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ABSTRACT

This report presents the status of the Nondestructive Assay R&D program of the LASL Nuclear Safeguards Research Group, R-1, covering the period September-December 1975. The topical content of this program report is summarized in the table of contents.

CONTENTS

I. NONDESTRUCTIVE ASSAY APPLICATIONS AND RESULTS

A. Holdup Measurements at the Kerr-McGee Nuclear Plutonium Facility, Crescent, Oklahoma (J. W. Tape, D. A. Close, and R. B. Walton)

An ERDA team consisting of members of TSO, Brookhaven National Laboratory; and members of Group R-1, Los Alamos Scientific Laboratory (LASL), is planning to independently verify the measured plutonium holdup at the Kerr-McGee Nuclear Plutonium Facility in Crescent, Oklahoma. In support of this effort instrument development, calibration, and general planning have been undertaken at LASL. A visit was made to the Kerr-McGee facility on December 15-16, 1975, to aid in the planning of the measurement program.

Gamma-ray measurements of plutonium will be made using NaI(Tl) shielded probes and SAM-II electronics. The organization of the gamma-ray equipment and calibration is now in progress.

Passive neutron measurements with SAM electronics will involve the use of two new detectors: the SNAP-II (see Section II.B) and a slab detector consisting of five ^{3}He tubes encased in polyethylene. The slab detector is designed to measure the neutron field near the center of a room with high sensitivity in an attempt to set an upper limit on the holdup for the entire room. Model calculations using Monte Carlo techniques are being employed to determine the neutron flux at the room center as a function of room size, wall thickness, and source distribution. Experimental tests in which discrete plutonium sources were distributed around a large room indicate that the detector is sensitive to small quantities of plutonium, but that obtaining an absolute calibration will be difficult. However, because the fast neutrons are very penetrating and because extraneous materials mixed with PuO_2 increase the total neutron yield, this technique will be effective in establishing an upper limit for the plutonium holdup.

The in-plant measurements are currently planned for February 1976.

B. Random Driver (D. A. Close and T. L. Atwell)

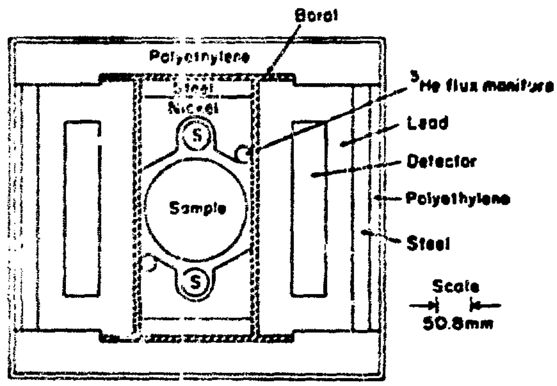

Fig. 1
Top view of Random Driver Mod-III.

A series of Monte Carlo calculations has been performed for the Random Driver Mod-III.[1] The geometry of this instrument is given in Fig. 1. The primary objective was to determine how to make the system relatively insensitive to neutron self-shielding in samples of high-fissile density. This effect was highly dependent on both the amount of moderating material in the sample and the container material. Minimizing the self-shielding effect required that a hard irradiation spectrum be maintained over a variety of sample types and containers. For these calculations, an HTGR fuel mixture was assumed: 322.58 g U (93% ^{235}U), Th/U = 4, gC/gU = 10 ρ = 1.5 g/cc. The HTGR fuel had a thin silicon-carbide coating, but within the accuracy of the calculations silicon had no effect, so the silicon-carbide coating was replaced with pure carbon. This material was assumed to be in a 3.5-ℓ aluminum can and a 3.8-ℓ polyethylene bottle. The following three effects were studied: (1) the effect of changing the amount of boral in the irradiation cavity liner; (2) the effect of using a boral-cadmium liner; and (3) the effect of putting an additional boral liner in front of each source.

