work with the UNIX operating system at NBS. We will install the current version of the KNS on our PDP 11/34 before applying it to the VAX 11/750. In this way, we can foresee any problems with the software driver before taking on the more complex VAX system. Motorola, Inc. will deliver

the Q-bus security modules for testing, and PE Systems will design the terminal KNS.

Another major effort was restructuring the Unibus Key Notarization Facility assembly language firmware to make it more reliable and understandable. This firmware will be rewritten in PASCAL, a high-level language.

Interconnection of the stand-alone Unibus Key Notarization Facility with a host computer and development of the driver processes were postponed because of late delivery of the LSI-11/23 computer system. We now have the LSI system working, and we will implement the KNS before the end of FY 81.

C. Secure Operating System Testing and Implementation (W. Brainerd)

We are monitoring the kernalized secure operating system (KSOS)-11 effort at LOGICON, a Navy support contractor, but the use of KSOS-11 in DOE facilities seems doubtful because of problems with the UNIX emulator and the specific implementation on the PDP 11/70. The Honeywell Secure Communications Processor (SCOMP) results should be more useful. We are looking forward to testing SCOMP for the Nuclear Materials Management and Safeguards System at Oak Ridge National Laboratory.

Much of our recent effort was spent in obtaining the VAX 11/750 and LSI 11/23 intelligent terminals, which will provide a basis for testing the terminal KNS. We plan to bring the Berkeley UNIX operating system up on the VAX and use it as our security development system, and we will be working to develop software drivers to interface the various operating systems with the KNS.

D. Network Security Controller (D. D. Nessitt and R. Watson, LLNL)

All the hardware (except the chassis) has been obtained for the prototype cryptographic controller, which will perform system encryption functions for the distributed security controller. Each component system in the distributed operating system will have attached to it a cryptographic controller that will securely maintain the

appropriate encryption keys and will perform the encryption functions necessary for safe communication of data. The prototype system will be designed so it can be connected to a CRAY-1.

Test circuitry for the Fairchild 9414 DES chip set has been built, and the chip set is undergoing engineering testing. Because the set is not working properly, we are checking into the possibility of Fairchild's halting production of this 9414 chip. We now are exploring the possibility of using American Micro Devices AMZ8068 instead of the Fairchild 9414.

Work continues on developing a security model for distributed operating systems. A precise definition of the Domain of Trust concept has been formulated that will be the basis of further investigation. The capability protection mechanisms have been investigated in more detail, and studying several engineering questions has yielded some proposed solutions.

E. Computer Security Management Guidelines (A. Parziale and G. Corynin, LLNL)

Copies of a major year-end report were mailed to DOE reviewers. This report discusses a spectrum of risk-analysis issues ranging from graph theory development to the application of our present framework to a specific facility at LLNL.

Further progress was made on the completion of the risk assessment model, and the graph-theoretic foundation for the model structure has been completed. The next step is to add the required probabilistic machinery that will allow us to address probabilistic risk.

In the next fiscal year, we will attempt to convert our results into a deterministic risk-assessment tool. This is an important first step towards developing a practical risk-assessment tool for facility managers in the field.

PART 2

NUCLEAR MATERIALS MEASUREMENT AND ENGINEERING

I. CHEMICAL AND ISOTOPIC ANALYSIS

Safeguarding uranium, plutonium, and thorium requires measuring the amounts and isotopic distributions of these elements for 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 with complex chemical compositions, and many scrap and waste materials. Projects to meet these needs involve (1) developing fast dissolution techniques and analytical methods for determining plutonium and uranium, with emphasis on scrap and difficult-to-dissolve materials, and (2) designing and constructing automated apparatus for determining plutonium and uranium.

A. Development of Assay Methods

1. Microgram-Sensitive Spectrophotometric Determination of Uranium (S. F. Marsh, D. J. Hoard, and J. E. Rein). To develop a better method for determining uranium at microgram levels with the Los Alamos automated spectrophotometer, a reagent, 2, 2'-(2,6-pyridinediylbis (methyldinenitrilo)diphenol), re-

ported to give a selective spot test for uranium,¹ was synthesized and evaluated. Many organic solvents were investigated for extracting the U(VI) complex of this reagent; however, time instability of the color was common. Because reasonable stability was attained only in solvents that provided very low sensitivity, investigation of this reagent was discontinued.

2. Microgram-Sensitive Spectrophotometric Determination of Plutonium (N. M. Saponara, S. F. Marsh, and J. E. Rein). An extraction-spectrophotometric method for determining 1 to 20 µg of plutonium was developed for use with the Los Alamos automated spectrophotometer or with a similar instrument in which all operations from addition of reagents through the absorbance measurement are performed in a single tube. This method is based on extraction of the intensely colored Pu(IV)-chlorophosphonazo III (CPA) complex into n-pentanol from 1.5 M HCl. To enhance selectivity for application to various nuclear materials, a prior anion exchange resin separation of the plutonium in 12 M HCl is made. Absorbance of the extracted complex is measured at two different wavelengths, 680 and 710 nm, and the difference of these two values is proportional to

the plutonium concentration. The two-wavelength technique corrects for baseline shifts when using the single-beam automated spectrophotometer and results in improved precision.

The Pu(IV) CPA complex develops slowly from an initial 93% of maximum absorbance in 2 h to a maximum in 12 to 16 h. This is not critical for analyses in an automated instrument, where all operations are reproduced precisely, but for manual operation a 2 h color development is recommended.

The method without initial anion exchange resin separation tolerates many metals and nonmetals (see Table I). The anion exchange resin separation of plutonium enhances tolerance to metals (see Table II).

The relative standard deviation, based on six measurements each of eight plutonium quantities, is 1.5% or less over the range of 2.5 to 20 μg of plutonium. Plutonium recovery through the anion exchange resin separation is 98%, but concurrent similar treatments of the reference materials eliminate any biases.

3. Controlled Potential Coulometric Determination of Plutonium (D. D. Jackson, R. M. Hollen, and J. E. Rein). The method² developed for the Los Alamos, automated, controlled potential, coulometric analyzer has three sequential electrolyses in a 5.5 *M* HCl to 0.015 *M* sulfamic acid electrolyte that uses a platinum gauze

working electrode. Plutonium is reduced to Pu(III) at 0.25 V (relative to the saturated calomel electrode). Some diverse ions, including iron, are oxidized at 0.57 V; phosphate is added to lower the Pu(III)-Pu(IV) potential; and the Pu(III) is oxidized to Pu(IV) at 0.68 V as the measurement step.

Accurate control of the electrode potential in the automated analyzer permits deletion of the 0.25-V reduction step, thereby simplifying the procedure and decreasing the analysis time. Both Pu(VI) and Pu(IV) reduce rapidly to Pu(III) at 0.57 V in the 5.5 *M* HCl to 0.015 *M* sulfamic acid electrolyte. To ensure continued tolerances of impurities having oxidation-reduction potentials within the range of potentials in the methods, samples were prepared with known molar ratios of impurities relative to plutonium and were processed by the automated analyzer. The level of plutonium for each test was 0.02 mmol (5 mg). Interference again was defined as a change significant at the 95% confidence level relative to plutonium alone. No interference occurred at molar ratios of 2 for iron, relative to plutonium, and of 1 for iridium, molybdenum, neptunium, osmium, rhenium, ruthenium, antimony, selenium, thallium, and vanadium. Gold and platinum interfered by plating on the platinum electrode. The tolerances did not differ significantly from those measured for the original method. Tolerances for Br⁻, BrO₃⁻, ClO₄⁻, I⁻, IO₄⁻,

TABLE I. Tolerances of Metals and Nonmetals Without Anion Exchange Resin Separation

Mole Ratio of:					
1000:1		100:1	50:1	10:1	1:1
Cd(II)	Pt(IV)	Cu(II)	Fe(III)	Sb(III)	Cr(III)
Co(II)	Re(IV)	Mn(II)		Mn(VII)	Cr(VI)
Hg(II)	Se(IV)	Mo(VI)			
Pd(II)		Ru(III)			
Acetate	Iodate	V(V)			
Borate	Nitrate	Zn(II)			
Bromide	Nitrate	Sulfide			
Bromate	Perchlorate	Thiocyanate			
Chloride	Peroxide				
Citrate	Sulfamate				
EDTA	Sulfate				
Fluoride	Sulfite				
Formate	Tartrate				
Iodide	Thiosulfate				

TABLE II. Tolerances of Metals with Anion Exchange Resin Separation

Mole Ratio of Metal to Plutonium:		
100:1		10:1
Ce(III)	Fe(III)	Rh(III)
Cr(III)	Mn(VII)	
Cr(VI)	Sb(III)	
Ir(III)		

NO_2^- , SO_3^{2-} , and SO_4^{2-} also were not significantly changed, but we found that H_2O_2 can be tolerated at molar ratios no higher than 0.1 in either the original or modified method. Like most nonmetallic impurities, H_2O_2 can be removed by perchloric acid fuming before analysis. The tolerance for NO_2^- , which had not been measured in the original method, is 50 molar ratio.

The precision of less than 0.1% standard deviation for weight aliquots of a plutonium reference solution is not different from that of the original method.

4. Separation of Trace Metal Impurities from Uranium, Plutonium, and Americium (S. F. Marsh). For the difficult quantitative measurement of trace metallic elements in nuclear materials, a prior separation of the actinides is being evaluated using the highly selective dihexyl-N, N-diethylcarbamylmethylenephosphonate extractant (DHDECMP) for the actinides and lanthanides. The purified compound, coated on an inert support of porous polymer beads on a 6- by 100-mm chromatography column, extracts more than 99.99% of 15 mg of uranium, plutonium, or americium from 5.5 M HNO_3 . Nearly 40 metallic impurity elements elute completely for their analysis by emission spectrography or other multielement techniques. A talk describing the applications of DHDECMP extraction chromatography to nuclear analytical chemistry will be presented at the 25th Conference on Analytical Chemistry in Energy Technology in Gatlinburg, Tennessee, and a topical report describing the purification, preparation, and use of DHDECMP extraction chromatography columns is being prepared.

B. Development of Automated Analyzers

1. Automated Controlled-Potential-Coulometric Plutonium Analyzer (D. D. Jackson, R. M. Hollen, and F. H. Kelly). A Los Alamos report³ has been published that describes the automated analyzer in detail, including the chemistry of the analytical procedure; the mechanical, electrical, and programmable calculator components and their operation; and test data showing precision capability. Appendixes provide detailed descriptions of preoperational adjustments and tests, procedures for determining plutonium, mechanical components including construction drawings, electrical components including circuit diagrams, and system software.

A movie⁴ was prepared that describes the operation and application of the automated electroanalyzer for determining plutonium. A patent application was filed covering the method and apparatus developed for cleaning the cells and electrodes of the automated analyzer.

The control program for the automated analyzer was changed to use the modified, simpler method described in Sec. A.3. Several minor modifications to the instrument improved dependability. Additional electrical shielding was added and grounding was modified to decrease noise pickup. A small reed delay to reset the coulometer was installed at the output connector to prevent false reset by spurious noise picked up by the cable running to the scanner relay.

Construction of an automated controlled potential-coulometric analyzer for plutonium determinations using the modified method at the Savannah River Plant (SRP) is progressing satisfactorily. The major portions of the sample transport mechanism and reagent dispensers have been completed. Hardware and software for switching between the two required potentials have been developed.

2. Automated Complexometric Titrator (D. D. Jackson). Several instruments, including probe colorimeters and motorized burets, are being evaluated for possible use in construction of an automated complexometric titrator for determining microgram levels of uranium. A Hewlett Packard (HP) 85F desktop, programmable calculator has been selected as the controller. This relatively inexpensive calculator can control the mechanical and electrical operations of the automated complexometric titrator and process the measurement data. Interfacing design for the various instruments is under way.

C. Evaluation of the Chemistry Preceding the Mass Spectrometric Ion Exchange Bead Technique (S. F. Marsh, R. M. Abernathy, and J. E. Rein)

Of the ten chemical treatments evaluated for attaining isotopic equilibration of plutonium before its absorption on resin beads, the one consistently reliable treatment was reduction to Pu(III) with heated ferrous sulfamate, followed by oxidation to Pu(IV) with heated sodium nitrate. The results of this investigation were published.⁵

II. STANDARDS DEVELOPMENT

Both physical standards and calibration methods are essential to provide highly reliable chemical analysis data for nuclear materials accounting and quality assurance. The following activities help to achieve these objectives: (1) prepare well-characterized, plutonium-containing reference materials for use in the Safeguards Analytical Laboratory Evaluation (SALE) program, for distribution by NBS and the New Brunswick Laboratory (NBL), and for calibration of nondestructive analysis (NDA) equipment at Los Alamos; (2) participate in an inter-DOE laboratory program for measuring half-lives of the longer lived plutonium isotopes; (3) evaluate mass spectrometric techniques for International Atomic Energy Agency (IAEA) verification of plutonium and uranium contents of reprocessing plant dissolver solutions; and (4) characterize, chemically, special lots of nuclear materials as required by DOE.

A. Reference Materials (J. E. Rein and Staff)

A sixth lot of highly pure, highly enriched ^{239}Pu metal was characterized extensively, packaged as 0.5-g weighed portions in individually sealed glass ampoules containing dry argon at reduced pressure, and 500 portions were shipped, after careful visual inspection, to NBL, now designated by NBS as the distribution facility. Additional information was sent to NBS concerning preparation of the certificate for this reference material designated SRM 949, Lot F.

A total of 796 1-mg portions of the new ^{244}Pu reference material, SRM 996, to be used as an isotopic diluent in mass spectrometric analysis, also was shipped to NBL for distribution. The material was purified and packaged at Los Alamos and characterized by NBS, NBL, and Los Alamos.

Shipment of plutonium oxide for use both as bimonthly distributed SALE samples and as secondary reference materials for worldwide distribution was completed to NBL.

Reference materials were prepared, packaged, and delivered, as specified, for calibration and testing of various NDA instruments. The reference materials included the following.

- Five plutonium solutions in HCl with varying levels of americium were prepared.

- Three uranium metal disks were unpackaged, cleaned, weighed, measured, and repackaged.
- One PuO_2 solid was prepared and characterized for total plutonium and isotopic content.
- Seven plutonium solutions were prepared; characterized for plutonium content, isotopic composition, and impurities; and specially packaged.
- Five U_3O_8 samples analyzed for uranium, thorium, impurities, and uranium isotopic contents were encapsulated in metal containers.
- Nineteen U_3O_8 samples were characterized and encapsulated in polyethylene bottles or metal cans.

B. Plutonium Isotope Half-Life Measurements (J. E. Rein and Staff)

The goal of a DOE-sponsored interlaboratory program to establish accurate plutonium isotope half-life values has been largely completed with publications of half-lives of 24 119 yr for ^{239}Pu (Ref. 6) and of 14.38 yr for ^{241}Pu (Ref. 7) and submission of papers for publication describing the preparation and characterization of the ^{240}Pu material and its mass spectrometrically measured half-life of 6574 yr. The alpha particle counting determination of the half-life of ^{240}Pu yielded a value of 6575 to 6580 yr. Completion of this phase of the project and a paper describing the measurements are in progress.

C. Americium-241 Content of NBS 948 (T. E. Sampson)

We have suspected for some time that the ^{241}Am separation date of late 1963 or early 1964 for the plutonium isotopic standard NBS 948, given in private communications with NBS, was in error. Either the date was in error or the separation at that time was incomplete.

Calibration measurements performed for the SRP gamma-ray isotopics system have enabled us to determine the current value of the ^{241}Am content of NBS 948 with an estimated accuracy of 1.5 to 2%.

The 125.3-keV/129.3-keV $^{241}\text{Am}/^{239}\text{Pu}$ ratio was measured for a variety of samples. By using known or

standard values for the ^{241}Am content, a weighted least squares fit was done to adjust the two constants in the ratio expression to provide the best agreement between the measured and known values. Consistent calibration curves were obtained for 300 to 20 000 ppm of ^{241}Am , with two calibrations being carried out corresponding to assumed $^{241}\text{Pu}/^{237}\text{U}$ equilibrium and nonequilibrium conditions. The two calibration curves represent two methods of treating the relative efficiency extrapolation to 125 keV. True equilibrium samples may be analyzed by either method.

The known or standard values for the ^{241}Am content of the samples were determined in several ways. One method assumes zero ^{241}Am on a known separation date. Other values were obtained by routine chemical analyses. All values were decayed to the gamma-ray measurement date by using the ISOPOW code with the following half-lives:

Isotope	$T_{1/2}$ (yr)
^{238}Pu	87.79
^{239}Pu	24 082
^{240}Pu	6 537
^{241}Pu	14.35
^{242}Pu	376 300

Table III shows the sample, description, method of determining the standard ^{241}Am value, and agreement between the measured and standard values after calibration.

The calibrations seem to be determined with a precision of 1.5 to 2%.

Table IV also shows our measurement results for NBS 948. (All dates in Tables III and IV are shown as month/day/year.) Parts per million is relative to total plutonium. The value in Table IV is also within 1% or less of the average values reported in the European Safeguards Research and Development Association (ESARDA) interlaboratory comparison.⁸

D. Consensus Standards (D. B. Smith)

Subcommittee Institute of Nuclear Materials Management (INMM)-N15.9, chaired by a member of the Los Alamos Safeguards Staff, submitted two proposed standards to the American National Standards Institute (ANSI) for concurrent N15 balloting and ANSI review. These standards are proposed ANSI N15.35 entitled "American National Standard Guide to Preparing Calibration Material for Nondestructive Assay by Counting Passive Gamma Rays" and proposed ANSI N15.36 entitled "American National Standard Nondestructive Assay Measurement Control and Assurance." The first of these proposed standards received all affirmative ballots and should be issued soon for use by the nuclear industry. Balloting on the second proposed standard is not yet complete.

TABLE III. Americium-241 Calibration for Plutonium Isotopi System

Sample	Description	Determination of Std. Value of ^{241}Am	^{241}Am (ppm)	Gamma Meas. Date	Ratio: Measured/Standard (after calibration)	
					Equilibrium	Nonequilibrium
P2117	~2 g PuO_2	Assume 0, 2/3/79 at ion-exchange time	331.7	4/05/81	0.9832	1.005
CAL400	454 g PuO_2 , calorimetry exchange std	Certificate value, 63 ppm, 6/12/79	360.0	4/22/81	1.0200	1.0056
J1325A	500 g metal	CMB-1 value, 295 ppm on 11/5/79	478.7	4/15/81	1.0012	0.9890
P2118	~2 g PuO_2	Assume 0, 2/6/79 at ion-exchange time	756.0	4/8/81	0.9811	0.9885
P2119	~2 g PuO_2	Assume 0, 2/2/79 at ion-exchange time	1169.0	4/9/81	1.0053	1.0120
8GMETA	8 g metal	CMB-1 value, 189 ppm on 7/25/74	1474.0	4/17/81	1.0249	1.0181
P2116	~2 g PuO_2	Assume 0, 3/19/79 at ion-exchange time	4914.0	4/22/81	0.9987	1.0066
SAL639	~1 g PuO_2	CMB-1 value, 4100 ppm on 3/13/73	9689.0	4/23/81	0.9641	0.9691
P2120	~2 g PuO_2	CMB-1 value, 7430 ppm on 4/17/79	12 087	4/10/81	0.9883	0.9969
NBS946	~0.25 g Pu sulfate	Assume 0, 7/12/70	17 425	4/13/81	1.0264	1.0163
NBS947	~0.25 g Pu sulfate	Assume 0, 11/16/70	19 024	4/11/81	1.0021	0.9946
Mean					0.9996	0.9997
std dev					0.0195	0.0143

TABLE IV. Americium-241 Content of NBS 948
by Gamma-Ray Spectroscopy

NBS 948, measured ^{241}Am	4473 ppm, 4/19/81
Implied separation date	8/28/62
Americium 241 implied at isotopic certificate date	2908 ppm, 9/1/72

III. DEVELOPMENT OF NEW NDA TECHNIQUES

A. Materials in Process

1. *In Line Measurement of Plutonium and Americium in Mixed Solutions* (T. K. Li). During the past several years, the Plutonium Processing Facility at Los Alamos has prepared ceramic grade plutonium dioxide in production quantities by purifying impure plutonium metal to plutonium oxide.⁹ In this process, large quantities of americium and other impurities are removed from the plutonium as peroxide filtrates. Because of the increasing demand for americium in oil well logging instruments and other uses, routine operations to recover americium have been developed and implemented.¹⁰ Figure 1 shows partial flow diagrams for a plutonium purification process based on selective precipitation and an americium recovery and purification process based on ion exchange and oxalate precipitation.