Figure 2 shows how the calculated number of fissions in the fissile fuel caused by neutrons of energy E_n varied with the amount of boral in the irradiation cavity liner. The fuel mixture was assumed to be in a 3.5-ℓ aluminum can. The solid curve in Fig. 2 represents the standard 0.635-cm-thick boral liner, the dashed curve represents no boral, and the dash-dot curve represents a liner with five times as much boral as the standard liner. The error bars are

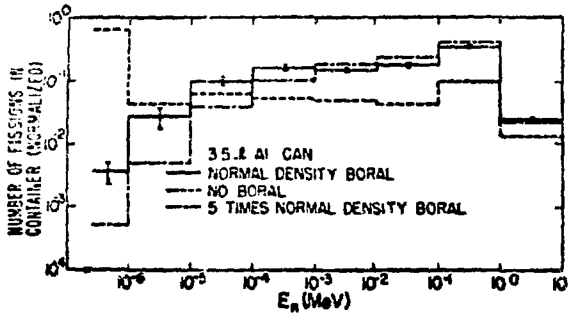

Fig. 2

Number of fissions in the ^{235}U material caused by neutrons of energy E_n. The material was assumed to be in a 3.5-ℓ aluminum can. The MCN results for a standard boral liner (0.635 cm), a no-boral liner, and a liner with five times as much boral are shown.

shown only for the standard boral liner, but they are representative of the other two cases.

Figure 3 shows how the number of fissions in the ^{235}U fuel caused by neutrons of energy E_n varied with the amount of boral in the irradiation cavity liner. The fuel mixture was assumed to be in a 3.8-ℓ polyethylene bottle. The solid curve represents the standard 0.635-cm-thick boral liner, the dashed curve represents no boral, and the dash-dot curve represents a liner with five times as much boral as the standard liner. Figures 2 and 3 show that the added boral not only absorbs the low-energy neutrons, but increases the number of fissions in the high keV energy region because of neutron scattering and reflection.

The effect a composite boral-cadmium irradiation cavity liner (0.483-cm boral plus 0.152-cm cadmium) had on the number of fissions in the material caused by neutrons of energy E_n is shown in Fig. 4. Figure 5 shows the number of fissions in the ^{235}U fuel caused by neutrons of energy E_n when an additional 0.635 cm of boral was placed in front of each source. These latter calculations assumed the material to be in a 3.8-ℓ polyethylene bottle.

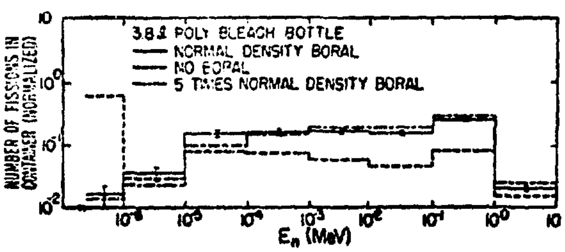

Fig. 3

Number of fissions in the fissile material caused by neutrons of energy E_n. The material was assumed to be in a 3.8-ℓ polyethylene bottle. The MCN results for a standard boral liner (0.635 cm), a no-boral liner, and a liner with five times as much boral are shown.

The standard boral liner dramatically reduced the effect of low-energy neutrons ($0 \le E_n \le 1$ eV) when the fissile material was in a 3.5-ℓ aluminum can (see Fig. 2). There was a further decrease in the effect of low-energy neutrons ($0 \le E_n \le 10$ eV) when five times as much boral was used.

From Fig. 3, it is seen that the standard boral liner greatly reduced the effect of low-energy neutrons ($0 \le E_n \le 1$ eV) when the material was assumed to be in a 3.8-ℓ polyethylene bottle. There was not a significant change when the boral liner was replaced with one having five times as much boral.

A comparison of Fig. 3 and Fig. 4 shows that for a 3.8-ℓ polyethylene bottle, the composite boral-cadmium liner does not significantly change the calculations from those for a standard boral liner. Similarly, a comparison of Fig. 3 and Fig. 5 indicates no improvement when additional boral is placed in front of each source. However, for irradiation cavities designed to accommodate larger diameter samples, such as Random Driver Mod-IV[2] and -V, experimental studies indicate that additional boral in front of the sources does minimize the number of low-energy neutrons.

These studies have shown that boral can be used very effectively in making the Random Driver and related instruments less sensitive to neutron self-shielding effects, and consequently in making the assays less matrix dependent.

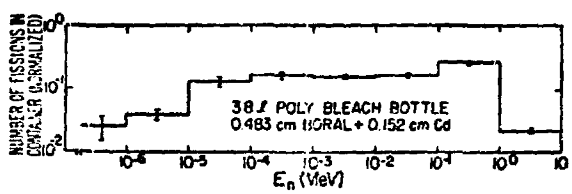

Fig. 4

Number of fissions in the ^{235}U material caused by neutrons of energy E_n. The material was assumed to be in a 3.8-ℓ polyethylene bottle with a composite 0.483-cm boral/0.152-cm cadmium irradiation cavity liner.