Solutions generated from the plutonium purification and americium recovery processes have a wide range of total (plutonium and americium) concentrations and americium/plutonium ratios. For solutions generated from the plutonium purification process, the total concentrations range from 0.1 to 500 g/L and the americium/plutonium 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 americium/plutonium ratios vary from 0.5 to 100.

It is important to both special nuclear material (SNM) control and process control to determine the plutonium and americium content of these solutions accurately and rapidly. The assay procedure should be as simple as possible for routine operation by process technicians. An in line solution assay instrument (SAI) that satisfies these criteria has been designed and installed in the plutonium purification and americium recovery process area in the Los Alamos Plutonium Processing Facility. Solution

types, with total concentration and americium/plutonium ratios, are shown at the SAI measurement points in Fig. 1.

The SAI is based on high resolution gamma ray measurements. Because the plutonium in the purification process is of known isotopic composition, the total plutonium concentration of the solutions can be determined by using the 413.7 keV gamma rays from ^{239}Pu . The americium concentration is determined by a weighted average based on the measured intensities of the 376.6 keV (after removal of the contribution from ^{239}Pu at 375 keV) and 419 keV gamma rays from ^{241}Am . High intensity, low energy gamma rays (for example, 125 keV, 129 keV) are not used because the correction factors and uncertainties become very large; the transmission of low energy gamma rays decreases dramatically as the total concentration increases to 200 g/L. Furthermore, as either the $^{241}\text{Am}/^{239}\text{Pu}$ or $^{235}\text{U}/^{239}\text{Pu}$ ratio in the solutions becomes very high, the Compton continuum from the intense 208 keV gamma ray makes the lower energy unusable. To maximize the SAI's sensitivity and precision at both low and high concentrations for such high ratios, a 0.76 mm thick tungsten filter was installed in front of the detector. This filter reduces the lower energy portion of the gamma ray spectrum and provides adequate reduction of the 416 keV sum peak that results from pileup of the 208 keV gamma rays and interferes with the 413.7 keV ^{239}Pu peak. The transmission correction is made by counting gamma rays emitted from an external transmission source (a 20 g nickel plated plutonium metal disk) through the sample. Corrections for rate related counting losses as a result of deadtime and pileup are made by counting the 88 keV gamma rays from a ^{109}Cd source attached directly to the detector housing.

The gamma ray net peak areas are calculated by using a straight line background subtraction method. Both the

Fig. 1. Flow diagram of the plutonium purification process and the americium recovery and purification process. Solution types, with total (plutonium and americium) concentrations and americium/plutonium ratios, are shown at the SAI measurement points.

413.7 and 419 keV gamma rays are clean single peaks, whereas the 376.6-keV gamma ray is part of an unresolved doublet that includes the contribution from ^{239}Pu at 375 keV. The americium content determined from the unresolved doublet can be expressed as

$$\text{Am (g)} = \frac{I}{I(376.6)} \frac{R}{K} - \frac{I(375.0)}{I} \times {}^{239}\text{Pu (g)}.$$

where

$I(375.0)$ and $I(376.6)$ = specific activity of 375.0 and 376.6-keV gamma rays, respectively, (in $\gamma/\text{g/s}$);

$I = I(375.0) + I(376.6)$;

R = count rate of (375.0 + 376.6) unresolved peak (in counts/s) with the corrections of transmission and rate-related counting losses;

${}^{239}\text{Pu} = {}^{239}\text{Pu}$ content (in g) in the sample as determined by the intensity of 413.7 keV;

K = calibration constant (in counts/G/s); and

$$G = \frac{I(375.0)}{I} \times {}^{239}\text{Pu(g)} + \frac{I(376.6)}{I} + {}^{241}\text{Am(g)}.$$

The SAI, whose hardware, software, and assay procedure have been described elsewhere,¹¹ was calibrated with solution standards prepared by the Analytical Chemistry Laboratory at Los Alamos. Six plutonium standards, with concentrations ranging from 0.45 to 261 g Pu/l, were used for the original plutonium assay¹¹ was less than 0.75% for concentrations greater than 30 g Pu/l and a 1000-s count time and about 4% for 0.5 g Pu/l and a 2000-s count time. Four americium

standards, with concentrations ranging from 0.1 to 4.5 g Am/L, were used for the americium calibration. The precision of the americium analysis was less than 1% for concentrations greater than 4.5 g Am/L and about 4% for 0.3 g Am/L with a 2000 s count time.

The SAI performance in assaying plutonium process solutions¹¹ was evaluated by comparing results from its analyses with results from photometric titration, which has a routine accuracy of about 0.5%. Figure 2 presents the percentage differences between the SAI and the photometric titration methods plotted vs plutonium concentration. The americium plutonium ratios are 100 to 1.5×10^4 ppm for peroxide precipitation feed (closed circles) and 0.1 to 10 ppm for oxalate precipitation feed (open circles). The error bars represent the total SAI uncertainty resulting from uncertain counting statistics, calibration, sample weight, and isotopic abundance. The solid line shows an average relative difference of only 0.03%, which indicates no significant bias between the SAI method and the photometric titration method. The dashed lines show a standard deviation of 0.82%, which is consistent with the SAI precision in this plutonium concentration range.

The SAI accuracy for simultaneous analysis of americium and plutonium in mixed solutions from process samples was tested by comparing the SAI results with those of radioanalysis. Radioanalysis, which is used routinely for analyzing process samples, determines plutonium content by alpha counting after the plutonium

is extracted with 2 thenoyl trifluoroacetone and americium content¹² by integral counting the gamma activity above 32 keV. The routine accuracy of the radioanalysis is about 1 to 5% for americium and about 5 to 10% for plutonium. Preliminary results for 15 process samples are summarized in Table V. The radioanalysis results and their associated uncertainties are the means and mean deviations of two measurements, respectively. Although the plutonium concentrations were relatively low (less than 1.2 g Pu/L) and americium plutonium ratios were relatively high (from 0.6 to 34.9), the average relative difference between the SAI and radioanalysis results was 0.21%, which indicates no significant bias between them. However, a negative bias of about 5% was observed for the SAI americium concentration results relative to the radioanalysis results. A possible explanation for the higher radioanalysis results may be the contribution of ²³⁵U to the 59.5 keV gamma ray, which is commonly associated with ²⁴¹Am. On the other hand, to obtain the best results from americium from radioanalysis, the uranium must be separated from the americium. Investigation is now under way to compare the radioanalysis results obtained after uranium removal.

2. Assay of Low Level Plutonium Effluents (S. T. Hsue, F. Hsue, and D. F. Bowersox). In the Plutonium Processing Facility at Los Alamos, two processes are used to recover or purify scrap plutonium: the lean

Fig. 2. Comparison of results from the SAI and photometric titration methods for concentrated plutonium solutions: peroxide precipitation (americium/plutonium up to 0.5) and oxalate precipitation (americium/plutonium up to 10 ppm) feeds.

TABLE V. Comparison of SAI Results with Radioanalysis Results for Simultaneous Americium and Plutonium Measurements

Sample	Am Concentration (g/l)			Pu Concentration (g/l)			Am/Pu ^c
	RAD ^a	SAI	Δ^b	RAD	SAI	Δ	
1	0.90 $\pm$ 0.02	0.86 $\pm$ 0.02	-4.4	0.88 $\pm$ 0.02	0.92 $\pm$ 0.04	4.6	0.9
2	0.41	0.39 $\pm$ 0.02	-4.9	0.098 $\pm$ 0.004	0.116 $\pm$ 0.04	18.4	3.4
3	3.10 $\pm$ 0.07	3.06 $\pm$ 0.03	-1.3	0.68	0.65 $\pm$ 0.07	-4.4	4.7
4	5.15	4.87 $\pm$ 0.05	-5.4	0.97 $\pm$ 0.02	0.89 $\pm$ 0.09	-8.3	5.5
5	3.10	2.68 $\pm$ 0.03	-13.6	0.83 $\pm$ 0.04	0.90 $\pm$ 0.07	8.4	3.0
6	1.56 $\pm$ 0.06	1.54 $\pm$ 0.02	-1.3	0.068 $\pm$ 0.02	0.053 $\pm$ 0.05	-22.1	29.1
7	2.39	2.20 $\pm$ 0.03	-7.9	0.99 $\pm$ 0.04	1.14 $\pm$ 0.06	15.2	1.9
8	3.95	3.87 $\pm$ 0.04	-2.0	1.02 $\pm$ 0.05	1.03 $\pm$ 0.08	1.0	3.2
9	1.90 $\pm$ 0.07	1.85 $\pm$ 0.02	-2.6	0.057 $\pm$ 0.002	0.053 $\pm$ 0.058	-7.0	34.9
10	2.36	2.39 $\pm$ 0.04	1.3	0.67 $\pm$ 0.02	0.80 $\pm$ 0.07	19.4	3.0
11	3.81	3.73 $\pm$ 0.04	-2.1	1.15 $\pm$ 0.05	0.93 $\pm$ 0.08	-19.1	4.0
12	4.41	4.10 $\pm$ 0.04	-7.0	0.91 $\pm$ 0.04	0.70 $\pm$ 0.08	-23.1	5.9
13	3.43 $\pm$ 0.04	3.35 $\pm$ 0.04	-2.3	0.54 $\pm$ 0.02	0.535 $\pm$ 0.076	-0.9	6.3
14	3.25	3.00 $\pm$ 0.03	-7.7	0.66 $\pm$ 0.01	0.59 $\pm$ 0.07	-10.6	5.1
15	0.049	0.044 $\pm$ 0.01	-10.2	0.060 $\pm$ 0.01	0.079 $\pm$ 0.035	31.7	0.6

^aRadioanalysis results and their associated uncertainties are represented by the means and mean deviations, respectively, of two measurements. Mean deviations less than 1% are not listed

^b Δ (SAI - RAD)/RAD.

^cAccording to SAI results.

residue process and the dissolver solution ion exchange process. The typical effluent solution generated from these processes contains a low plutonium concentration (less than 100 mg/l) and a relatively high americium concentration (americium/plutonium less than 20). Because this waste solution is generated in quantity, timely assays of the plutonium content are important from the process control point of view, as well as for safeguards. Chemical analysis is too time-consuming; it requires extracting the plutonium from the americium¹³ and measuring the plutonium content by alpha spectrometry.

To measure this type of solution by NDA, we need signals that are more abundant than the passive gamma-ray activities emitted from plutonium. The gamma rays—for example, the 413.7-keV line of ²³⁹Pu—are useful for determining plutonium concentrations above 100 mg/l. Although alpha emission has higher specific activity than does gamma-ray emission, the alpha activity from americium in these solutions dominates, accounting for 80% or more of the alpha signal. One possible solution uses L x rays following the alpha decay.¹⁴ Because the L x rays from the decay of

plutonium and americium are different in energy, in principle they can be separated.

Figure 3 depicts an L x-ray spectrum from a plutonium-americium mixed solution with an americium/plutonium ratio of about 2000 ppm. The spectrum from an americium foil also is shown in Fig. 3. Plutonium and americium emit uranium and neptunium x rays, respectively, following the alpha decay. The uranium 17.22-keV L_{B1} x ray, as well as the 20.17-keV L_{γ1} x ray, can be used to monitor the plutonium concentration. A Si(Li) detector, with a resolution of 207 eV at 5.9 keV, was used. (Note that x rays generated from fluorescence do not have much effect because they are plutonium or americium L x rays and are energy separated from the uranium and neptunium x rays used in the measurement.)

In developing the passive L x-ray technique for quantitative assay of low-level plutonium solutions, two technical problems were encountered: sample self-attenuation and americium subtraction.

At low energies, that is, 17 to 20 keV and plutonium concentrations less than 1 g/l, the sample

Fig. 3. L x-ray spectrum from plutonium and americium between 11 and 23 KeV.

self-attenuation is dominated by the matrix concentration. By choosing a relatively thin sample (0.5 cm), the effect caused by matrix variation is minimized. At 18 keV, the correction factor (CF), as a result of sample self-attenuation, varies from $CF = 1.245$ to $CF = 1.282$, a change of less than 3% when the nitric acid normality varies from 2 to 7 N, respectively. Because the required accuracy for this effluent solution is about 20%, the self-attenuation variation is negligible.

The americium subtraction is more complicated. At 17.22 and 20.17 keV, the major peaks are the uranium L_{β_1} and L_{γ_1} x rays, but there also are contributions from the neptunium L x rays following the americium decay. When the americium/plutonium ratio is greater than or equal to 1, the neptunium L x rays can contribute a significant portion of the 17.22- and 20.17-keV peak areas. The neptunium contribution to the 17.22-keV peak is determined by the neptunium L_{β_1} peak at 17.75 keV; the contribution to the 20.17-keV peak is determined by the neptunium L_{γ_1} peak at 20.78 keV. The relative efficiency of the detector (including the relative sample self-attenuation) is determined by the neptunium L_{α_1} , L_{β_1} , and L_{γ_1} line intensities. The system dead-time and pileup correction are determined by counting the 5.9-keV line emitted by a ^{55}Fe source.

To investigate the plutonium assay by L x rays, we prepared a set of standard solutions containing 10 to 200 mg/l of plutonium with americium/plutonium ratios ranging from 60 to 0.15 ppm. The solutions, with their plutonium and americium concentrations known to 1%, were doubly contained in sample vials. Figure 4 shows the sample vials, the sample holder above the Si(Li) detector, and the shielding housing. Results indicate that the plutonium concentrations can be determined to 2% for this set of well characterized solutions.

Effluent solutions were drawn from actual processes in the plutonium recovery section, and measurements were made with the L x-ray detector. Duplicate samples were sent for chemical analysis, which was performed approximately 1 day later. The chemical analysis includes plutonium extraction,¹³ evaporation of plutonium solution onto a disk, and alpha counting. The americium concentration of the original solution is determined by integral counting of the signal above 30 keV. The claimed precision of the plutonium concentration determination is about 10%.

The L x-ray assay precision is dominated by the americium subtraction (therefore the americium/plutonium ratio) and to a lesser degree by the plutonium concentration. Figure 5 shows the percentage

Fig. 4. Inner and outer sample vials and the sample holder on top of the Si(Li) detector.

Fig. 5. Comparison of L x-ray and alpha counting as a function of plutonium concentration and as a function of americium/plutonium ratios.

deviation as a function of the plutonium concentration and also as a function of the americium/plutonium ratio. For plutonium concentrations less than 10 mg/L, the average deviation is 42%; for plutonium concentrations greater than 10 mg/L, the average deviation is 30%. In each case the deviation is greater than that obtained for the standard solutions. Possible causes for the larger deviations, which are being investigated, are

- isotopic variations between samples.
- higher americium/plutonium ratios than in the standard solutions, and
- postprecipitation in plutonium from the solutions.

An advantage of the L x-ray system is that by monitoring the 26.3 and 59.5 keV gamma ray peaks, the americium concentration in the solution (from 0.01 to 10 mg/L) can be determined accurately to a small percentage. In conclusion, the L x-ray system can provide a timely go/no-go measurement for deciding whether to recover the plutonium or send the solution to acid evaporation. As shown in Fig. 6, both the L x-ray and alpha counting arrived at the same conclusion for the 13 samples compared. The sensitivity of the L x-ray system is about 1 mg Pu/L.

Further developments are under way to improve the method's precision and also its capability to assay higher americium/plutonium ratio samples.

3. An Active Neutron Holdup Measurement Technique (N. Ensslin and E. Sanford). An active neutron technique suggested by the Safeguards Assay staff has been used to measure uranium held up on graphite surfaces and concrete walls. The technique uses

Fig. 6. Plutonium concentration by L x-ray counting vs plutonium concentration by alpha counting. The diagonal line designates perfect agreement between the two methods.

americium lithium neutron sources to induce fissions in ^{235}U . The induced-fission neutrons then are detected by ^4He gas proportional counters. Because the americium lithium sources emit neutrons whose energy is typically 0.3 to 0.5 MeV, whereas the fission neutrons are typically 1 to 2 MeV, it is possible, in principle, to detect the relatively small number of fission neutrons by setting an appropriate threshold.

Figure 7 shows the top of the source and detector holder used for the measurements described here. Two 5×10^4 n.s. americium lithium sources and six 18-atm ^4He tubes were mounted on this frame. Polyethylene shielding was used to reduce direct counting of source neutrons, and some polyethylene surfaces were covered with cadmium to maintain a harder interrogation spectrum. Also, a 5 cm thick aluminum reflector was added to increase the measured response by about 50%.

The data from this detector were collected and displayed on a multichannel analyzer (MCA). From the analyzer spectra, it was apparent that the americium lithium sources emitted some neutrons with energies as high as those from fission events. Also, both source and fission neutrons produced a spectrum of energy pulses that resulted from the folding of neutron energies with the distribution of recoil events in the ^4He gas. Thus, assays had to be based on a difference

Fig. 7. Top view of the active neutron detector for holdup in walls or other surfaces.

measurement between the energy spectra obtained with and without uranium. To determine those regions of the energy spectrum most sensitive to the presence of uranium, a series of U_3O_8 standards were assayed. The data were analyzed in the following three energy regions:

- medium energy (0.2 to 0.5 MeV),
- high energy (0.5 to 1.0 MeV), and
- high energy tail (0.8 to 1.0 MeV).

Figure 8 illustrates the results of these measurements in which the U_3O_8 standards were 30.5 cm (1 ft) from the detector. On the basis of these data, the high-energy region was selected for holdup measurements because it provided good counting statistics and used relatively high-energy neutrons.

Measurements of U_3O_8 standards with and without graphite matrix material (Fig. 8 is with graphite) demonstrated that moderating material increased the detector response but made the calibration nonlinear. Also, the reflection of source neutrons back into the detector (when it was next to a water tank or concrete wall) yielded a net "signal" equivalent to several hundred grams of ^{235}U . Another geometrical effect was the detector response as a function of distance from a localized uranium sample. This dependence was between

Fig. 8. Active neutron detector response as a function of ^{235}U content in U_3O_8 mixed with graphite.