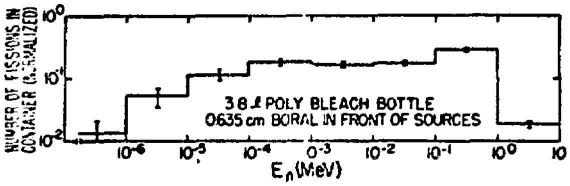

Fig. 5

Number of fissions in the ^{235}U material caused by neutrons of energy E_n. The material was assumed to be in a 3.8-ℓ polyethylene bottle with an additional 0.635-cm of boral in front of each source.

II. INSTRUMENT DEVELOPMENT AND MEASUREMENT CONTROLS

A. The CMB-8 Uranium NDA Material Balance System (T. L. Atwell, T. R. Canada, N. Ensslin, and H. R. Baxman*)

The CMB-8 uranium reprocessing procedures encompass a complex multistep process, involving a variety of material forms and a wide range of special nuclear material (SNM) concentrations. The old associated accounting system is a handwritten, paper-storage transaction procedure, summaries of which are available on a biweekly basis.

A nondestructive assay (NDA) material balance system has recently been installed that measures the uranium content of the solids and solutions within the plant at various points in the process.[3] These measurements are used as the input data for the accounting transactions that are now stored on-line in the system computer and are available for immediate recall by the plant personnel. The physical layout of the system hardware is shown in Fig. 6.

The cost-effective computer system consists of a 28-k PDP-11/10 minicomputer, a 256-k dual floppy disk for mass storage, a hard-copy line printer (not shown), a high-speed paper tape reader/punch for media transfer, and two CRT data display terminals. The terminal shown in Fig. 6 is used for data acquisition, data reduction, and control of the assay instrumentation. The other terminal is dedicated to the materials management and accounting system. The software for this highly interactive time-sharing system is programmed in Multi-User Basic and runs under the RT-11 Operating System.

The NDA system consists of a Random Driver[4,5] and a uranium solution assay device (USAD). The former is an active neutron interrogation instrument that measures the ^{235}U mass (50-5000 g) present in solid compounds and metal alloys by counting coincident fission neutrons. The USAD measures either the ^{235}U or the total uranium density in solution by state-of-the-art gamma-ray assay techniques over a range of 10 ppm to 400 g/ℓ. Through the computer interfacing, the system actively questions the assayist, checks his responses, detects errors made in the assay procedures, and performs a variety of diagnostic checks on instrument performance.

*LASL Group CMB-8.

Fig. 6
Recently installed computer system and NDA instrumentation for LASL's CMB-8 uranium reprocessing operation. The Random Driver and USAD assay instruments are shown on the far left and far right, respectively.

The accounting program closely parallels that of the laboratory—the operator can

(1) enter a transaction at the terminal, where (a) the transaction goes into a list of transactions, (b) "to" material category is incremented; "from" material category is decremented, (c) running sums of total mass, material in or out of process (MIP or MOP), and MUF are updated;

(2) call a list of all transactions sorted by account number, receipt area, material category;

(3) call a list of ending balances in each material category sorted as above;

(4) get sums of material in all material categories sorted by receipt areas;

(5) clear or close-out (clear but save MUF) a receipt area;

(6) call for material sum and MUF by receipt area and get total MUF.

Work is proceeding on the study of experimental biases, on general software modifications, and on the drafting of a detailed operation manual.

B. Upgrading of Snap Neutron Detector SNAP II (H. O. Menlove, T. L. Atwell, and A. Ramalho*)

Recent modifications have been made on the SNAP portable neutron probe[6] to improve the following characteristics: (1) overall efficiency, (2) flat neutron energy response, (3) flat vertical position response, (4) capability to measure the incident neutron energy, and (5) close coupling to plutonium fuel rods and cans. For most applications the detector is used to measure neutrons for (α,n) reactions from PuO_2 and UO_2 and spontaneous fission from ^{240}Pu and ^{242}Pu. The average energy of these neutrons is ~1-2 MeV. Thus the efficiency should be optimum for this energy range, whereas the background neutrons entering the shield should have somewhat lower energy because of multiple scattering in the room. The previous detector moderator wall thickness of 2.5-cm CH_2 was too thin for optimum counting of the signal neutrons.

The primary modifications to the SNAP unit were to increase the central moderator core (CH_2) from 7.6- to 12.7-cm diameter and to increase the active length of the ^3He detectors from 12.7 to 20.3 cm. The overall diameter of the shield was increased from 22.9 to 24.1 cm and the length was reduced from 33.0 to 30.5 cm to better distribute the shielding without increasing the weight.

Figure 7 gives a plot of relative efficiency vs energy for the different detector configurations listed in Table I. The neutron source standards were positioned 30 cm from the detector center. The bottom curve (A) represents the original SNAP configuration and the upper curves represent the modifications listed in Table I.

The total efficiency increase of a factor of 4.51 resulting from the modifications (SNAP-II) makes it possible to use the instrument in applications with lower amounts of plutonium. The SNAP-II version also has three times less efficiency drop between 1.5 MeV and 0.5 MeV making the detector less sensitive to small amounts of hydrogen in the sample matrix or container.

In addition to increasing the total efficiency, the effect of increasing the active length of the ^3He tube is to center the maximum response with the center of the SNAP shield as well as giving a broader flat region.

The primary reason for the smaller core (12.7 cm) in the original unit was to leave room for the exterior shielding without going to a larger diameter and thus heavier unit. The shielding effectiveness was

Visiting scientist from the International Atomic Energy Agency (IAEA).

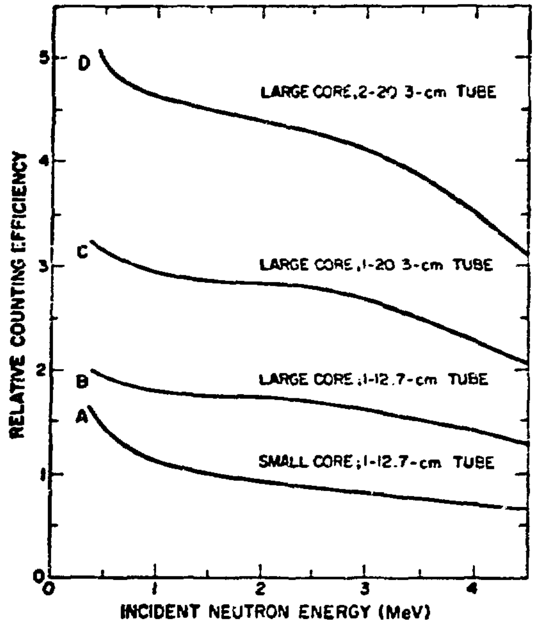

Fig. 7
Relative efficiency as a function of energy for different core and ^3He tube configurations in the SNAP neutron detector.

measured by the front-to-back ratio (F/B) for the neutron sources. Figure 8 shows the F/B ratio for the original shield and the modified unit with a 25.4-cm-diameter shield. It can be seen that for this larger diameter the F/B ratio of the new version was better than the original unit but there was a weight increase. The modified SNAP-II external diameter was reduced to 24.1 cm to lower the weight and yet give a F/B ratio equivalent to the original shield.

To determine the average energy of the incident neutrons, the F/B ratio can be used as illustrated in Fig. 9. In addition, the ratio of the front-to-back ^3He tube (f/b) in SNAP-II can be used to estimate the average energy as shown in Fig. 9. This tube ratio is not as sensitive as the F/B shield ratio; however, it is not as subject to background problems and it is available with the normal assay measurement.

To reduce the shielding weight on SNAP-II and make it possible for close coupling to large containers, the front lip (top and bottom) of the CH_2 shield has been removed as shown in Fig. 9. This resulted in ~10% reduction in weight from the original SNAP configuration. Also a small hole (1.9-cm diameter) has been drilled through the interior to give higher efficiency counting for FBR- and LWR-plutonium recycle fuel rods as well as other small

TABLE I

EFFICIENCY VS NEUTRON ENERGY FOR SNAP MODIFICATIONS

Curve	CH_2 Core diam (cm)	Number of ^{3}He Tubes	Active Length (cm)	Relative Eff. At 1.5 MeV	Eff. Ratio 1.5 MeV / 0.5 MeV
A	2.6	1	12.7	1.00	.70
B	12.7	1	12.7	1.74	.90
C	12.7	1	20.3	2.85	.90
D	12.7	2	20.3	4.51	.91

samples. This close coupling also makes possible neutron coincidence counting using SNAP-II to separate the spontaneous fission neutrons from the (α,n) neutrons.