$1/r^3$ and $1/r^4$, which limits useful detector response to distances of 60 cm (2 ft) or less. Therefore, the active neutron detector is very sensitive to geometry and matrix effects.

Consequently, the detector has been used for quantitative measurements only where the holdup geometry and matrix can be duplicated during calibration. This was possible for large, flat graphite surfaces and concrete walls. The response to uranium was determined by placing sheet standards between the detector and the surface. Even this procedure is not exactly correct because the response from uranium within the wall may be slightly different. In principle, the detector was sensitive to uranium at greater depths than can be reached with passive gamma-ray detectors because of the greater penetrability of neutrons. For the graphite and concrete surfaces actually assayed, this technique was comparable to the gamma-ray technique, suggesting that the uranium was held up near the surface.

4. Software for Enrichment Plant UF₆ Monitor (S. Bourret). Software for the UF₆ gas phase monitor, which will be installed and evaluated at the K 25 Oak Ridge Gaseous Diffusion Plant (ORGDP), has been written and implemented. The diagnostic program is designed to exercise all or most of the UF₆ monitor hardware. The main diagnostic program has an executive that lists a menu of diagnostics, monitors the keyboard, and dispatches control to a selected diagnostic. Eleven hardware areas that can be checked are the

- CRT character generator.
- CRT data display.
- clock.
- EROM.
- printer.
- front-panel LED display.
- front-panel key pad.
- source wheel.
- analog-to-digital converter.
- random-access memory (RAM), and
- central processing unit (CPU).

The menu lists a check for each area except the CPU. The check of the CPU and the serial link demonstrates that the program is running.

The UF₆ diagnostics program is contained in four 2532 EROM chips and occupies about 15 k bytes of memory. These memory chips are used and present only during diagnostics. The program is a combination of

FORTRAN and assembly language drivers and occupies memory locations 2000 to 5453.

A fifth EROM chip is present at all times. This program is an octal debugging one much like the on line debugging tool present with the LSI 11. This routine can load and verify a program and examine and change data in memory and registers. It has break points, can punch or dump a program, and can free run or trace through a program.

The UF₆ CRT display board with 6 k bytes of RAM has been designed, built, and programmed. A total of 2000 flicker free points can be displayed at once on the screen in one of seven different programmable intensities, and an on-board character generator can display 128 different characters. This board will be used to display the spectra.

B. Spent Fuel Assay

1. Spent Fuel Storage Facility Project (Los Alamos Safeguards Staff). An interim report was completed that reviews the potential role of nondestructive measurements on irradiated light water reactor (LWR) fuels at spent fuel storage facilities.¹⁵ Until recently, the scope of this study was to develop the conceptual design of a spent fuel nondestructive verification and assay system (VAS) for future away from-reactor (AFR) spent fuel storage facilities and to fabricate, test, and demonstrate a prototype VAS system. The design of a prototype VAS is about 80% complete, and some of the hardware already has been procured.

Nuclear materials accounting and control for safe guards and nonproliferation reasons and support of new spent-fuel management technologies require nondestructive measurements that span a wide range of fuel characteristics and fuel types. Several nondestructive techniques are already in an advanced stage of development, primarily through the efforts under this project and under the US program of technical assistance to the IAEA. This new measurement technology should be transferred to the nuclear industry.

Specific recommendations of this interim study include the following.

- The Cerenkov viewing device (CVD) and the neutron ring detector, both already developed for the IAEA, should be transferred to the domestic nuclear industry. These devices are portable, easy to use, and provide confirmatory evidence regarding the integrity and burnup of spent-fuel assemblies.

- The prototype spent fuel nondestructive VAS should be completed, and a program of field testing and demonstration should be carried out. This system would be the prototype for future in plant, nondestructive measurement systems at spent fuel storage and reprocessing facilities. A program of field testing and demonstrating the prototype is needed at an actual spent fuel storage or reprocessing facility before a final system can be considered ready for in plant use.
- The study of applying nondestructive measurements for spent fuel should be extended to explore the potential of these measurements in supporting new methods for fuel management and storage currently under development by the DOE, Electric Power Research Institute (EPRI), and the nuclear industry.

2. Calculated Neutron Source Spectra from Irradiated Pressurized Water Reactor (PWR) Fuel Assemblies (P. M. Rinard, G. E. Bosler, and J. R. Phillips). Neutrons from an irradiated fuel assembly provide an important signature that can be used to determine fuel parameters. Interpreting a detector's response must begin with a knowledge of the emission rate and energy spectrum of the neutrons produced within the assembly. These can depend on the initial enrichment, exposure history, and cooling time. The spectra presented here illustrate the effects of these three parameters. The additional concern of neutron transport to a detector is not considered at this time.

The important sources of neutrons are those nuclides that fission spontaneously and those that emit alpha particles for (α, n) reactions with oxygen in the fuel. The atom densities of these nuclides were obtained from calculations by the Nuclear Data Group at Los Alamos National Laboratory, using EPRI CELL¹⁶ and EPRI CINDER^{17,18} codes. The calculations were performed for a 15 × 15 fuel pin array of the PWR, characteristic of the H. B. Robinson plant (740 MWe). The initial fuel enrichment in the assembly modeled was 2.561%. The power level was held constant and the linear power density was 200 W/cm. The calculated atom densities are in good agreement with destructive analysis results on this type of fuel.

A Maxwellian shape was assumed adequate to describe a spontaneous fission spectrum, but two different "temperatures" were used. For plutonium and uranium nuclides, 1.2 MeV was used; but for curium nuclides, the temperature was increased to 1.5 MeV. These values were assigned after studying calculated spectra.¹⁹

Little information is available on (α, n) spectra for several alpha emitters of interest. Considering the best available data^{20,22} and calculated spectra,^{23,24} we concluded that a single spectrum can be used regardless of the energy of the alpha particle emitted. This spectrum fits the data, for the most part, and is augmented by calculated spectra at the lowest and highest energies.

With the atom densities as weighting factors, the two spontaneous fission curves and the single (α, n) curve were combined to generate the total neutron spectra from the fuel after irradiations and cooling times. An example of the exposure effect on the spectra is given in Fig. 9, in which the initial enrichment and cooling time are fixed. Near zero exposure, the spontaneous fission neutrons from ²³⁸U are almost the only ones present. After exposures between 750 and 10 000 MWd/tU, the (α, n) neutrons are nearly equal in number to those from spontaneous fission, and the curves almost show individual peaks from the two types of sources. After higher exposures, the ²⁴²Cm and ²⁴⁴Cm nuclides become the most important sources of neutrons, and the spectra become more nearly a purely spontaneous fission spectrum.

The effect of cooling time on the neutron spectrum is illustrated in Fig. 10, where the exposure level of the assembly is important. For example, after a low exposure, the long lived plutonium nuclides are the most prominent sources, and cooling time makes little difference. After high exposures, larger amounts of ²⁴²Cm are produced, and, because ²⁴²Cm has a 163 day half life, the cooling time affects the spectra markedly.

The effects of both exposure and cooling times are summarized in Fig. 11. The fraction of all the neutrons from spontaneous fission is shown as a surface and is never less than one half for this type of assembly. For exposures less than 11 000 MWd/tU, the spontaneous fission fraction increases with cooling time monotonically; for greater exposures, the fraction increases for 2 or 3 yr then decreases very slowly. This transition occurs when ²⁴⁴Cm begins to be produced in large amounts and grows in relative importance with cooling time because of its long half life (18.1 yr).

Calculations were made with higher initial enrichments (see Fig. 12). Note that the atom densities were calculated at a fixed power level; with a higher enrichment, the neutron fluence had to be lower, thereby producing less of the important curium. This accounts for the high enrichment curve lying lowest in the figure.

Near the work's conclusion, a Russian study²⁵ was published on the same topic; its results are similar to

Fig. 9. The neutron spectrum depends on the exposure given to the 256% enriched fuel. In these graphs, the enrichment and cooling time are not changed, and the exposure is increased. Also given are the fraction of the neutrons from spontaneous fission and the average energy of all neutrons.

Fig. 10. For each of five exposures, the effect of cooling time on the spectrum is shown.

Fig. 11. The fraction of neutrons from spontaneous fission is shown as a surface function of exposure (in GWd/t) and cooling time (in years). Notice that after an exposure below 11 GWd/tU, the fraction decreases with cooling time, above 11 GWd/tU, it increases.

Fig. 12. The source strength (but not the spectral shape) is strongly affected by the initial enrichment. Only one exposure is shown here, but the curves are typical of all exposures studied. Note that the linear power density was a constant during the atom density calculations, so the neutron flux was lowest for the highest enrichment case. This means that the 4.5% curve always corresponds to the weakest source strength in this figure.

those found here, but with some differences. Because they used the "harder" (α, n) spectrum and a "softer" spontaneous fission spectrum,²⁴ the two maxima in their total spectra appear more prominently over a wider range of exposures than shown in Fig. 9. We feel that the typical (α, n) spectrum and the Maxwellian temperatures used here are more appropriate; the (α, n) spectrum data are followed more closely, and the temperatures are for spontaneous rather than induced fission.

3. PWR Fuel Assembly Assay: Fissile Content Sensitivity Studies (G. W. Eccleston and M. P. Baker). Research and development efforts are in progress to design an assay system for direct nondestructive measurements of the exposure and residual fissile content of spent LWR fuel assemblies.^{15,26} This system, which will operate underwater in storage basins containing LWR fuel, will incorporate both gamma ray and neutron measurement instrumentation. Passive and active measurements will be used to collect neutron data. The active system will incorporate a californium neutron source to induce fissions in an assembly producing a measurable steady state neutron response.

Sensitivity measurements were conducted to determine the effects of matrix positions and small changes in fuel pin loading on an active neutron assay of a

water immersed PWR type fuel assembly. The 120 cm long fuel pins were loaded with 3.2%-enriched UO_2 and arranged in a 15×15 array (Fig. 13) comprising 204 fuel pin positions and 21 guide-tube locations. An underwater environment for the experiment was obtained by securing the fuel assembly in a 50 cm diam by 140 cm high cylindrical water-filled aluminum barrel. A 4 atm 3He detector (2.5 cm diam by 30 cm active length) wrapped in a watertight sleeve was immersed in a vertical position in the barrel and positioned at the midplane of the fuel. A small californium spontaneous fission neutron source (3.5×10^6 n/s) also was positioned at the fuel midplane in the water but on the assembly side opposite the neutron detector.

The sensitivity measurements consisted of comparing neutron count rates from the assembly before and after one of the 204 3.2% enriched UO_2 fuel pins was replaced by a pin loaded with natural UO_2 (0.7% enriched). This pin substitution represents a 0.38% decrease in the total ^{235}U loading of the fuel array. Data were collected as the pin substitution was made throughout the fuel array. Matrix assignments corresponding to fuel pin locations (Fig. 13) consist of column and row positions, each ranging from 1 to 15; for example, the fuel pin adjacent to the source is in column 8 of row 1, and the pin adjacent to the detector is in column 8 of row 15.

The count rate differences for fuel-pin-substitution measurements along column 7 of the PWR type assembly are shown in Fig. 14. The ^{252}Cf neutron source was positioned 4 cm from the assembly face, and the detector center was 3 cm from the assembly. An average

Fig. 13. Top view of the PWR 15×15 rod fuel assembly.

Fig. 14. Count rate differences resulting from substituting one 0.7% enriched fuel pin for a 2.3% enriched pin along fuel column 7.

count-rate difference (indicated by the dotted line in Fig. 14) of $0.93 \pm 0.17\%$ was observed. Almost all pin positions along a column produced the same count-rate difference, within statistics, indicating that the measurement sensitivity to small changes in the ^{235}U loading of a pin is independent of matrix location. Exceptions to this observation occur for fuel pins adjacent to the neutron source. These pins produced a significantly larger count-rate difference than the average, indicating an increased sensitivity to changes in ^{235}U loading. The same trend was observed in fuel columns 1 through 6; however, the average count-rate difference became less (compared to column 7) for columns further from the source and detector.

These measurements indicate that an unirradiated PWR fuel assembly can be tested underwater with an active neutron source measurement for a uniform fissile loading. Scanning measurements collected along the assembly length will show changes greater than 0.4% in uniform fissile loading.

4. PWR Fuel Assembly Assay: Modeling and Fuel-Pin Fission Calculations (G. W. Eccleston, G. E. Bosler, and R. G. Schrandt). Calculations also are being used in designing the neutron assay portion of the PWR fuel assay system. Computer modeling enables parametric studies and provides information not easily attainable through experimentation, such as fuel-pin fission densities.

For modeling LWR fuel assemblies, a lattice version of the Monte Carlo Neutron-Photon (MCNP)²⁷ transport code incorporating special tallies was developed. The general code is limited to a region of 175 cells, which is not sufficient to describe a fuel array. The new lattice capability, pointwise continuous cross sections, and extensive tally options enable detailed modeling and neutronics simulations of LWR fuel.

The lattice option allows a repeating geometry of parallelepipeds to be specified within the existing geometry, particle tracking, and tally treatments of the code. Ten different geometries can be specified to occupy the parallelepipeds, and each geometry can be placed at selected positions throughout the lattice array. The lattice option was used to specify an assay geometry incorporating a 15×15 PWR fuel assembly containing 204 fuel pins and 21 guide-tube locations, which is equivalent to a region specification of approximately 6500 cells. The lattice version was compared to the regular code where the lattice region was reduced to one large homogeneous cell. The lattice version required

about twice the computer time to track an equivalent number of histories. Approximately 85% of this time increase was due to particle tracking through the detailed cell and surface regions of the fuel array; the other 15% was due to overhead computing required by the lattice treatment.

Special tallies were developed and added to the lattice version of the MCNP code. These tallies are used to calculate

- fuel pin fission events by isotope and lattice location.
- detector responses according to the origin of the neutron (fissionable isotope or source) causing the response.
- detector responses tallied as a function of lattice location corresponding to multiplication or direct counts caused by sources internal to the lattice, and
- fission events produced in the fuel pins of a lattice by an external source.

The tallies are computed by using a track length estimate of the flux times the appropriate reaction cross section. This last tally has an option of tracking only neutrons produced by the external source, with the additional capability of terminating particles following the first fission. This feature enables quick runs that provide tallies of fuel-pin fission density resulting from an external neutron source.

Design of the active neutron assay portion of the spent-fuel measurement system is aided by calculations that show the effects of an external source in inducing fissions in the fuel. The MCNP code was used to calculate source neutron induced fissions in a 15×15 PWR fuel assembly immersed in water. The array was filled with 3.2%-enriched zircalloy clad UO_2 fuel pins. The neutron production from fission at pins throughout the fuel array is shown in Fig. 15 for a californium source located at a point 7.62 cm from the assembly edge. The top and bottom surfaces represent ^{235}U and ^{238}U fissions, respectively, throughout the fuel array. Note the large neutron production rate in fuel pins closest to the californium source.

5. Analysis of Spent-Fuel Specimen Data Obtained at the Battelle-Columbus Laboratories (BCL) (G. E. Bosler and J. R. Phillips). At BCL, Columbus, Ohio, fuel rods from spent-fuel assemblies are sectioned for eventual dissolution and destructive isotopic analysis. Before dissolution, these rod sections are available for NDA with instruments such as the High-Level Neutron Coincidence Counter (HLNCC). Neutron measurements on

Fig. 15. Calculated fissions in pins of 15×15 rod PWR fuel array per californium source neutron.

these sections, which include total counts from all neutrons emitted by the sample and coincidence counts from spontaneous fission neutrons, are valuable because direct correlations can be made between the neutron count rate data from the HLNCC and the actual isotopic composition determined by destructive means. The destructive analyses, which include most transuranic isotopes from uranium through curium and some fission products, also are valuable for comparison with calculationally estimated quantities.

The rod sectioning, isotopic analysis program is being performed at BCL as part of a joint Baltimore Gas and Electric Company, Combustion Engineering Company, and EPRI Demonstration Fuel Assembly Program. Through contractual agreements with BCL, Los Alamos is responsible for performing nondestructive neutron analysis on the samples. The particular fuel being analyzed is from a specially adapted PWR assembly at the Calvert Cliffs Reactor Facility. Under the specifications of the overall fuel evaluation program, this assembly was designed to be in the core through four cycles. At the end of each cycle, rods are removed from the assembly and replaced with new rods. The reconstituted assembly then was returned to the core for further irradiation, and in this manner rods from the same assembly could be examined at various stages of irradiation. In April 1981, the fourth cycle was com-

pleted, and in the near future neutron measurements will be made on rods with exposures from 40 to 50 GWd tU.

Unfortunately, Los Alamos involvement in this program did not begin until after rods were removed at the end of three cycles, and no neutron data are available for the first two cycles. For the three cycle rods, exposures ranged from 34 to 37 GWd tU. These rods had an initial ^{235}U enrichment of 2.45% and cooling times of 17 months. We have computed the neutron emission rates for typical PWR fuel assemblies using the EPRI CFFL¹⁶ and EPRI CINDER^{17,18} codes. The calculated transuranic inventories are based on various fuel parameters including initial ^{235}U enrichments. Figure 16 shows the relative neutron emission rate for fuel material with an initial ^{235}U enrichment of 2.56% and with selected cooling times. From Fig. 16, we estimate the neutron source rate for samples with 36 GWd tU exposures and 17 month cooling times to be about $7000 \text{ n s}^{-1} \text{ cm}^{-1}$. From the experimental measurements, the neutron source rate is about $6700 \text{ n s}^{-1} \text{ cm}^{-1}$. This experimental value was determined by assuming a 12% (which is typical of an HLNCC) detector efficiency and a fuel mass density of 10.3 g cm^{-3} .

The model used for the calculational results in Fig. 16 is for a 15×15 instead of a 14×14 array like the Calvert Cliffs fuel. Also, the calculational source rate terms do not include any transport effects. Self shielding in the

Fig. 16. Neutron source rate as a function of exposure for four cooling times.

actual samples probably would lower the experimental count rates. However, the good agreement between the estimated calculational and experimental values promotes confidence in the accuracy of the calculational model. Data from the four cycle analysis are needed to confirm this conclusion.

Comparison of calculated data and destructive data for the Calvert Cliffs rod sections is shown in Fig. 17, where the boxed area is enlarged and shown as Fig. 18. In these figures, the ^{244}Cm ^{239}Pu ratio is represented as a function of exposure. This ratio is used because the destructive data for the curium isotopes were reported in this fashion. Curium 244 is the dominant neutron source in fuels with high exposures and long cooling times. Proposals²⁸ have been made to relate neutron count rates to ^{244}Cm content and then to correlate the ^{244}Cm to parameters such as total fissile, total plutonium, or total plutonium minus ^{239}Pu . Such correlations depend on accurate calculational models that can be used with experimental measurements to estimate the fissile or plutonium content of spent fuel assemblies.

For the limited data available, information in Figs. 17 and 18 tend to confirm calculational predictions of ^{244}Cm concentrations, at least for exposures up to $3 \times$

GWd/tU. As shown in Fig. 18, most destructive data are within the indicated error limits of the calculational curves. When available, the four cycle data will be important in this analysis. As seen in Fig. 17, the ^{244}Cm ^{239}Pu ratio increases dramatically for exposures between 35 and 50 GWd/tU. If the calculational models are incorrect, a larger error between experimental and calculational results probably would occur for the high exposures of the four cycle rods.

An additional comparison between calculated data and destructive analysis data is shown in Table VI, where plutonium isotopic data are compared, showing reasonably good agreement. Certainly the calculated data have the proper trends, indicating that calculational techniques are effective in predicting spent fuel parameters. Discrepancies, particularly in the ^{240}Pu and ^{241}Pu comparisons, may have resulted from use of a 15×15 instead of a 14×14 calculational model for the Calvert Cliffs fuel. Further calculations will be performed using specific Calvert Cliffs models to determine if better agreement on these plutonium isotopic data can be attained. In some cases, for example the ^{244}Cm ^{239}Pu ratio, the value may be insensitive to specific reactor parameters, and a model of a generic type reactor may be sufficient to generate calculated quantities.

Fig. 17. Comparison of destructive and calculated results for 2.56% and 2.2%.

Fig. 18. Expanded curve showing a detailed comparison of destructive and calculated results

TABLE VI. Plutonium Isotopic Comparisons for Rod Sections with Exposures of Approximately 36 GWd/tU % of Total

Isotope	Calculated Value	Measured Value
²³⁸ Pu	1.7	2.277
²³⁹ Pu	49.9	48.533
²⁴⁰ Pu	24.3	28.216
²⁴¹ Pu	15.4	12.553
²⁴² Pu	8.7	8.421

C. Nuclear Waste Assay*

1. **A High Efficiency Modular System for the Assay of Plutonium in Large Waste Packages** (E. R. Shunk, S. W. France, G. C. Herrera, and J. T. Caldwell). A 4 π coincidence neutron counting system with assay chamber dimensions of 1.2 $\times$ 1.2 $\times$ 2.4 m and a 15% detection efficiency has been designed, constructed, and used to determine the plutonium content in large crates of miscellaneous nuclear waste.

A significant economic and environmental concern to the nuclear industry is determining the transuranic nuclide content in bulk wastes. No instrumentation exists to screen such wastes in large packages at the required burial limit sensitivity of 10 nCi/g of waste. (A typical waste crate is 1.2 $\times$ 1.2 $\times$ 2.4 m and weighs 1000 kg; about 100 mg of plutonium in a 1000-kg waste package results in 10 nCi/g.) Such bulk waste either is assumed to contain more than 10 nCi/g, which entails expensive retrievable storage, or is grab-sample assayed assuming matrix homogeneity, a procedure that can lead to serious errors in transuranic-content estimates. We report here on a direct, large package transuranic assay system that we have designed, built, and used to assay plutonium content in typical large waste crates.

Our design goals included the following: (1) portability—we are scheduled to use this system in assay campaigns at locations several hundred kilometers apart; (2) overall detection sensitivity of 100 mg of plutonium; and (3) flexibility—the waste crates to be assayed in our program are not a standard size. Our system uses neutron total and coincidence counting of the contained ²⁴⁰Pu, with a 4 π neutron detection sensitivity of about

15% to obtain the 100-mg plutonium sensitivity. The total plutonium mass then can be determined by using the ²⁴⁰Pu isotopic abundance, if it is known.

We incorporated these design goals with six large, stand alone, detection modules that may be assembled into large 4 π counting geometries to accommodate various waste package sizes. Each detection module is a 1.27 or 2.54-cm thick polyethylene boxlike enclosure in which high efficiency ³He proportional counters are placed. Each has two chambers separated by 2.54 cm thick polyethylene and is reinforced structurally with aluminum rods and unistrut. Seven ³He proportional counters (four in one chamber, three in the other) that measure 2 atm of fill pressure, 5 cm in diameter, and either 1.83 or 0.91 m long are contained within the module.

This moderator-detector configuration has been chosen to maximize counting efficiency per ³He tube. Four detection modules have external dimensions of 2.49 $\times$ 1.22 $\times$ 0.33 m and two have dimensions of 1.22 $\times$ 1.22 $\times$ 0.33 m. In our nominal 4 π configuration, the large modules form the sides and the small modules form the ends of an assay chamber with internal dimensions of 1.2 $\times$ 1.2 $\times$ 2.4 m. Our measured 4 π detection efficiency in this configuration is 15%. Alternate detection module configurations (with less detection efficiency) are easily arranged to accommodate packages up to 2.4 $\times$ 2.4 $\times$ 2.4 m.

Separate counting electronics channels are provided for each chamber in each of the 6 modules for a total of 12 individual logic pulse outputs. These 12 separate logic pulse signals are routed to our LeCroy 3500 data-acquisition system by two Kinetic Systems 3610 hexscalars. These 12 signals are recorded and stored separately on permanent disk files. The relative singles count rates from different portions of the 4 π system are used for geometric and matrix compensation algorithms. The 12 signals are summed by the software and the summed values are used for neutron coincidence determinations.

2. **Passive and Active Assay Measurements on Crate-Size Waste Packages** (J. T. Caldwell). Much of the plutonium and uranium-containing wastes and scrap generated in the nuclear industry is ultimately packaged in large crates of 1.0 m or more on a side. We have developed three independent neutron detection measurement techniques to perform assays on such large containers and have built field worthy instrumentation to

*The principal sponsor of this work is the DOE Transuranic Waste Management Program.

implement these techniques. This instrumentation has been used to perform independent plutonium, ^{235}U , and ^{238}U assay measurements on waste crates ranging in enclosed volume from 0.85 to 1.61 m³.

We have developed three neutron detection assay techniques: (1) passive 4π neutron counting,²⁹ (2) active 4π pulsed neutron interrogation (differential die away^{30,31}), and (3) 8 MeV bremsstrahlung photofission. (The instrumentation for these measurements is discussed in the references.) The neutron detection instrumentation is the same for techniques (1) and (3). This instrumentation has been designed to be operationally flexible²⁹ in that the 4π detection system is built from discrete moderated ^3He proportional counter modules, as described above for application to plutonium assay in large waste packages.

For passive assay of crates, our measured 3σ detection sensitivity is about 10 mg of ^{240}Pu for an unshielded detector with sea level cosmic ray background levels. In the assay campaign, we deduced individual crate ^{240}Pu mass contents ranging from 10 mg to 2.5 g. Individual crate assay times were about 1 h. Matrix compensation techniques²⁹ were used in deducing ^{240}Pu masses.

The differential die away pulsed neutron technique is discussed in detail in a recent paper.³² Thermal neutron fission is used here. Generally, ^{235}U and ^{239}Pu are measured, as well as other fissile species. Under favorable conditions, prompt to delayed ratios may be used to obtain separate ^{235}U and ^{239}Pu mass estimates. A crate size version of the differential die away instrumentation has been built but was not available during our crate assay campaign. The measured detection limit is 5 to 10 mg for ^{239}Pu or ^{235}U .

Bremsstrahlung from a commercially available 8 MeV medical radiography electron linac was used to perform saturation irradiation photofission delayed neutron time history measurements on the large crates. These time histories differ markedly among the three isotopes ^{235}U , ^{238}U , and ^{239}Pu , in both yield and shape. With calibration samples, we observed that ^{235}U and ^{238}U have about the same total yield per gram but differ greatly in shape and that the shape of the ^{239}Pu time history is similar to that of ^{235}U but the yield is only 40% of the ^{235}U value. Thus, by combining the photofission delayed neutron time history data with the passive neutron measurements (same detection system used), we obtained estimates for all three isotopes. The observed minimum detectable ^{235}U or ^{238}U mass for the photofission measurement was about 1 g placed anywhere within a crate.

3. Multi Isotopic Transuranic Waste Assay System
(W. E. Kunz, J. D. Atencio, W. Bernard, G. C. Herrera, H. H. Hsu, J. C. Pratt, and J. T. Caldwell). Oak Ridge National Laboratory (ORNL) has accumulated about 3000 barrels of transuranic wastes in retrievable storage. We have been commissioned to construct an assay screening system to measure the ORNL waste and in particular to determine whether the alpha activity of each barrel is above or below the 10 nCi/g maximum level required for low level permanent burial. These wastes, which were produced by several sources, present a complex assay problem. For instance, the Oak Ridge high flux isotope production reactor, whose products are one of the waste sources, generates wastes containing several higher atomic number transuranics, such as curium, californium, and einsteinium. Other waste streams generate fissile transuranics, such as ^{239}Pu , ^{241}U , and ^{241}Pu .

The assay system that we are constructing, called the differential die away system, will perform functions such as passive neutron counting, including totals, coincidences, and event multiplicities; measurement of total fissile material content; measurement of fissile prompt to delayed neutron ratios; and determinations of the effects of the waste matrix on the measurements. An earlier publication contains more information on these measurements.³²

This assay system will have an assay chamber that is similar to our current operational system³² and that is large enough to accommodate a standard 208 L barrel and a barrel rotator. The chamber will have an inner liner of 0.11 m thick graphite and an outer layer of 0.17 m thick polyethylene. A small, pulsed D + T neutron generator, encased in a 0.10 m thick lead shield, is located in the chamber floor. The 15 μs long pulses of 14 MeV neutrons interact with the lead by inelastic and (n,2n) reactions, which increase the number of fast neutrons trapped in the chamber and walls. These neutrons are thermalized within a few tenths of a millisecond. The resultant thermal neutrons have a characteristic lifetime within the chamber of about 1 ms.

Cadmium shielded fast neutron detector packages will surround the assay chamber within the moderating layer. If no fissile material is in the chamber, the detector record indicates only the background level after 0.7 ms because the detectors are insensitive to thermal neutrons. However, when the interrogating neutrons induce fission reactions in fissile material within the waste or scrap package, ensuing prompt fission neutrons are detected

with high probability. To monitor the interrogating thermal-neutron flux, we have placed a low-efficiency, bare ^3He proportional counter in the assay chamber. The ratio of the prompt fission neutron detector counts to the interrogating flux monitor counts is proportional to the amount of fissile material present. The measured sensitivity of this system is 1 mg of fissile material (^{239}Pu , ^{235}U , and ^{233}U) located anywhere within a 208-L barrel of typical transuranic waste matrix.

The counting efficiency of the cadmium-wrapped, fast neutron detectors for fission neutrons will be about 5%. This efficiency is low for coincidence or event multiplicity determinations from low intensity neutron sources or for prompt-to-delayed neutron ratio determinations for small amounts of fissile material. For this system, we will increase the total counting efficiency to about 15% by adding bare ^3He proportional counters. The bare counters, when incorporated into the chamber walls, have almost three times the counting efficiency for fission neutrons as do the cadmium wrapped counters. These counters, which are sensitive to thermal neutrons, will be useless during fissile material neutron interrogation until all pulse produced thermal neutrons have died away; on the average, this takes about 20 ms after the pulse. Thus, for prompt to delayed determinations, we would pulse at about 20 Hz, count prompt neutrons from 0.7 to about 4.7 ms with the cadmium wrapped counters, and count delayed neutrons with both the bare and cadmium wrapped counters from 20 to 50 ms. We measure both fissile amount and the prompt to delayed ratio during the same interrogation period.

For passive measurements of neutron coincidences and event multiplicities, we will be limited to low neutron source strengths when using the combined cadmium-wrapped and bare counters. The thermal neutron lifetime for the chamber, and thus that for the bare counters, also is near 1 ms, thereby requiring a coincidence gate 1 to 2 ms long. For strong neutron sources, the accidental coincidence rate obtained with this gate length would be prohibitive. However, if we use only the cadmium-wrapped detectors (with a neutron lifetime of about 35 μs), we can use a gate length of only 35 to 70 μs . A strong spontaneous fission source is easily measured with the less efficient cadmium-wrapped detection system.

A combination of the passive and active neutron techniques should enable us to characterize most of the Oak Ridge transuranic wastes. The basic differential die away measurement combined with the

prompt to delayed ratio data for fissile isotopes will enable us to separately quantify ^{239}Pu and ^{235}U or ^{233}U in mixed wastes. The passive coincidence counting, including neutron multiplicity measurements, will enable us to separately quantify mixed spontaneous fission isotopes such as ^{244}Cm , ^{252}Cf , and ^{240}Pu . The overall system sensitivity for these measurements is 10 nCi/g or better for most transuranic isotopes thought to be in the Oak Ridge wastes.

4. Experimental Evaluation of the Differential Die Away Pulsed Neutron Technique for the Fissile Assay of Hot Irradiated Fuel Waste (J. T. Caldwell, W. E. Kunz, J. D. Atencio, G. C. Herrera, and J. C. Pratt). The high sensitivity fissile assay system being developed at Los Alamos for the routine assay of fissile isotopes in 208 L barrels^{10,12} also has been evaluated for the fissile assay of hot irradiated fuel waste. This system uses thermalized pulses of neutrons produced originally in a portable 14 MeV neutron generator to interrogate waste packages. Prompt neutrons from fission events produced in this pulsed interrogation are detected in shielded, high sensitivity, fast neutron detection packages.

In previous publications^{10,12,13} we described our system's performance for the fissile assay of waste packages that, for the most part, contained insignificant sources of neutrons or gamma rays. Because a significant segment of waste is accompanied by large self neutron sources as well as high gamma radiation fields, we evaluated the system for assay of such wastes.

First, to simulate the effects of self sources of neutrons on our fissile assay sensitivity, we placed a strong isotopic neutron source within the assay cavity and performed the usual differential die away pulse measurements with known amounts of fissile isotopes in the cavity. With a 1.26×10^6 n/s source in the cavity, we observed 1.763×10^6 source background counts during our usual 0.7 to 4.7 ms count period and 1×10^4 pulses from the neutron generator. With the strong source and a 500 mg sample of ^{235}U in the cavity, a net induced fission signal of 74 400 counts was observed for the same number of interrogating pulses and the same count interval. Thus, the experimental 3σ above background detection limit for this situation is 27 mg of ^{235}U , or about 18 mg of ^{239}Pu .

This measured 3σ limit is readily scaled to different steady source backgrounds or to a different number of total pulses. For example, for a 10^4 pulse interrogation

(about 167-s elapsed time under usual conditions) and a 1.0×10^7 n/s steady source neutron background, the 3σ detection limit is 50 mg of ^{239}Pu . For a run of 4×10^4 pulses and a 1.0×10^7 n/s background, the 3σ detection limit is 25 mg of ^{239}Pu .

A strong gamma radiation field affects fissile assay sensitivity only in an indirect fashion. The ^3He proportional counters in our assay system are not highly sensitive to gamma radiation fields. With no system modifications, we can tolerate waste barrels producing up to 10 R/h contact gamma radiation fields. Counting electronics (discriminator levels) can be adjusted to bias out the gamma radiation effects with no appreciable loss in neutron counting rate. The fast counting electronics in our system are advantageous in this situation.

We have evaluated the effects of replacing segments of graphite in our assay chamber with lead to directly shield the ^3He detection packages from gamma rays. An amount of lead sufficient to provide 10 cm of direct shielding for the ^3He counters produces no significant

change in observed fissile assay sensitivity. Experimental measurements indicate that this amount of shielding will result in a gamma dose rate reduction factor of about 10^3 for typical hot irradiated wastes. Thus, with no design change other than replacing the graphite directly in front of the detection packages with an equivalent thickness of lead, the differential die away assay system can be operated successfully (no performance degradation) with hot waste barrels producing contact dose rates up to about 10^4 R/h. Operation with even hotter barrels is possible with a design change in which the detection packages are shielded with greater amounts of lead.

We have shown that the differential die-away pulsed neutron assay system is suitable for the fissile assay of hot irradiated fuel wastes, even under conditions of extremely high self neutron and gamma-radiation levels. Screening measurements near the 10 nCi/g level appear feasible for 208 L waste barrels having self neutron sources up to 10^7 n/s and contact gamma fields up to 10 000 R/h.

IV. DEVELOPMENT OF SAFEGUARDS SYSTEMS TECHNOLOGY

A. Design of Integrated Materials Control and Accounting and Physical Security Safeguards (J. T. Markin, A. L. Baker, R. G. Gutmacher, E. A. Hakkila, J. P. Shipley, W. J. Whitty, and T. R. Canada)

The joint Los Alamos Sandia integrated system study is developing a methodology to design and evaluate safeguards systems for nuclear facilities. Because the first part of this study will consider only a generic nuclear facility, we have developed an organizational diagram for a generic facility that shows the relationships between management, process, and safeguards components. A significant feature of this diagram is the recognition that many process activities that are in place before any safeguards overlay have intrinsic safeguards value. This diagram is useful for eliciting relationships between facility activities and safeguards components and in identifying potential integration points.

Because the design process may generate many candidate safeguards systems that must be evaluated against threat scenarios, we have developed a framework to deal with this complexity. The sequence of threat activities and corresponding safeguards countermeasures are represented in a threat countermeasure diagram that allows the analyst to identify the contribution of each

safeguards component to the system's total performance. This diagram is used to develop rules for combining individual component performance measures to find the system's effectiveness. Work is in progress to combine the threat countermeasure diagram with optimization methods so that an optimal safeguards design can be selected from candidate designs. Specifically, the proposed method would select a design that maximizes the probability of interdicting the threat while constraining resource expenditure. Alternative formulations in which resource expenditure is minimized while performance is constrained are also possible.

B. In Process Inventory in Solvent-Extraction Contactors (C. A. Ostenak, D. D. Cobb, and E. A. Hakkila)

Los Alamos continues to sponsor development of techniques for estimating in-process nuclear materials in solvent extraction contactors. These techniques, comprising computer based chemical models and actual process measurements, will be essential for optimizing the sensitivity of near-real-time materials accounting in future fuels reprocessing plants.

During this reporting period, Clemson University issued a draft report³⁴ that describes the mathematical model used in the PUBG computer code, which simulates mixer settler operation. PUBG is applicable to extraction, scrub, partitioning, and strip contactor operations; it allows for deviations from mass transfer equilibrium and accounts for random fluctuations in feed stream concentrations. For partitioning operations, PUBG is applicable to both the hydroxylamine-hydrazine reductant system and the U(IV) reductant system. For the latter, the autooxidation rate of U(IV) is modeled to compete with the reduction rate of Pu(IV).

In addition to the mathematical model in PUBG, the user's manual describes the numerical methods of solution and the organization and structure of the code. Results from PUBG are compared with measured data and with the results from other contactor models.

Obtaining the computer codes for modeling pulse column solvent extraction contactors was discussed with Iowa State University personnel. The code and user's manual will be available in December 1981.

C. Implementation of Optimization Codes (E. A. Kern)

Safeguards systems designs frequently require optimizing a payoff function with respect to several variables. The multiple variable optimization code

PCON was obtained and successfully implemented on the PRIME 750 computer from a listing of the code. PCON was converted to double precision to improve convergence. In addition, the code's dimensions were increased, and the convergence algorithm was modified. The modified PCON code then was used successfully in solving the optimal allocation of measurement uncertainty problems associated with the Hot Experimental Facility (HEF). In one case, PCON minimized a function of 43 variables subject to 88 inequality constraints. Computation times with the Prime 750 for the more difficult problems varied from 30 to 60 min.