C. Photoneutron (Sb-Be) Assay System for Small Samples (M. S. Krick and H. O. Menlove)

The photoneutron assay system described previously[7] was updated by the installation of two five-channel-amplifier NIM modules designed by J. E. Swansen of Group R-1 and described previously.[7] The photoneutron system used 30 ^{3}He proportional counters arranged in 10 groups with 3 tubes per group. Each group of tubes was connected to a

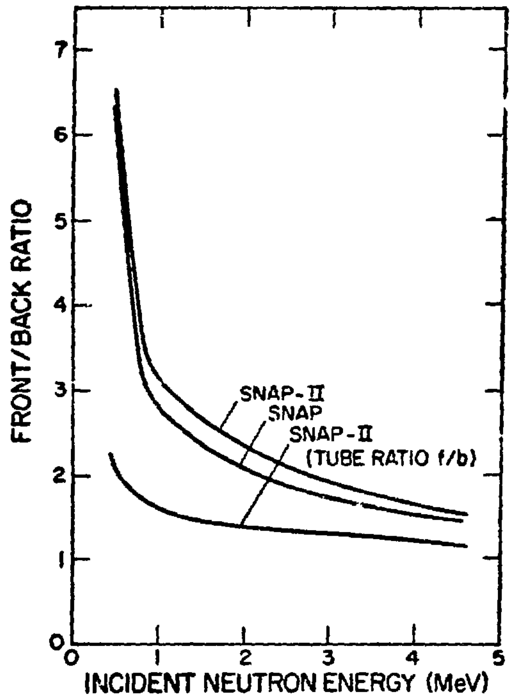

Fig. 8
Shielding effectiveness vs incident neutron energy and neutron energy measuring capability of SNAP-II.

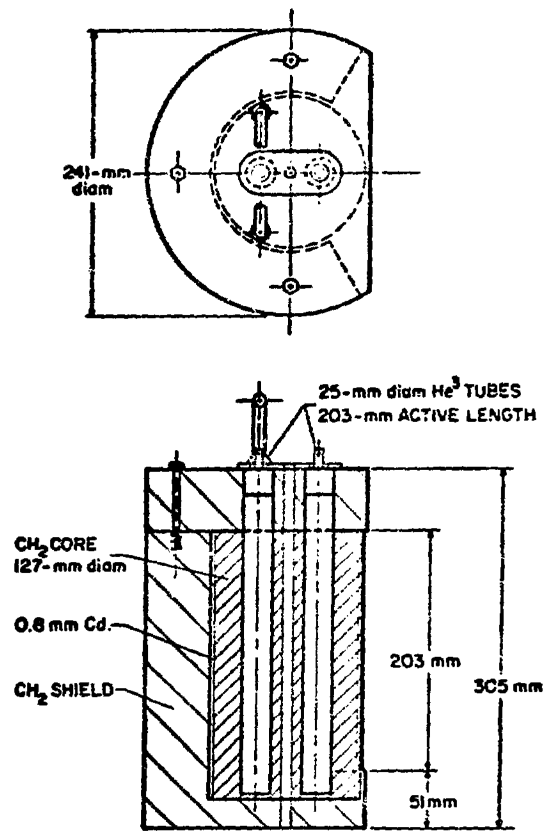

Fig. 9
Schematic diagram of SNAP-II portable neutron detector with two ^{3}He tubes for increased efficiency.

charge-sensitive preamplifier and to one channel of a five-channel NIM module. The pulse shaping time constant, the amplifier gain, and the discrimination level were set independently for each channel. The logical OR output from the second of the series-connected five-channel amplifiers was connected to a scaler. The amplifiers have performed flawlessly and require only two single-width slots in a NIM bin.

The sample size dependence of the assay system for small samples was studied by measuring the positional sensitivity of the system for a small sample (1.8 g) of 93.1% enriched ^{235}U. The response vs position is shown in Fig. 10 where the curve is a quadratic least-squares fit to the measured response normalized to 100% at the position of maximum response. Integrating this curve over appropriate intervals produces the curves of Fig. 11. The upper curve shows the sample height dependence for a given mass when the sample is centered at the position of maximum response. The lower curve shows the dependence when the bottom of the sample is at the position of maximum response. Both curves are normalized to 100% at zero height.

D. ^{252}Cf-Based Hydrogen Analyzer (D. A. Close, R. C. Bearse*, and H. O. Menlove)

After the basic design features of the instrument were established and tested,[9] additional computer calculations were performed to determine if further improvements could be made. These calculations investigated four possibilities: (1) using a ^{10}B sleeve around the source cylinder; (2) using a ^{10}B sleeve around the sample-detector cylinder; (3) positioning the sample directly between two detectors; and (4) using different combinations of materials.

Visiting staff member from the University of Kansas, Lawrence, Kansas.