D. Simulation Upgrades (E. A. Kern)

We accomplished some initial planning for future modeling and simulation activities associated with the Fuels and Materials Examination Facility (FMEF) at Hanford. Westinghouse Hanford is modeling a portion of the FMEF with the Q GERT simulation language, and they want us to use Q GERT in our modeling to make the codes transportable. After a short investigation, we concluded that Q GERT does not provide enough flexibility. However, a new simulation package, SLAM II, provides all the Q GERT capabilities as well as the increased flexibility required for our modeling. A purchase order for the SLAM II software was placed with Pritsker and Associates.

PART 3 NUCLEAR FACILITY SAFEGUARDS SUPPORT

I. TEST AND EVALUATION PROGRAMS AT NUCLEAR FACILITIES

A. A Plutonium Isotopic System for SRP (T. E. Sampson, S. T. Hsue, E. L. Sandford, S. S. Johnson, D. L. Cramer, and K. Kroncke)

In early February 1981, we decided to accelerate an FY 81 program milestone of planning a plutonium isotopic system test and evaluation at SRP to actually deliver the system in FY 81. The instrument will be used to measure scrap, waste, and JB line buttons as well as incoming feed. Using existing equipment, we constructed, calibrated, and documented a system that was ready for delivery by the end of May 1981. Delays were caused by SRP's request that the system be housed in an

air conditioned cabinet and by the lack of physical facilities at SRP to accommodate the instrument. Both items have been addressed and shipment to SRP is planned during August 1981.

The system consists of a single 200 mm² × 10 mm deep intrinsic germanium detector with conventional nuclear instrument module (NIM) electronics that incorporate pulse pileup rejection and digital gain and zero stabilization coupled to an LSI 11 computer based MCA for data acquisition and reduction. All input/output is through a hard copy terminal.

The techniques used for the isotopic measurements have been described previously.¹⁵⁻¹⁷ For the SRP system,

the software was modified to improve the $^{239}\text{Pu}/^{241}\text{Pu}$ ratio measurement for low (0.3 wt%) ^{241}Pu content samples as well as to allow the assay of ^{241}Pu - ^{237}U non equilibrium samples (age since uranium separation less than 45 days).³⁸ The software also allows the use of multiple constants files (regions of interest and calibration constants) so the best calibrations for specific material types may be used. Acceptable results for many cases are obtained by using fundamental published half life and branching ratio data without recourse to any standards whatsoever. However, the best results are obtained only after calibration with standards to remove small (a few per cent, typically) biases caused by branching ratio errors and die specific peak integration and relative efficiency determinations used.

In Fig. 19, calibration results for the SRP system are shown for a wide range calibration covering ^{240}Pu contents of 6 to 19 wt%. For ^{239}Pu , ^{240}Pu , and ^{241}Pu , scatter in the points comes mainly from the difficulty in calibrating for the wide range of isotopes and sample sizes (0.25 to 500 g Pu). For ^{238}Pu and ^{241}Am , a significant contribution to the observed standard deviation arises from the uncertainty in the "accepted value." Counting statistics are, in general, better than the observed scatter. The precision of the individual isotopic results depends on many factors, including count time, count rate, sample plutonium mass (smaller samples generally give better precision than larger samples), sample geometry, and sample isotopic distribution. In Figs. 20 and 21, we show the observed precision compared with that predicted from counting statistics for two samples. The precision was determined from 30 measurements for each counting time at a count rate of about 13 kHz. These precision determinations combine to give the observed precision for the specific power shown in Fig. 22. For the specific power, a precision of about 1 RSD can be obtained in a count time of 1/2 h for either sample type.

B. Test and Evaluation of the In-Line Plutonium Solution Densitometer at SRP (H. Smith, T. Marks, S. Johnson, R. Slice, J. Sprinkle, L. Cowder, C. Shonrock, and D. L. Garcia)

The off-line testing of the SRP K-edge plutonium solution densitometer³⁹ is essentially complete. In addition to tests reported earlier,⁴⁰ the long-term stability of the instrument has been demonstrated through periodic measurements of both the unattenuated photon in-

Fig. 19. Calibration of SRP isotopic system.

tensities through an empty cell and the effective thickness of a standard plutonium foil permanently installed in the instrument. A plot of the foil assay data over the first 8 months of the off-line testing phase (Fig. 23) illustrates the densitometer's stability over this extended time.

Investigation of plutonium assay precision achieved at plutonium concentrations from 5 to 200 g Pu/l demonstrated the densitometer's usefulness far above and below its design range of 20 to 40 g Pu/l.³⁶ Further application on mixed uranium and plutonium solutions has shown that accurate plutonium assays are possible on solutions with uranium/plutonium concentration ratios up to 2:1 and possibly higher, through use of a transmission extrapolation technique⁴¹ (see Fig. 24).

Fig. 20. Observed isotopic precision for 500 g sample of low burnup plutonium metal (6% ^{240}Pu). Thirty measurements were performed at a count rate of 13 kHz at each point.

Fig. 21. Observed isotopic precisions for 1000 g PuO_2 sample (12% ^{240}Pu). Thirty measurements were performed at a count rate of 13 kHz for each point.

Fig. 22. Observed precision of the specific power determined by gamma ray spectroscopy for two samples. Thirty measurements were taken at a count rate of 13 kHz for each point.

Fig. 23. A summary of all individual assays of the standard plutonium foil over the 8 month period from May - December 1980. The rms scatter of the assay points is within 0.5% of the average foil assay value of 29.375 g Pu/L (effective). The error bars indicate the quoted statistical precision of an individual 40 min measurement ($\pm 3\%$).

Fig. 24. A comparison of the SRP K edge densitometer plutonium assays on mixed uranium-plutonium standards with the analytical chemistry results. The assays without the transmission extrapolation procedure exhibit increasing inaccuracy for increasing uranium-plutonium concentration ratios. The extrapolation procedure seems to remove the inaccuracies successfully.

A mock-up of the solution plumbing system to be used in-line was constructed for the off-line tests. All hardware components (valves, pump, flow meter) were duplicates of the devices to be used in the process installation. The solution tubing and sample cell were transparent to facilitate full observation of the solution flow. The complete plumbing system was exercised in a cold test

mode (that is, with water), both manually and under computer control, by performing all possible assay sequences, error conditions, interlock criteria, and observation of system responses. We estimated solution flow rates and set preliminary software delay times in preparation for application in the in-line plumbing. Both the control software and plumbing circuit performed satisfactorily, with all tests carried out successfully.

Installing the densitometer solution plumbing system in the process line and in line test and evaluation depend on finding available time in the process schedule for connection of the plumbing hardware to the process holding tanks.

C. L_{III} Edge Densitometer Evaluation at NBL (J. K. Sprinkle, Jr., S. S. Johnson, D. G. Langner, T. Marks, and P. Russo)

The off line evaluation⁴⁰ of the L_{III} -edge densitometer⁴² at NBL is in progress. The uranium assays are complete (see Fig. 25). Six samples were made of each of six different solution concentrations, and each of these 36 reference solutions was analyzed once by a wet chemical technique⁴³ and three times by the densitometer. These data will be used for calibration and, consequently, the average bias of approximately 2% will be removed. These results represent the assay precision over this concentration range. The concentration dependent calibration is not well understood,

Fig. 25. Summary of uranium assay data by L edge densitometer compared with destructive chemical analysis

although it has been consistently observed. It is a small effect that can be taken into account by using more than one calibration constant. The data in Fig. 25 show that this instrument in its present development requires standards (during calibration) that span the range to be assayed, unlike many NDA devices.

Evaluation of the densitometer's capabilities for plutonium assay is nearly finished. The next feature to be evaluated is the assay of mixed uranium and plutonium solutions. These results will be presented later.

The in-line version of the instrument will be installed at Allied General Nuclear Services (AGNS) in August 1981. This version has been modified for near-real-time assay of flowing streams as described in the next section.

D. L-Edge Densitometry for Real-Time Concentration Measurements of Flowing Streams (J. K. Sprinkle, Jr. and P. Russo)

Application of NDA to in-line or on-line measurements of flowing solution streams for near-real-time accounting of SNM requires modifications in procedures and technologies applied in instrumentation designed for analysis of static solutions. Assay techniques such as absorption-edge densitometry or transmission-corrected gamma-ray assay use measured transmissions of gamma rays through the SNM to determine total or isotopic SNM concentrations. Because gamma-ray transmission is a negative exponential function of the SNM concentration, the analysis of data accumulated during finite

counting periods, when SNM concentration varies, will give a result that is biased low compared to the actual average concentration of the counting period. The relative bias increases in magnitude with the relative variation in concentration for two reasons:

- Measurement with a fixed intensity transmission source leads to losses in statistical precision that increase with concentration, resulting in the heaviest weighting of data obtained with the lowest solution concentrations.
- The absolute change in the logarithm of the measured transmission is equivalent to the relative change in the measured transmission. This also results in an effective weighting in favor of lower solution concentrations.

A practical approach to these problems involves reducing the data-acquisition time so the expected variation in concentration is sufficiently small to cause a negligible bias. This requires both a knowledge of the process (to predict the range and period of the fluctuations in concentration) and of the bias magnitude in a given concentration range.

The L-edge densitometer,⁴² scheduled to operate on line at the AGNS facility in Barnwell, South Carolina for uranium concentration assay of flowing streams, must be optimized for this operation mode. Therefore, calculations have been performed that evaluate the bias in two operating circumstances. The first assumes that the transmission source intensity is fixed during the data-acquisition period. Concentration $\rho(t)$ varies linearly during the acquisition period T such that

$$\begin{aligned}\rho(0) &= \rho_{LO} \\ \rho(T) &= \rho_{HI} \text{, and} \\ \rho(t) &= a + bt\end{aligned}$$

Therefore, the average concentration $\bar{\rho}$ during the counting period T is

$$\bar{\rho} = \frac{(\rho_{LO} + \rho_{HI})}{2}$$

The second circumstance consists of varying transmission source intensity so the detector count rate remains constant despite the same change in concentration.

In Fig. 26 results are plotted for different average concentrations as the percentage negative bias in the L-edge assay result vs the concentration range $\Delta\rho$. The quantity $\Delta\rho$ is one half the percentage variation in concentration:

$$\Delta\rho = 100\% \times \frac{\rho_{HI} - \rho_{LO}}{2\rho}$$

The dashed curves show results for the fixed transmission source intensity. The solid curves represent results for a constant detector count rate.

Fig. 26. The percentage negative bias as a function of one half the percentage variation in concentration.

Operation with a fixed detector count rate reduces the expected relative bias by a factor of 3 to 4. The advantage of maintaining constant detector count rate has prompted the effort for automatic control of the current on the x-ray generator to maintain a constant detector count rate during the L-edge assay. This instrument will operate on line at AGNS with a fixed, predetermined count time that is short compared to the expected period of the concentration variations.

Perhaps the results of the AGNS exercises (or of other such experiences of real-time monitoring of flowing process streams) will demonstrate enough concentration change so the use of fixed counting periods may be undesirable. A possible solution for instruments designed to operate in real time is to vary automatically the counting period to minimize changes in concentration within the period.

Finally, because the real-time results at AGNS will be averaged over a material balance period for demonstration of materials accounting, the real-time variations in concentration should be used to correct the highly precise average for biases of the magnitude indicated in Fig. 26.

E. Advanced Materials Accounting System Demonstration at AGNS (Los Alamos Safeguards Staff)

A series of *miniruns* demonstrating the applicability and effectiveness of near-real-time accounting to reprocessing plants was conducted jointly by AGNS and Los Alamos personnel in 1980. The results were presented at the Third Annual ESARDA Safeguards Symposium at Karlsruhe⁴⁴ in an AGNS/Los Alamos poster session, which drew considerable interest from reprocessors around the world. The presentation illustrated that process control data can be applied to detecting abrupt and protracted diversion and process upsets.

In a continuation of this work, the first of two *miniruns* for 1981 was held during this reporting period. This experiment had three major objectives: (1) refine the decision analysis programs for eventual transfer to safeguards operating personnel at AGNS and other facilities; (2) determine the location, schedule, and logistical support required to install an L-edge densitometer at AGNS before the next *minirun*; and (3) perform a pulsed-column inventory experiment at the end of the *minirun*. All three objectives were completed successfully. The next *minirun* is scheduled for mid-August 1981.

To provide more timely measurements of process streams for the August minirun, an L_{III} -edge densitometer will be installed in the 2BP stream. As discussed above, the densitometer will measure the concentration of flowing solution streams.

F. Evaluation of NDA Capabilities for Inventory of Uranium in Solvent-Extraction Columns (P. Russo and R. Strittmatter)

During a recent visit to the Oak Ridge Y-12 facility for enriched uranium scrap recovery, measurements were performed to evaluate the capability to inventory the SNM content of the solvent-extraction systems by using NDA techniques with adequate precision for materials accounting. The equipment suitable for such purposes must be low in cost, easy to maintain, and compatible with hostile nitric acid environments.

The two solvent-extraction systems each have three columns identified as the extraction, strip, and backwash columns. These stainless steel columns are 9.14 m (30 ft) high and 10.2 cm (4 in.) in diameter and contain thin stainless steel sieve plates at 2.54-cm (1-in.) intervals for phase dispersion during pulsing. The aqueous, acidic uranium feed is input near the top of the extraction column. The uranium partitioning favors the organic phase that is input near the bottom of the column and extracted near the top. This extracted organic phase is input near the bottom of the strip column where the partitioning still favors the organic rather than the still-acidic aqueous phase. The organic extracted near the top of the strip column is input at the bottom of the backwash column where partitioning favors the aqueous phase, which is no longer acidic. The aqueous product is extracted near the bottom of the backwash column.

Column inventories are estimated by sampling at a single point. As a result of column geometries and partitioning effects, uranium concentration gradients along the columns are expected to be steep during pulsing operations. If these gradients persist after shut-down during inventory, the sampling approach can cause large inventory uncertainties. The Y-12 measurements were designed to examine these gradients in the static configuration and to determine the accuracy of the column inventory that might be feasible in this configuration.

A 5.1-cm (2-in.) $\times$ 1.3-cm (1/2-in.)-thick NaI(Tl) detector, collimated to a 1-in. diam, was used with a portable, stabilized assay meter (Eberline SAM-II) to

obtain the ^{235}U 186-keV-photopeak count rates at various vertical heights along the columns of one of the secondary solvent-extraction systems. The calibration for uranium concentration was based on a calculation that assumes a known ^{235}U enrichment. The calculated calibration incorporates the following effects:

- Small-angle scattered 186-keV events that fall within the finite width of the 186-keV photopeak in the NaI(Tl) spectrum. These events are estimated to represent 35% of the detected rate for the 186-keV photopeak.
- Sample self-attenuation of the 186-keV gamma rays. These effects vary, roughly, by a factor of 2 over the range $20 \text{ g/l} \leq \rho_u \leq 200 \text{ g/l}$ where ρ_u is uranium concentration.
- Changes in effective sample geometry as a result of the concentration-dependence of the 186-keV gamma-ray mean-free path. This varies by a factor of 1.6 over the same range of uranium concentration.
- Gamma-ray attenuation caused by the column walls.
- Efficiency of the detection system for counting the 186-keV gamma rays.

The calculated calibration was verified with measurements of enriched uranium solutions stored in 15.2-cm (6-in.)-diam glass columns at the Los Alamos Group CMB-8 uranium scrap recovery facility at Los Alamos. Reference values for these solutions were obtained by NDA of column samples using the CMB-8 uranium solution assay system.⁴⁵

Figure 27 shows the calculated calibration curves for the CMB-8 and Y-12 columns plotted as the observed 186-keV count rate vs uranium concentration. The data points are the observed CMB-8 count rates plotted at the reference concentrations. The solid points correspond to an initial set of measurements that were subject to errors caused by significant and varying 186-keV backgrounds and inadequate detector shielding.

The measurements at Y-12 were obtained in 60-s counting periods; each measured count rate had a precision resulting from counting statistics of about 1%. The concentration profiles (uranium concentration vs height) based on these measurements are plotted in Fig. 28. The heights were estimated to within about 3 ft of the true vertical position. The measurements were performed on static solutions in the three columns of one of the solvent-extraction systems that had been shut down 24 h earlier. Therefore, the aqueous and organic phases were separate as indicated by the solid and open points and by

Fig. 27. Calculated count rate for 186 keV gamma rays plotted vs uranium concentration for solutions of 42% ^{235}U in stainless steel columns assayed by a NaI(Tl) detector. The data points were obtained from measurements of uranium solutions stored in glass columns.

Fig. 28. Measured uranium concentration plotted vs column height for the three columns of one of the secondary solvent extraction systems of Y-12. The data correspond to measurements in the aqueous phase, except for the open points and dashed lines that indicate the organic phase. The curves drawn to smoothly connect the data points have been further smoothed (dotted lines) in two regions where anomalous dips in measured concentration are observed.

the solid and dashed lines, respectively. Because the shutdown was caused by a routine occurrence of normal operation rather than termination of normal operation for inventory, we cannot expect the absolute results of these measurements to be the same as the average results obtained during inventory. However, the steepness of the concentration gradients after the 24 h shutdown period implies that the gradients during inventory also might be steep.

The dips in measured concentration near the bottom of the strip and backwash columns have been smoothed over with dotted lines so the magnitude of the dips is estimated at $\pm 2\%$ (relative to the dotted line). Fluctuations of this magnitude may result from impediments in the columns, whose effects were not incorporated in the calibration and were not considered during the measurements. The two types of impediments in the columns are the sieve plates and the 9.5 mm diam stainless steel stator rods (at 120° intervals at the periphery column) that position the sieve plates at 2.54 cm intervals. The rods and plates extend from the 46 cm vertical position to the 6.4 m position in the column. Thus, the two measurements at the top of each column and the several measurements at the bottoms of the strip and backwash columns are unaffected by attenuation caused by the rods and plates. The maximum effect of the plates, assuming that the 2.54 cm detector collimator is centered between two plates, and the rods, assuming that a rod occupies the collimated region at the region's center, has been calculated as a function of uranium concentration. The results are plotted in Fig. 29 as the percentage loss in the 186 keV count rate vs uranium concentration. The loss caused by the plates (dashed line) decreases with increasing concentration because the fraction of events originating outside the collimated region decreases with the mean free path. The loss caused by the rods (solid line) increases with concentration because the fraction of sample displaced by a rod increases as the effective sample volume is reduced. The sum of the two losses (dash-dot line) is a relatively constant effect of about 35% for all concentrations. Thus, the dips in the observed concentration profiles probably are caused by these losses.

The sharp drop in uranium concentration between the aqueous and the organic phases in the extraction and strip columns results from saturation of the organic solvent with uranium. Although partitioning favors the organic phase in these two columns, solubility favors the aqueous phase. The uranium concentration in the saturated organic (30% TBP in kerosene) is 120 g/L in

Fig. 29. Maximum percentage loss in count rate caused by the attenuation effects of the sieve plates (dashed line) and stator rods (solid line) in the solvent extraction column plotted as uranium concentration. The sum of the two effects is indicated by the dash-dot line.

good agreement with the measured concentrations of the organic phases.

The trend of increasing concentration with height in the extraction column is the result of uranium feed input at the top of the column and extraction of the uranium into the organic phase as it rises to the top. The saturated organic phase is removed near the top of the extraction column and fed into the strip column. The high uranium concentration at the bottom of the strip and backwash columns is the result of input of a saturated organic phase at the bottom of both columns. The uranium remains with the organic phase in the strip column, therefore a high concentration is at the top of this column. The uranium is extracted from the organic phase in the backwash column, so a low concentration is at the top of this column.

The smoothed curves in Fig. 28 (including the dotted segments) were integrated graphically over height (that is, volume) to give the uranium content in the three columns of this system. The result is 2.54, 8.60, and 3.54

kg for the extraction, strip, and backwash columns, respectively, for a total of 14.7 kg of uranium in the three columns. This quantity is subject to uncertainties estimated at the 10% level resulting from neglect of the impediments effects (plates and stator rods) in the columns, as well as uncertainties in the vertical positions of each measurement. These uncertainties can be eliminated in controlled measurement situations in which detectors are positioned at well-defined heights along the column and at equivalent positions with respect to the baffles and stator plates. Comparison of the measured and expected concentrations of the saturated solution phase indicates that the calculated calibration is good to within a few percent. A refinement of this calibration using the standards of nearly identical geometry, combined with further measurement at the Y-12 facility, may be achieved.

The steep concentration gradients in the static Y-12 columns indicate that inventory based on a measurement at a single point in the columns is subject to large errors. The Y-12 measurements also indicate that simple NDA techniques (such as those in these exercises) might achieve a measurement of the SSN contents of the static solvent extraction columns with five or six detectors per column. This assumes that the gradients are relatively smooth, as indicated by the dotted lines in the regions where dips in count rate were observed and that volumes can be specified to ± 1 or 2%. The influence of the plates and stator rods on the count rate must be verified, as must the absence of sharp drops in concentration to insure the capability of a near-inventory measure with only a few detectors. The possibility of improving the columns for destructive analysis of the concentrated solution concentration, in an effort to determine the absolute calibration and for verification of the volume estimates should be explored. The calculated column inventories and profiles should be compared with the calculated results by using the solvent extraction column model (see Part 2, Sec. IV, B) to guide future implementation of on-line monitoring and verification.

G. DOE System Concepts: Analysis of the Plutonium Facility Los Alamos Safeguards System (PF LASS) Data Base (R. C. Bearse, A. J. Baker, D. G. Shirk, and C. C. Thomas, Jr.)

Programs have been written and tested that translate data from the Data General computer at Technical Area

(TA) 55 into compatible language for our Prime computer. Using these programs, the PF LASS transaction file for February 1980 to February 1981 has been read into the Prime. Only those transactions that involve one of the seven receipt areas used in fast flux test facility (FFF) fuel fabrication have been retained for further analysis.

A second series of programs has been developed to analyze this FFF data base. We now can assemble those transactions leading to generation of a particular material in process (MIP). We also have demonstrated rudimentary error propagation (assuming no covariance), more accurate propagation including covariance terms is being developed. An existing plotting program has been adapted to enable us to provide MIP and CUSUM charts (with error bars). The generation of MIPs and CUSUMs for all receipt areas can be accomplished in less than 2 h.

The present data base does not include data before February 1980. Thus, contributions of shipper/receiver differences to some MIPs are not included because of the long time periods required for chemical analyses. We now are correcting this deficiency by selecting the necessary transactions from early data tapes and including them in our data base.

H. Neutron and Gamma Ray Measurements of Spent Fuel Assemblies at the General Electric (GE) Morris Storage Facility (J. R. Phillips, J. K. Halbig, G. E. Bosler, S. E. Klosterbuer, F. A. Duran, and O. R. Holbrooks)

A series of nondestructive neutron and gamma ray measurements was performed on a set of 14 PWR fuel assemblies with exposures ranging from 18.5 to 40.5 GWd/t. This measurement exercise had two objectives:

1. to collect neutron and gamma ray data from each of the four corners and four sides to investigate correlations between operator declared values of exposure and cooling time; and
2. to demonstrate applicability of the Los Alamos developed instrumentation to IAEA, DOE, and private industry representatives.

Each of the 14 fuel assemblies listed in Table VII was measured with the γ positioning system for measuring the corners⁴⁴ and with a square ring detector (Fig. 30) for measuring each of the four sides simultaneously. The ring detector was mounted on a support bracket fitted to the fuel storage baskets at the facility. (Electronics for

TABLE VII. Fuel Assemblies Measured During the June Exercise at the GE Morris Storage Facility

Fuel Assembly	Discharge Date	Initial ^{235}U Enrichment	Declared Exposure (GWd/tU)
A 72	10 16 74	3.40	18.47
A 54	10 16 74	3.40	20.81
A 74	10 16 74	3.40	20.84
A 73	10 16 74	3.40	20.91
D 31	11 16 75	3.04	23.43
C 32	4 6 74	3.40	23.57
D 26	11 16 75	3.04	26.91
C 51	2 26 76	3.40	28.90
B 52	2 26 76	3.02	31.71
C 52	3 4 77	3.40	32.82
C 70	3 4 77	3.04	33.08
C 72	3 4 77	3.04	37.85
C 56	3 4 77	3.40	40.07
C 65	3 4 77	3.40	40.55

Fig. 30. Square ring detector mounted on the support bracket for holding the detector on top of the storage baskets

the detectors are described in Part 4, Sec. III. A.) The fuel assembly first was lowered into the ring detector and each of the four sides was measured at designated axial positions; then the fuel assembly was moved to the V detector and each corner was measured separately. This process was repeated for each of the 14 fuel assemblies.

The neutron results (summarized in Table VIII) were corrected for differences in the cooling time by using the ^{235}U half-life because it is the source of over 95% of the neutrons for these ranges of exposures and cooling times. Data from the four sides and four corners were summed in each analysis. The square ring data seem slightly better, with an average absolute difference between the regression time and the measured values of 2.92%, whereas the V detector had an average absolute difference of 4.41%. In the square ring detector the neutron detectors (14 fission chambers) were placed horizontally so the entire side of the fuel assembly was measured. Asymmetries in the neutron signals were more evident in the V detector measurements because the neutron detectors in this case were placed vertically. These measured asymmetries can be directly related to the irradiation histories of the specific fuel assemblies (for example, for an assembly located near the periphery of the core).

Table IX compares results from the neutron measurements for various measurement combinations. The side measurements generally were in better agreement with the declared values than were the corner measurements. Two opposite side measurements from a "fork" detector had average differences of 2.6 and 3.3%. This type of measurement is easier to use because the fuel assembly can be moved to the detector or the detector can be placed around the fuel assembly without requiring vertical movement. Single corner results show a wider range in the differences than the summed results from all four corners.

Additional analyses will be performed on these neutron results. However, measuring fuel assemblies with neutron detectors placed horizontally on the side of the assemblies appears to give more representative results than do vertical detectors placed at the corners. Average differences of 2.92 and 4.41% were measured for the square ring detector and V detector, respectively. Both results demonstrate the applicability of passive neutron measurements to verify operator declared exposure values for materials accounting and control.

Gamma ray measurements were obtained simultaneously with the neutron measurements. Similar

variabilities between the corners and sides of the fuel assemblies were measured. The internal consistency of a set of similar fuel assemblies can be verified by calculating the correlation between the measured gamma ray dose divided by the declared exposure vs the declared

cooling time of the individual fuel assemblies (see Tables X and XI). The average differences in the calculated and declared cooling times were 2.7% and 3.3% for the side measurements and corner measurements, respectively.

TABLE VIII. Comparison of Declared Values with Results from Least Squares Regression for Power Function

Fuel Assembly	Declared Exposure (GWd:tU)	Square Ring		V Detector	
		Value	Percentage Difference	Value	Percentage Difference
A 72	18.47	17.90	3.17	18.75	1.5
A 54	20.81	21.49	3.18	22.41	7.14
A 74	20.84	21.35	2.41	22.41	7.00
A 73	20.91	21.41	2.34	21.38	2.18
D 31	23.43	21.84	7.27	22.58	3.75
C 32	23.57	24.47	3.69	20.31	16.07
D 26	26.91	25.46	5.69	26.22	2.63
C 51	28.90	30.16	4.17	26.89	7.49
B 52	31.71	33.62	2.78	33.60	5.63
C 52	32.82	31.91	2.86	32.82	0.73
C 70	33.08	32.50	1.80	32.64	1.36
C 72	37.85	37.83	0.06	38.80	2.44
C 56	40.07	40.42	0.86	41.14	2.59
C 64	40.55	40.79	0.58	41.04	1.20
Average absolute percentage difference			2.92		4.41
Average percentage difference			-0.06		-0.17
Parameters					
R ²	0.982	0.952			
α (scaling factor)	0.11015	0.11950			
β (power)	2.81	2.93			
Fitted function					
(count rate) = α(exposure) ^β					

TABLE IX. Comparison of Results from Neutron Measurements

Measured Data	Parameter Value			Average Absolute Difference (%)
	R ²	α	β	
All four sides	0.982	0.11015	2.81	2.92
Sides 1 and 3	0.986	0.05580	2.79	2.57
Sides 2 and 4	0.976	0.05401	2.82	3.27
Side 1	0.957	0.03387	2.72	4.26
Side 2	0.977	0.02186	2.83	3.35
Side 3	0.959	0.02238	2.87	4.42
Side 4	0.961	0.03148	2.82	3.57
All four corners	0.952	0.11950	2.93	4.41
Corner A	0.952	0.02825	2.97	4.41
Corner B	0.918	0.02788	2.96	5.92
Corner C	0.910	0.02774	2.93	6.71
Corner D	0.932	0.03805	2.85	6.10

TABLE X. Relative Gamma Ray Measurements of Fuel Assemblies

Fuel Assembly	Declared Exposure (GWd/tU)	Cooling Time (Days)	Measured Dose ^a		Dose/Exposure	
			Side	Corner	Side	Corner
A 72	18.47	2445	457	207	24.7	11.2
A 54	20.81	2445	517	253	24.8	12.2
A 74	20.84	2445	517	251	24.8	12.0
A 73	20.91	2445	514	228	24.6	10.9
D 31	23.43	2049	666	323	28.4	13.8
C 32	23.57	2638	499	237	21.2	10.1
D 26	26.91	2049	783	375	29.1	13.9
C 51	28.90	1947	857	417	29.7	14.4
B 52	31.71	1947	1035	506	32.6	16.0
C 52	32.82	1575	1266	590	38.6	18.0
C 70	33.08	1575	1275	613	38.5	18.5
C 72	37.85	1575	1529	746	40.4	19.7
C 56	40.07	1575	1602	776	40.0	19.4
C 64	40.55	1575	1616	780	39.9	19.2

^aRelative units.

TABLE XI. Cooling Time Consistency Calculations

Fuel Assembly	Declared Cooling Time (days)	Calculated Cooling Times			
		Sides		Corners	
		Days	Percentage Difference	Days	Percentage Difference
A 72	2445	2404	1.7	2478	1.3
A 54	2445	2397	2.0	2329	-4.7
A 74	2445	2397	2.0	2358	3.6
A 73	2445	2411	1.4	2525	3.3
D 31	2049	2151	5.0	2115	3.2
C 32	2638	2681	1.7	2657	0.7
D 26	2049	2107	2.8	2103	2.6
C 51	1947	2070	6.3	2041	4.8
B 52	1947	1901	-2.4	1858	-4.6
C 52	1575	1594	1.2	1654	5.0
C 70	1575	1599	1.5	1606	2.0
C 72	1575	1512	-4.0	1497	-5.0
C 56	1575	1530	2.9	1524	3.3
C 64	1575	1534	2.6	1542	2.1
Average absolute percentage difference			2.7		3.3

Parameters	Sides	Corners
A	42.97	46.65
B	5.5×10^{-4}	5.8×10^{-4}
r^2	0.9773	0.9679

Equation used to approximate dose rate/exposure vs cooling time: $\text{Days} = a e^{bt}$

II. DIRECT SUPPORT TO NUCLEAR FACILITIES

A. The United Nuclear Corporation (UNC) End-Crop Assay Device (J. K. Sprinkle, Jr., J. E. Stewart, C. O. Shonrock, R. Siebelist, and M. Hykel)

In March 1981, Stewart, Sprinkle, and Shonrock visited the UNC facility at Hanford to discuss possible locations for the end-crop assay device in terms of physical restrictions such as temperature stability, "clean" power, and materials handling. Three locations suggested by UNC personnel were surveyed for neutron background, and shielding measurements were made in one location next to hundreds of metric tons of finished fuel. Collimated slab detector measurements (1000 s) of

two typical end-crop boxes indicated that a counting precision of 1% is easily obtainable for half-filled boxes.

In June and July 1981, the feasibility of assaying shipping boxes filled with end crops was evaluated at Los Alamos. Our 4π neutron barrel counter was modified to use ^3He tubes and to hold the UNC fuel rods. Several measurements made with various uranium loadings indicated that singles counting will yield 2% accuracy over a range of 75 to 526 kg (165 to 1158 lb) of uranium fuel rods. The effects of a nonuniform geometric response of the counter, multiplication, spectral softening, cosmic-ray response, and a copper matrix also were investigated. These data indicate that the design goal of

10% accuracy can be met by the assay device when it is applied to actual fuel element end crops containing significant quantities of zirconium and copper.

A preliminary design was presented to UNC personnel, and a cost estimate was prepared. A final design awaits UNC guidance on loading procedures.

B. The Fluorinel and Storage (FAST) Facility Delayed-Neutron Interrogator (G. W. Eccleston, P. Collinsworth, D. C. Garcia, K. Kroncke, S. Johnson, and T. Van Lyssel)

The FAST facility delayed-neutron interrogator has been fabricated and reassembled in a special cubicle at Los Alamos. The instrument will remain here until final installation at the FAST facility in Idaho Falls, scheduled for July 1982. The housing has the same dimensions as the shielded cubicle that will contain the instrument. This cubicle was constructed to ensure that all instrument components will fit through the door and into the tight spaces allotted for the interrogator. Construction in this housing enabled assembly procedures to be developed and installation problems to be identified and solved. Maintenance and working procedures on instrument components in the cubicle will be monitored and developed during check-out and operation at Los Alamos.

The electronic system has been completed and interfaced to the instrument. It comprises two computer systems with dual system electronics that collect neutron signals from either the spent-fuel or waste-canister assay regions of the instrument. The electronics provide back-up instrumentation that permits the instrument to be made operational immediately following failure of any one component.

Software programs that communicate with systems components control the interrogator. Software is being developed to achieve automatic assays, calibration, diagnostics, and analysis of samples measured in the spent-fuel or waste regions.

Initial measurement tests are being conducted on the system. The ^3He detection efficiency measured for neutrons from a californium source located at the detector midplanes and centered in the waste and fuel tubes was 7.4 and 1.2%, respectively. Vertical profile responses measured as the source was lowered into the center of the through tubes are shown in Figs. 31 and 32 for the waste and fuel sections of the instrument.

Fig. 31. ^{252}Cf source located along the waste tube center.

Fig. 32. ^{252}Cf source located along the fuel tube center.

C. Safeguards Systems Design for the FMEF (R. G. Gutmacher, A. L. Baker, K. C. Greenaugh, J. J. Malanify, J. T. Markin, and R. S. Marshall)

A preliminary review of present nuclear materials safeguards concepts at the FMEF at Hanford was completed.⁴⁷ This review focused on the secure automated fabrication (SAF) line; the computer systems; the vault, in-process storage, and shipping/receiving areas; and the measurements for materials accounting. Insufficient documentation was available on the nondestructive evaluation and destructive evaluation cells and on the test-pin line. However, measurements discussed for the

SAF line are to some degree applicable to the test pin line.

Specific concerns for several areas of the FMEF include

- possible delays because of the inability of vault systems to meet demands;
- the length of time required for vault inventories;
- the vulnerability of in process storage areas;
- the vulnerability of containers in the shipping and receiving measurement area;
- possible inadequacy in the capacities of process vessels and of the scrap storage and processing areas for the SAF process; and
- holdup in the SAF line, especially in those areas where pneumatic powder transport is used.

Suggestions were given about reducing the impact of these concerns, and further studies are planned.

The Hanford Engineering Development Laboratory (HEDL) FMEF staff understands that to integrate effective safeguards systems with efficient nuclear material processing, safeguards and processing considerations must be coordinated from the earliest possible moment. This preliminary review concluded that the present FMEF safeguards concepts are an excellent beginning for an effective integrated safeguards system.

In future work, vulnerabilities in the FMEF safeguards system design and modeling and simulation of the SAF line will be studied.

D. Conceptual Design for the HEDL/FMEF Receiving Station (T. E. Sampson, L. G. Speir, N. Ensslin, S.-T. Hsue, S. S. Johnson, S. Bourret, and J. L. Parker)

A conceptual design report has been completed for a receiving station (see suggested floor plan in Fig. 33) for input accountability measurements on PuO_2 received at the FMEF at HEDL. The report discusses design guidelines proposed by HEDL in mid 1980, technique selection, instrument design and performance characteristics, system integration, mechanical layout, maintenance features, additional development and key interfaces, cost estimates and design and construction schedules, and cost savings areas.

Three demonstrated NDA techniques, neutron coincidence counting, calorimetry, and isotopic determination by gamma spectroscopy, integrated into a versatile

data acquisition system are proposed. This system should perform nondestructive input accountability measurements with precision of better than 1% (1σ) at throughputs of up to 2 metric tons of PuO_2 per year.

Three flexible measurement strategies are proposed using various combinations of the NDA techniques. These strategies can be adapted to current facility requirements regarding accuracy, precision, throughput, and measurement backlog. These strategies, outlined in Table XII, are based on assaying a "batch" of several 2 kg canisters of PuO_2 . Each batch is assumed to have the same isotopic composition and impurity content.

The system also has redundancy features arising from back up hardware components and from the versatility of combining the three techniques. Accountability measurements can continue if any one technique is inoperable.

A detailed design, construction, and a 24 month delivery schedule were proposed. System cost, including hardware procurement, mechanical design and fabrication, and personnel, totaled about \$900 000 (1981 dollars), including a 15% contingency.

This estimate does not include gloveboxes. Significant cost savings can be realized by conducting the assays on doubly contained samples outside the gloveboxes. Additional savings of approximately \$150 000 result from having individual stand-alone instruments, rather than an integrated computer controlled system, although this approach has significant disadvantages.

The conceptual design report in draft form was delivered to HEDL on schedule, 6 months after the start of the work.

E. SRP Support (H. A. Dayem, M. Jain, K. C. Greenough, and D. P. Martinez)

The JB line at SRP converts plutonium-nitrate solutions to high purity plutonium-metal buttons. The process consists of purification and concentration of plutonium nitrate by cation exchange, precipitation of plutonium as trifluoride, drying and conversion to tetra fluoride and oxide, and calcium reduction and finishing. A detailed description of the process and flow sheets were completed and sent to SRP for review.

The process was divided into 22 unit process areas for modeling and simulation. Also, tables for process heel,

Fig. 33. Suggested floor plan for FMEF receiving station.

TABLE XII. Receiving Station Measurement Strategies

I.	II.	III.
Neutron Coincidence Count (all cans)	Gamma-Ray Isotopic Calorimetry (1 can)	Gamma Ray Isotopic Calorimetry (all cans)
	Neutron Coincidence Count (all cans)	
1. Accept shipper ^{240}Pu	1. Gamma-ray isotopic plus calorimetry—total plutonium, 0.5%, 1 σ , plus plutonium isotopic including americium	1. Independent total plutonium measurement on each can. 0.6%, 1 σ
2. Neutron verification, sample-to-sample within a batch, 0.5%, 1 σ , 30-min count time	2. Rapid verification of remaining cans in batch by neutron coincidence count	2. Throughput: 3 cans per day per system
3. Total plutonium, batch-to-batch, 2%, 1 σ , affected by impurities	3. Throughput: similar to strategy I	
4. Throughput: 12 cans per 8 h		

input, and output concentrations or weights were prepared for input to the MODEL⁴⁸ code. Programming for the simulation code is about half complete.

Some information needed to complete process data tables for the simulation will be obtained from SRP personnel.

F. TA-55 Applied Systems Integration (C. C. Thomas, Jr., A. L. Baker, R. C. Bearse, W. Ford, R. S. Marshall, and D. G. Shirk)

1. Development of Distributed Processing. Distributed processing is required to interface on-line NDA

instrumentation with facility safeguards systems such as the TA-55 PF/LASS. As the first step in demonstrating this capability, a PDP 11/34 was received and installed as a preprocessor. Systems performance and timing studies are being performed to evaluate the system's response time for distributed data processing. A PDP 11 760 (VAX) will serve as the main computer for the demonstration system.

2. Graphics Development for PF/LASS Data Analysis. The TA-55 measurement control program generates an instrument history base that can provide the process and accountability personnel with valuable information on instrument performance and trends. Currently, there is no graphics capability for on line display of the information in the PF LASS instrument history data base. Programs for processing the histories were written, tested, and used to generate both CRT and hard-copy plots for the FFTF process balances.

Graphics display (both CRT and hard copy) technologies were developed and demonstrated for both MIP and CUSUM MIPs. This effort, which uses the PF/LASS data base,⁴⁹ the PRIME 750, the Audit Trail program,⁵⁰ and a program for generating CUSUM MIPs, is directed to characterizing the PF/LASS system, identifying R & D efforts needed for more advanced material control and accountability systems, and providing input to our FFTF simulations.

3. Voice Systems. Voice systems are being investigated for potential use in near real-time, on-line NDA instrument networks and access control.⁵¹ Two simple voice controlled systems have been demonstrated. The first, for instrument control, accepts voice commands and issues audible operator prompts. The second is for access control and uses the speaker's voice to verify identity and to actuate external devices.

Experience with the voice-actuated instrument-control system over 3 months with two validated users in the laboratory indicated that the system is reliable and efficient. Statistics were not collected on detailed operation. The incidence of outright rejection of valid words remained acceptable (less than one out of four), and misrecognition was never a problem.

The prototype access control system, still under development and evaluation, is being used to evaluate strategies, vocabularies, and equipment. In one configuration selected for extensive testing and data collection, access occurs after three recognitions before four rejections, or denial after four rejections before three

recognitions. Seven valid users (six male and one female) trained the system, that is, produced vocabulary templates. Sixteen different individuals, 13 males and 3 females, were used as impostors trying to gain access with a valid ID. The impostor success rate was 2.8%. Refinements to the system should reduce this rate by an order of magnitude.

4. Electronic Balance Recalibration History. A set of 5.5 kg range digital electronic balances has been in operation in the Los Alamos Plutonium Facility since early 1978. Balance installation performance and measurement control procedures have been described previously.^{52,53} Recent work on development of a measurement model for the reactor grade plutonium dioxide production has prompted a review of the balance recalibration history.

The measurement control program was conducted from initial balance installation until mid-1980 by the Los Alamos Maintenance and Standards Group (E 1). The weekly logs maintained from August 8, 1978 through June 27, 1980 were reviewed to obtain recalibration data. The balances were divided into two groups. The first group includes balances that have not been modified since installation, and the second group includes balances that have been modified to improve their stability.⁵² The modified balance recalibration history was analyzed before and after modification.

Both the modified and unmodified balances exhibit a wide variation in recalibration frequency, ranging from once a week to once in about 33 weeks. One way analysis of variance and the Wilcoxon-Mann-Whitney test indicate no change in balance performance as a result of the modification, although the recalibration frequency distribution was affected because the performance of some balances is improved after modification whereas that of others deteriorates. The majority of the balances (61.5%) had a recalibration frequency of less than 0.2% recalibrations/week (less than once in 5 weeks).

A detailed technical memorandum describing the results of recalibration history analysis is being prepared.

G. TA-55 Implementation Support (C. C. Thomas, Jr., W. Ford, and L. Osborne)

Plutonium concentration in solutions contained in vertical Pyrex tanks is determined in weighed samples with a SA1. The mass of the solution in the tank is

needed to calculate the total plutonium content. A mass-measurement system based on the *pneumatic bubbler-tube method* was selected for test and evaluation under the conditions encountered in the Los Alamos Plutonium Facility (TA 55). Although the measurement pressure range is 0 to 5 PSID, the instrument must withstand pressure excursions in the range 11 PSID vacuum to 30 PSID pressure. Five transducers were tested, with four units performing satisfactorily under plant conditions.

Resolution of the electronics is 10 out of 32 000 g; however, drifts, noise, quantization error, and other effects limit the accuracy to 30 g. Calibration runs have been made by the gravimetric method. An additional calibration on the experimental test bed was made with a precision pressure gage (Ruska DDR 6000) that has been incorporated into the tank calibration system.

H. Consolidated Fuel Reprocessing Program--HEF (H. A. Dayem, C. A. Ostenak, E. A. Kern, J. T. Markin, D. P. Martinez, and C. C. Thomas, Jr.)

The Safeguards Systems Group was asked to provide technical assistance in materials measurement and accounting technology for the HEF. We have completed the conceptual design of a near-real-time accounting system for the HEF.⁵⁴ The overall study objective was to calculate optimal measurement uncertainties to meet given performance goals while minimizing the total system development cost. This objective required completion of the following tasks:

- formulate near-real-time accounting performance goals,
- define materials accounting strategies,
- identify measurement points,
- specify a reference materials measurement system that represents current technology,
- deduce cost functions for instrument measurement uncertainty improvement, and
- construct an optimal measurement allocation technique.

In previous materials accounting systems studies for various facilities in the nuclear fuel cycle, we calculated the near-real-time accounting system's performance by proposing a measurement system based on current technology or reasonable extrapolations of current technology. The measurement uncertainties of the proposed

instruments were combined and propagated to obtain an overall materials loss detection sensitivity. In this study the approach is reversed. We hypothesize near real time accounting performance goals and, using optimization techniques, we calculate measurement uncertainties required to meet these goals while minimizing the total cost of the system. In this way we can answer the questions: Given limited resources, in which measurement technologies should R & D funds be invested, what measurement uncertainties are most important, and what measurement uncertainties are required to meet a given performance goal?

We applied optimization techniques to satisfy four levels of performance that represent a range of measurement capabilities. The range includes measurements assumed attainable with existing technology (Goal 1) to those desired by IAEA and Nuclear Regulatory Commission (NRC) guidelines. Goal 1 is based on what should be possible if state of the art instrumentation is used. Goal 2 assumes that correlated errors will be reduced by a factor of 4 and random errors by a factor of 2. Goal 3 is based on IAEA criteria, and Goal 4 is based on NRC goals that are being considered.

Near real time accounting can be applied to the chemical separations process of the HEF. For the chemical separations unit process accounting area, an abrupt loss detection sensitivity of 15 kg of plutonium and a protracted loss detection sensitivity of 150 kg of plutonium are obtainable with current measurement technology. These loss detection sensitivities have a 50% detection probability and about a 2% false alarm probability. If subdividing the chemical separation process while maintaining loss detection sensitivity is desirable, then a flow meter with comparable measurement uncertainties to the accountability tanks 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. This requires in-process inventory measurement uncertainty improvements to about 1% precision for process tank volume and concentration measurements. Accountability and plutonium sample tank transfer measurement uncertainties require improvements to about 0.04% volume calibration and standards, about 0.1% concentration calibration, and about 0.05% concentration standards.

I. Verification Experiments and Instrumentation for Enrichment Plant Safeguards (J. E. Stewart, J. C. Pratt, and R. B. Strittmatter)

Verification experiments for radiation monitoring strategies to detect misoperation of centrifuge enrichment facilities were designed and conducted at the Advanced Engineering Test Facility (AETF) in Oak Ridge. Los Alamos radiation monitoring systems used at the AETF included a neutron detector array, gamma detector array, high resolution gamma detector, and two data acquisition systems. Detection sensitivities were measured as highly enriched uranium production was simulated by processing high assay (16, 50, and 90% ^{235}U enrichment) U₆F₈ in a dedicated group of centrifuges. Thousands of measurements were taken, which will serve as benchmarks for calculational studies of the effectiveness of radiation monitor deployment strategies for the Portsmouth Gas Centrifuge Enrichment Plant (GCEP). For example, the response of one neutron detector (corrected for variations in cosmic ray induced neutron background using a reference detector) exceeded a threshold value (alarm limit) 0, 4, and 23 times during processing of the 16, 50, and 90% materials, respectively. During the 6 wk period of these measurements, two false alarms would have registered for the same detector and could have been triggered by normal facility operations.

Los Alamos also participated in the design and execution of a comprehensive series of neutron background measurements at the URENCO-Centec Separation Plant 4 stage 1 (SP4-1) facilities at Almelo, Netherlands. These activities were in collaboration with representatives from the Netherlands Energy Research Foundation (ECN), URENCO Nederland Operations BV (UOBV), and Ultra Centrifuge Nederland NV (UCN). Neutron monitoring equipment developed by the Los Alamos Safeguards Assay Group was used for the measurements and is similar to that deployed in centrifuge development facilities (AETF) at the ORGDP. The Almelo experimental activities represent a necessary first step toward quantifying the effectiveness of neutron detection systems for detecting hypothetical misoperation of SP4. They are but one part of a cooperative United States Dutch program of enrichment plant safeguards. The SP4 preparations for initial operations offer us a unique measurement opportunity.

In another enrichment plant safeguards project, the design of the in line gas phase enrichment meter for

installation in the ORGDP K 25 gaseous diffusion plant was completed and subsequently (early April) reviewed in detail with K 25 personnel. Agreement was reached on a number of issues, including safety certification, mechanical design details, data readout, and ORGDP Los Alamos responsibilities following installation, which is scheduled to begin in September 1981. (The software for this system is described in Part 2, Sec. III, A, 4.)

J. SNM Monitor Applications (P. Fehlau)

Part of our study of monitoring vehicles to detect gamma ray and neutron emitting SNM motivated a new vehicle monitor design that uses gamma radiation detectors placed beside a vehicle. The new monitor components, shown in Fig. 34, look much like those of a conventional personnel SNM doorway monitor but differ from them in sensitivity. The new monitor has large, effective scintillators and digital logic circuitry that make

Fig. 34. Components of a new vehicle SNM monitor include two detector enclosures and a NIM bin with signal conditioning, logic, and power circuitry.

Fig. 35. The insensitive detector photomultiplier and light pipe regions in each column overlap.

it appropriate for personnel monitoring applications as well, where low-intensity signals must be detected. We investigated two personnel monitoring applications during the initial indoor testing period: diversion monitoring for SNM at emergency exits and monitoring for undeclared feed in international safeguards of enrichment plants. A report on the investigation is being prepared for publication; highlights are given here.

The new vehicle portal monitor has plastic scintillation detectors with dimensions of $30 \times 3.8 \times 91$ cm; a 23-cm-long light pipe is used between the scintillator and the photomultipliers. Two detectors are mounted in each column with their insensitive photomultiplier-light pipe regions overlapping, as shown in Fig. 35. For all applications measurements, the detector columns were placed 76 cm or 2 m apart.

Measurements to evaluate emergency exit monitoring used the 2-m spacing; the data included enriched uranium source scans through the monitor and similar scans with people moving through the monitor to

determine the effect of body shielding on detector background count rate. A running speed of 2.54 m/s was the fastest speed observed in a test to determine evacuation speed; that speed was used to convert the scanning data to results expected in an evacuation. A figure of merit (FOM S^2/B), which is the signal squared divided by the background, of about 18 was obtained for a 10-g ^{235}U sphere in a $22\text{-}\mu\text{R/h}$ background. The 50% detection level requires a FOM of 16. The new monitor would meet the usual personnel SNM monitoring requirements when used at a 2-m-wide double emergency door for monitoring running individuals.

The second application uses standard, 76-cm spacing. The objective is to detect recently separated natural uranium in the form of UF_6 . Separation of the UF_6 from uranium daughters reduces gamma-ray emission to a minimum and makes it difficult to detect, particularly when lead shielding also is used.

We obtained a separated sample and monitored its gamma ray activity over a period of time. The observation and a fitted theoretical curve for three types of measurements are shown in Fig. 36. We used these data,

Fig. 36. Growth curves for the ^{234}Pa daughter radiation building up in freshly separated natural UF_6 .

adjusted for the worst case attitude in the monitor, to calculate the mass of ^{235}U that would be detectable 95% of the time in a $15\ \mu\text{R/h}$ background. The results are presented in Table XIII. The sensitivity for the fresh material is less than desired but can easily be improved by using separate detector logic for upper and lower pairs of detectors, narrower spacing, or longer occupancy time.

TABLE XIII. The 95% Detection Masses in $15\text{-}\mu\text{R/h}$ Background

	UF, Age		
	24 h	50 day	Shielding
Quantity of ^{235}U	1.4 kg	64 g	Bare
	23 kg	1.04 kg	1.25 cm of lead

PART 4 INTERNATIONAL SAFEGUARDS, TECHNOLOGY TRANSFER, AND TRAINING

I. SAFEGUARDS TRAINING

A. Safeguards Technology Training Courses

The Los Alamos Safeguards program conducts the following formal training courses each year for representatives from domestic and international nuclear communities:

- Fundamentals of Nondestructive Assay of Fissionable Materials,
- Gamma Ray Spectroscopy for Nuclear Materials Accountability,
- Advanced Neutron Assay Methods, and
- Integrated Safeguards Systems.

Additionally, four special courses on NDA for inventory verification are conducted annually for new IAEA inspectors, and a Nuclear Nonproliferation Safeguards course, which resulted from the US Non-Nuclear Proliferation Act (NNPA) of 1978, is offered each year to representatives of countries that may be planning national safeguards systems. These courses provide training for approximately 150 attendees per year.

The training sessions that took place during this reporting period are described in greater detail below.

B. Safeguards Inspector Training (T. D. Reilly)

Los Alamos has developed the Basic NDA Inspector Training Course for IAEA safeguards inspectors. All new inspectors attend this course at Los Alamos after completing an extensive training program in safeguards

technology in Vienna. They visit nuclear facilities worldwide to verify inventories of nuclear materials and certify that none is being diverted for military purposes. This is a major part of the IAEA effort to verify compliance with the Treaty of Nonproliferation of Nuclear Weapons.

This course was presented at Los Alamos February 18-26, 1981, and June 14-25, 1981. Each time, 12 participants spent 7 days in the NDA training laboratories. They came from 11 countries and represented the six inspection sections of the IAEA. Since 1978, this course has been presented eight times to 100 inspectors; a majority of the present IAEA inspectors has received NDA instruction at Los Alamos.

This laboratory course provides instruction in the use of NDA instrumentation available at the IAEA, most of which has been developed at Los Alamos. Working in pairs with a Los Alamos instructor, the participants perform neutron assay experiments, largely with the HINCC, for 3 days then spend 3 days using gamma ray assay instrumentation. This time the gamma ray assay experiments included work with the new portable MCA being developed at Los Alamos, in addition to the SAM 2, BSAM, and Silena that always have been used. The nuclear materials available for measurement include cans of bulk UO_2 and PuO_2 (up to 1 kg), small samples of PuO_2 with different isotopic composition, LWR fuel pellets and rods, and PWR and BWR fuel assemblies.

Future training sessions are scheduled for September 1981 and January 1982.

C. NNPA Course on Nuclear Materials Accountability (G. R. Keepin, C. R. Hatcher, and T. D. Reilly)

An Advanced International Training Course in State Systems of Accounting for and Control of Nuclear Materials was held April 27 - May 12, 1981, at Santa Fe and Los Alamos, New Mexico and at Richland, Washington. The course, sponsored by the US DOE in cooperation with the IAEA, was developed "to provide practical training in the implementation and operation of a national system of accounting for and control of nuclear materials that satisfies both national and IAEA international safeguards objectives."

Seventy people (including course attendees, lecturers, and equipment demonstrators) participated in the 16 day course. Nations represented included Belgium, Brazil, Canada, Egypt, France, India, Iraq, Israel, Italy, Japan, Korea, Luxembourg, Malaysia, Mexico, Pakistan, Poland, Sweden, Switzerland, Taiwan, Turkey, and Yugoslavia. Participants also came from the

co sponsoring organization - the IAEA in Vienna, Austria - and from the EURATOM Organization of the Commission of the European Communities in Luxembourg.

The course was conducted by Los Alamos National Laboratory, Battelle Pacific Northwest Laboratory, and the Exxon Nuclear Company, Inc. The course staff included G. Robert Keepin, Course Director, Bernardino Pontes, IAEA Scientific Advisor, and Course Coordinators, Charles Hatcher (Los Alamos), Douglas Reilly (Los Alamos), Richard Schneider (Exxon Nuclear), and Robert Sorenson (Battelle).

The principles and practical methods used in establishing and operating nuclear material accounting and control systems at bulk handling facilities were emphasized - particularly low enriched uranium (LEU) conversion and fuel fabrication plants. Also emphasized was the interaction between facility safeguards, national system safeguards, and international (IAEA) safeguards.

II. TECHNOLOGY TRANSFER

A. International Safeguards for Reprocessing Plants (E. A. Hakkila, R. G. Gutmacher, J. T. Markin, J. P. Shipley, and W. J. Whitty)

A Los Alamos-Sandia report entitled "Integrated International Safeguards Concepts for Fuel Reprocessing" that describes a design approach to integrating nuclear materials accounting and containment surveillance concepts for an internationally safeguarded fuels reprocessing facility was completed.^{55,56} The study evaluates safeguards concerns and IAEA verification activities relating to materials accounting and containment surveillance for the fuel storage chop leach, chemical separations, and plutonium nitrate storage areas of an AGNS-type plant. It shows that if the inspector verifies all operator near real-time accounting measurements, he can meet IAEA goals for detecting abrupt diversion in the chemical separations material balance area. Substantial probabilities for detecting diversion in the headend and product storage material balance areas can be attained, but IAEA abrupt diversion goals cannot be met rigorously. IAEA goals for detecting protracted diversion

cannot be met in any of the material balance areas because the goal quantity is only 0.05% of the annual throughput.

Enhanced containment surveillance concepts in the headend and product storage material balance areas were developed. This methodology is based on the probability of generating an alarm during a diversion occurring over a specified time. The evaluation suggests that a high level of assurance for detecting an abrupt diversion can be attained in both material balance areas. In addition, a high probability exists for detecting protracted diversion in the fuel storage-chop/leach material balance area. Protracted diversion remains a significant safeguards problem in the plutonium-nitrate storage material balance area. Although a methodology for combining the assurance for materials accounting and containment/surveillance systems was developed in this study, quantifying that assurance was not technically possible.

We attended a meeting of the Editorial Committee for the overview report of the International Working Group on Reprocessing Plant Safeguards (IWG-RPS) in Vienna.⁵⁷ This report summarizes the 2 yr of IWG-RPS

activities for the Director General of the IAEA and recommends future work of the group.

B. Joint US Federal Republic of Germany (FRG) Program (J. P. Shipley)

From June 15 to July 10, D. Seifried (KFK-ERG) was a guest scientist in the Safeguards Systems Group under the US-ERG cooperative program. He investigated and extended Los Alamos developed sequential analysis procedures. One such extension is based on the innovations sequence associated with the Kumar filter. In addition, a statistical test boundary based on the law of the iterated logarithm was determined, and the complete method was installed as an additional test in DECANAL.

The Los Alamos Safeguards Systems Group is participating in a US-ERG Near Real Time Accounting Working Group. The Los Alamos contribution includes analytical and empirical studies of false alarm rates, with special emphasis on the effect of changing loss patterns in specific tests.

C. Safeguards Cooperative Program with EURATOM, Ispra (A. L. Baker, J. T. Markin, E. A. Kern, D. B. Martinez, and J. P. Shipley)

Two Los Alamos staff members provided the Ispra safeguards group with the computer program DECANAL under the joint US-JRC-Ispra program for safeguards technology exchange. The Ispra safeguards group develops materials accounting techniques for use by EURATOM inspectors. This group needs to perform sequential analysis of materials accounting data to detect missing material and wants to use DECANAL for that purpose. When DECANAL was made operational on the JRC computer, the Ispra group was instructed in the theory and practical use of the program, and an interface program that allows DECANAL to be used with their materials accounting program ISADAM was written.

As a result of this trip, two possibilities emerged for continued interaction with the JRC-Ispra. The Ispra

group suggested that someone from Ispra visit Los Alamos to install the ISADAM program on our PRIME computer and to provide instruction. Also, Ispra representatives indicated an interest in exchanging information about methods for near real-time accounting in a laboratory facility.

D. Technical Support and Technology Transfer (Los Alamos Safeguards Staff)

Members of the Japanese Energy Research Institute (JERI) and PNC were invited to Los Alamos on new developments in near real-time accounting and processes inventory estimation in solvent extraction circuits.

Members of the DWK reprocessing design team of the ERG and of the Cogema reprocessing facility in La Hague, France, were invited on near real-time accounting for reprocessing facilities. Both teams expressed interest in joint test and evaluation experiments with the Los Alamos Safeguards Systems Group for demonstrating near real-time accounting.

During this reporting period, nine papers were presented at the Tenth Second Annual Meeting of the INMM,^{13,14,15} and a session on near real-time accounting was organized and chaired by a Los Alamos Safeguards Staff member. Two Los Alamos papers on safeguards technology were presented at a meeting of the Operations Research Society of America, the Canadian Operational Research Society, and the Institute of Management Sciences in Toronto, Canada.^{16,17} Los Alamos staff members also chaired two sessions of the Toronto meeting entitled "Energy Studies" and "Decision and Risk Analysis in the Energy Field." A Los Alamos paper on isotopic analysis by gamma-ray spectroscopy was presented at the Fifth Symposium on X- and Gamma-Ray Sources and Applications at Ann Arbor, Michigan.¹⁸

A member of the Los Alamos Safeguards Staff was selected general chairman for a safeguards measurements meeting to be sponsored by the American Nuclear Society and the INMM in 1983.

III. INSTRUMENT DEVELOPMENT FOR INTERNATIONAL SAFEGUARDS

A. Spent Fuel Verification Development

1. **Cerenkov Spent Fuel Instrument Upgrade** (N. Nicholson, E. Dowdy, and S. France). We are preparing an instrument for the IAEA to monitor irradiated fuel assemblies stored in an underwater storage pool. The instrument will be able to qualitatively image the Cerenkov light emitted by the assemblies as well as quantify the light intensity. Earlier instrumentation supplied to the IAEA was capable only of qualitative information.

A Varo Noctron V CVD has been selected as the basis for the improved Cerenkov viewing device (ICVD). The Varo has a generation 2 type image intensifier, 25 mm in diameter, and is less expensive than the discontinued Javelin model 222 that the IAEA is now using. The Noctron V provides a high resolution image, high gain, and very little barrel distortion. The add on package that converts this CVD to an ICVD has been designed. Weight estimates for the add on package assure that the overall package will weigh less than 4 kg. A digital readout will be located above the viewer and at the same focal plane as the image rather than in the field of view of the binocular viewer.

The add on package will contain the necessary optical components to transport a fraction of the incoming light through a prismatic mirror onto an aperture and through a field lens; it then will be reflected again onto the input of a photomultiplier tube. 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 the digitizing circuitry will be within the add on package, which will be powered by an external battery. Standard 25 mm single lens reflex lenses made by Zuiko (Olympus) will be used for the objective lenses and will include telephoto and zoom telephoto lenses. Figure 37 shows how the add-on package will fit onto the Varo CVD. Figure 38 shows the components of both the add-on package and the CVD, and Fig. 39 shows a more detailed sketch of these components. The major components for the two ICVDs have been ordered and most have arrived. Fabrication of the add on packages should begin shortly. The design of the ICVD allows removal of the add on package when not needed, and the Varo CVD can be used for qualitative viewing of the irradiated fuel.

2. **Ion or Neutron (ION-1) Electronics for Spent Fuel Verification** (J. K. Halbig, S. F. Klosterbuer, J. Caine, F. Duran, D. Peterson, and R. Whitehill). Two ION-1 units delivered to the IAEA in 1980 measure neutrons in the pulse counting mode and gamma ray dose in the current mode. Although some problems caused by printed circuit layout were encountered, the third unit delivered in March 1981 was modified to eliminate these problems. In addition to solving these problems, the modifications added memory, and the original 8 bit analog to digital converter (ADC) was replaced with a 12 bit ADC. The new board makes the unit extremely reliable, and the increased memory will be used to automate specific spent fuel measurements. One of the two original units was modified in Vienna in May 1981, and the other will be shipped to Los Alamos for modification when it is available. In addition to previously described functions,²⁶ the microprocessor is used to correct for internal electronics offsets caused by age, jarring in transit, or high humidity. The offset range that can be corrected is ± 80 to $\pm 10\%$ of full scale. Using the microprocessor significantly decreases the hardware complexity and increases the reliability of the instrument's analog channel, which is used to resolve currents to fractions of pico amperes.

Fig. 37. Outside view of the ICVD.

Fig. 38. Inside view of the ICVD.

Fig. 39. Schematic of the ICVD's functions.

3. Instrumentation for Spent-Fuel Measurements at GE Morris Facility (J. K. Halbig, S. F. Klosterbuer, P. Rinard, F. Duran, O. R. Holbrooks, and D. Peterson). The ION-1 units have been used worldwide by IAEA inspectors and development personnel. One instrument was used in June 1981 at the GE Morris spent fuel storage facility in Illinois during a joint exercise with IAEA personnel (the data are discussed in Part 3, Sec. 1.H.). This exercise used equipment

previously installed at the facility and new equipment designed to test methods for rapidly measuring fuel assemblies at a reactor storage pond.

The principal new piece of equipment was the square ring detector. Each of the four sides housed an ion chamber and a fission chamber, and all eight signals were connected to one of two electronics packages. The first, a summer box, allowed the signals to be read individually or in a summed mode by ION-1 or a

microprocessor-based data-acquisition unit. The second package interfaced these signals to the microprocessor-based data-acquisition unit used at previous exercises. The latter unit was used to rapidly acquire and log data for exposure vs detector response correlations and to study result uncertainties of the detector. This unit took data from the individual channels, summed them, and calculated averages.

The summer box was used with ION-1 to demonstrate the square-ring and portable-electronics packages to the IAEA. The results should determine if the summer box is necessary or if all signals can be summed electrically and fed directly to ION-1. At this exercise, personnel from IAEA inspector and development sections, International Safeguards Project Office, and Los Alamos were able to use the equipment and suggest hardware and software modification to improve ION-1.

A commercial cassette tape drive was interfaced to a Nuclear Data-60 analyzer to record the collected high-resolution spectra of the tops of the fuel assemblies in the pool taken through an air-filled collimator. For future exercises we plan to interface an HP-85 to the microprocessor-based data-acquisition unit.

B. Portable MCA (J. K. Halbig, S. F. Klosterbuer, T. D. Reilly, J. Caine, F. A. Duran, O. R. Holbrooks, D. Peterson, and R. Whitehill)

Two portable MCAs were completed and delivered to the IAEA in May. The MCAs feature a 1000-channel ADC with a conversion oscillator frequency of 32 MHz, internal high-resolution gamma-ray spectroscopy amplifier, NaI amplifier stabilizer, CRT display (3.5 × 5.5 cm), interactive keypad and 32-character display, magnetic tape storage, RS-232 serial interface, and internal battery power. The microprocessor-based MCA uses 17.5 k memory bytes for basic analyzer functions and 2.5 k memory bytes for specific measurements and can add up to 40 k bytes for future implementations. All interfaces are accomplished through the microprocessor software, and all calculations are done with a floating-point mathematics package with 7-digit precision and 10^{+38} range.

The MCA, designed for use with NaI and high-resolution detectors, provides preamplifier and bias power for these detectors. When operated with a 1.9 × 1.9-cm Bicron NaI detector with embedded americium seed and thermistor for stabilization, resolutions of 61-keV full width at half maximum (FWHM) at 661.6 keV

were achieved. No degradation in resolution was seen up to 20 000 cps, and detector resolution was quoted as 8.5% FWHM by the manufacturer. By using a good coaxial Ge(Li) detector and the internal high resolution gamma-ray (HRG) amplifier, resolution of 1 keV was achieved at the 185.7-keV peak of ^{235}U with a count rate of approximately 6000 cps.

The MCA is powered by an internal rechargeable battery pack of C size nickel cadmium batteries. If the batteries or charger fail, the batteries can be replaced with alkaline cells. All necessary voltages are generated internally, including those used by the NaI probe and HRG preamplifier. The bias for the NaI probe runs nominally at 750 V and is adjusted automatically by internal circuitry to keep the americium peak at the proper pulse height. The bias for the HRG detector is adjustable from about 500 to 3000 V; the polarity is selected by an internal jumper.

The small magnetic tape wafer drive, mounted on the front panel, and the serial port are used to read data from or into the MCA. The RS 232 serial port can be connected to a printer or central computer, and both data and commands can be transferred over this link.

The inspector-instrument interface is an extension of that implemented in the electronics unit for verification of spent fuel assemblies.⁶⁶ Operator interaction with the MCA is effected through the keypad, LCDs, CRT, and knobs on the front panel (Fig. 40). The current spectrum is displayed on the CRT, which has 256 vertical by 1024 horizontal resolution; the V SCALE knob is used to adjust this vertical scale. The MCA contains a 4 × 4

Fig. 40. Portable MCA.

keypad (Fig. 41), whose entries include the digits 0 to 9, INC, and DEC to increment or decrement any numerical value and to scroll the display; six upper-case commands—STRT, which is the most frequently used command; STOP, used to end or abort any sequence; ENTR, used to register a new numerical value that was keyed in or entered by using INC or DEC; and SHFT, which must be pressed before accessing any upper-case key.

Following is a brief description of the basic analyzer functions available in the initial software:

- STRT Acquire data for time specified by livetime.
- ROI Assign or change regions of interest. A maximum of 99 regions can be assigned, and regions may overlap.
- CALC Total or background subtracted areas of assigned regions and their associated uncertainties are calculated and displayed.
- STAT Examine or change instrument parameters. Those included in the STAT function are preset count time and elapsed time in livetime seconds, count rate, percentage deadtime, detector being used (HRG or NaI), amplifier input polarity, amplifier gain, high-voltage setting, upper and lower level discriminators, memory subgroup, and battery and digital supply voltages.
- TAPE Read spectra from the tape, write spectra to the tape, position the tape, erase the tape,

STAT 7	CALC 8	ROI 9	STRT
ENRH 4	DUMP 5	TAPE 6	STOP
1	2	3	ENTR
0	↓ DEC	↑ INC	SHFT

Fig. 41. MCA keypad labels.

and examine instrument settings for the last spectrum read. The full spectrum and all parameters—high voltage, gain, count time, inspector identifier, etc.—are recorded on the cassette. The identifier and the date and time uniquely identify the spectrum.

DUMP Write the current spectrum and instrument setting to the serial port.

In addition, the number of counts in any channel is available when the MCA is waiting for a new command to be entered.

The seventh command is the first automated measurement implemented:

ENRH Perform ^{235}U enrichment measurement. The calibration constants used in the calculation are obtained either by entering previously determined constants from the keypad or by prompting the user through a calibration procedure using two known samples. Once the calibration constants are known, the user is prompted through the steps to determine the enrichment of an unknown. Once the calibration constants are entered, each measurement of an unknown is accomplished by a single button push. Results include the enrichment estimate with its calculated uncertainty.

The MCA delivery included demonstrating and training IAEA development and inspector personnel on MCA use. We also used the instrument with the IAEA project officer to make enrichment measurements at the Seibersdorf Laboratory near Vienna (see Tables XIV and XV).

C. Coincidence Measurements of FFTF Breeder Fuel Subassemblies (G. W. Eccleston, J. Foley, M. Krick, H. Menlove, A. Ramalho*, and P. Goris†)

A fast breeder reactor (FBR) fuel assembly coincidence counter was designed, constructed, and used to measure FFTF subassemblies. The data were collected in a field exercise at the HEDL in Richland, Washington.

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†Hanford Engineering Development Laboratory, Richland, Washington.

TABLE XIV. Enrichment Test of Mini-MCA 80-4^a

Sample Enrichment	No.	Window 1 ^d Counts	Window 2 ^d Counts	Measured Enrichment	Count Time
1.2	6	38 769 ± 197	14 482 ± 120	* ^c	500 s
9.0	35	129 183 ± 359	15 221 ± 123	* ^c	500 s
2	10	9175 ± 96	2672 ± 52	1.99 ± 0.06	100 s
2	10	18 279 ± 135	5415 ± 74	1.95 ± 0.04	200 s
3	20	24 086 ± 135	5908 ± 77	3.03 ± 0.04	200 s
5	31	32 661 ± 181	5857 ± 77	4.93 ± 0.05	200 s
6	32	38 119 ± 195	6199 ± 79	5.99 ± 0.05	200 s
7	33	42 496 ± 206	6085 ± 78	6.99 ± 0.05	200 s
8 ^{b,c}	34	46 850 ± 216	5192 ± 72	8.28 ± 0.05	200 s
8 ^c	34	16 128 ± 127	1631 ± 40	6.33 ± 0.07	93 s
10 ^{b,c}	36	28 070 ± 168	2361 ± 49	10.50 ± 0.82	101 s
8 ^{b,c}	34	23 383 ± 153	2619 ± 51	8.248 ± 0.077	101 s

Calculated calibration constants:

$$A = 4.375 \times 10^{-2}$$

$$B = -7.570 \times 10^{-2}$$

^aUsed with Los Alamos small detector and battery charger plugged in.^bContainer on edge—different geometry.^cContainer had only small amount of material.^dWindow 1—Channels 342 to 484

Window 2—Channels 601 to 758

Gain = 16

^eUsed as calibration source.

The FBR fuel assembly coincidence counter, shown in Fig. 42, contains 12 ³He tubes, with active length of 121.9 cm, equally spaced in a polyethylene ring. Initial tests of the counter were completed with a small californium source. The die-away time (31 μs), deadtime (2.3 μs), and detection efficiency (8.4%) were measured with the californium source located at the midplane and positioned at the counter center. The vertical response profile, shown in Fig. 43, is flat over about 80 cm of the active counter length. The response profile at the counter base was remeasured with a set of boiling-water reactor (BWR) fuel rods placed in the end. The rods reflected neutrons, similar to the blanket regions at the ends of the FBR assembly, and increased the region of flat response in the counter.

HEDL made available four FFTF fuel subassemblies for measurements in the FBR coincidence counter: Table

XVI lists the fuel material in each subassembly. The subassemblies were hexagonal and contained 217 fuel pins. The central fuel region consisted of mixed oxide with depleted uranium outside this region.

Measurements were collected after centering the mixed oxide fuel region of a subassembly in the counter. Two sets of data were collected, corresponding to all 12 tubes connected or with only every other tube connected. Data were obtained from only half the tubes to lower the count rate and reduce deadtime effects associated with the coincidence circuits, which possibly could have produced bias effects. A plot of the coincidence rate vs plutonium content for the measured subassemblies is shown in Fig. 44. Data analysis and corrections for multiplication and americium loadings are in progress.

TABLE XV. Enrichment Test of Mini-MCA 80-3^a

Sample Enrichment	No.	Window 1 ^c Counts	Window 2 ^c Counts	Measured Enrichment	Count Time
1.2	6	41 145 ± 202	18 421 ± 135	^{ad}	500 s
9	35	142 944 ± 378	19 274 ± 139	^{ad}	500 s
2	10	10 089 ± 100	3452 ± 59	2.05 ± 0.05	100 s
3	20	12 963 ± 114	3793 ± 62	2.98 ± 0.06	100 s
5	31	17 949 ± 134	3602 ± 60	5.01 ± 0.06	100 s
6	32	20 727 ± 144	3748 ± 61	6.01 ± 0.07	100 s
7	33	23 539 ± 153	3858 ± 62	7.04 ± 0.07	100 s
7	33	23 131 ± 152	3848 ± 62	6.89 ± 0.07	100 s
7	33	23 373 ± 153	3685 ± 60	7.07 ± 0.07	100 s
8 ^b	34	25 685 ± 160	3569 ± 59	8.03 ± 0.07	100 s
10 ^b	36	30 686 ± 175	3623 ± 60	9.94 ± 0.08	100 s
6	32	20 330 ± 143	3765 ± 62	5.85 ± 0.06	100 s
6	32	20 629 ± 144	3920 ± 63	5.88 ± 0.06	100 s
6	32	20 717 ± 144	3783 ± 62	5.99 ± 0.06	100 s

Calculated calibration constants:

$$A = 3.876 \times 10^{-2}$$

$$B = -5.401 \times 10^{-2}$$

^aUsed with modified B-SAM detector and battery charger plugged in.^bContainer on edge—different geometry.^cWindow 1—Channels 423 to 548

Window 2—Channels 575 to 700

Gain = 16

^dUsed as calibration source.

Fig. 42. FBR coincidence counter.

Fig. 43. FFTF assembly counter vertical response profile.

TABLE XVI. FFTF Fuel Subassembly Material Loadings

Assembly Number	16 483	16 471	8195	8281
Mixed oxide (g)	37 428	37 436	37 669	37 589
Plutonium (g)	7395.86	8385.74	9108.35	9746.86
Plutonium isotopic (wt%)				
^{238}Pu	0.06	0.05	0.06	0.07
^{239}Pu	86.35	86.85	86.47	86.47
^{240}Pu	11.82	11.62	11.67	11.89
^{241}Pu	1.56	1.27	1.61	1.43
^{242}Pu	0.21	0.21	0.19	0.22
^{241}Am (g) 1/1/80	29 ± 3	---	34 ± 3	---

Fig. 44. Coincidence count rate vs ^{240}Pu effective loading of FFTF fuel assemblies.

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