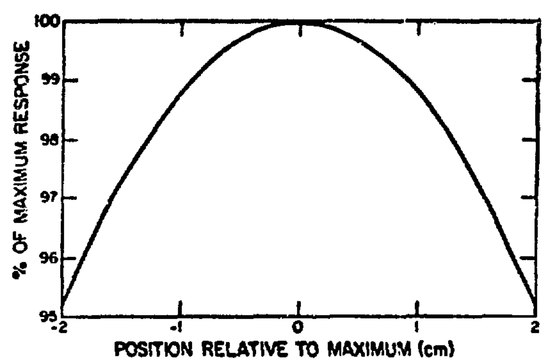

Fig. 10
Dependence of Sb-Be small sample assay on sample position.

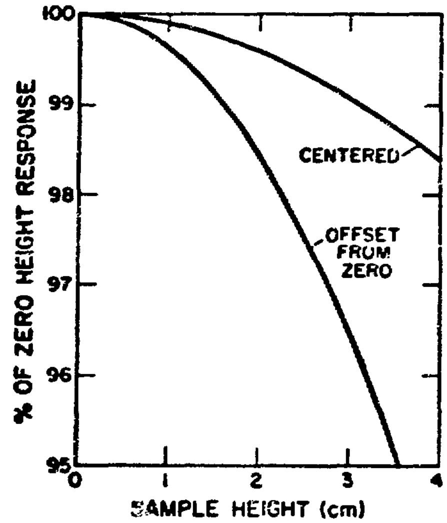

Fig. 11
Dependence of Sb-Be small sample assay on sample height

It was proposed that a boron sleeve around the source cylinder or around the sample-detector cylinder might increase the net signal-to-background ratio. Lower energy neutrons have a higher probability of having a collision with hydrogen, and further, have a higher probability of being detected by a ^{3}He detector. Ideally then, boron, having a high thermal neutron capture cross section, would remove the low-energy neutrons contributing to the background and would not remove the slightly higher energy neutrons that would be slowed by collisions with hydrogen. Thus signal rates might not be greatly affected while background rates might be decreased. This should be true particularly for the beryllium source cylinder, since it is the best neutron moderator.

Calculations were performed for two thicknesses of ^{10}B, 3 mm and 2 mm. The 3-mm thickness was the maximum that could be inserted around either of the cylinders without altering the design of the instrument. No differences were noted between these two thicknesses, so further calculations were performed with 2-mm-thick ^{10}B sleeves.

First, a ^{10}B sleeve was assumed to be placed around the sample-detector cylinder, and several combinations of cylinder materials were investigated. The results are shown in Table II and are to be compared with those for the Ni(2)-Ni system (0.486 ± 0.112). When the 2-mm ^{10}B sleeve was assumed to be around the beryllium source cylinder

TABLE II

MONTE CARLO COMPARISON OF VARIOUS CYLINDER MATERIALS AND GEOMETRIES[a]

Sample cylinder material	Number of detectors	Source cylinder material	Net signal/ background
Fe	3	Be	0.144±0.015
Ni	2	Ni	0.522±0.115
Be	2	Ni	0.273±0.052
Be	2	Be	0.106±0.015
Be	2	Fe	0.088±0.011
Fe	2	Be	0.414±0.076

[a]A 2-mm ^{10}B sleeve was assumed around the sample-detector cylinder.

with an iron sample-detector cylinder, there likewise was no improvement in the net signal-to-background ratio.

A calculation was performed on the Fe(2)-Be arrangement with a ^{10}B sleeve around the iron cylinder, and with the sample assumed directly between the two ^{3}He detectors. This geometry did not increase the net signal-to-background ratio.

Calculations were made to see if replacing the iron reflector with nickel would yield higher net signal-to-background ratios. These calculations assumed Ni(2)-Ni geometry with no sleeves. The iron reflector system proved better than the total nickel system. It was found that the total iron system was not as good as the Ni(2)-Ni system in the iron reflector. We were unable to improve our original system.

E. Plutonium Solution Analysis System (J. L. Parker)

Work has continued on the plutonium solution analysis system based on high-resolution gamma-ray spectroscopy, to be installed in the ash-leach line at the LASL plutonium recovery plant. The sample holder shield and the detector shielding have been designed and fabricated, plutonium solution standards have been acquired from LASL Group CMB-1, and the system has been assembled in a temporary glovebox in the Group R-1 laboratories. Studies are underway to determine the optimum method of analysis and the precisions and accuracies to be expected for various solution concentrations. Using a 30-m sample, it appears that an accuracy in the ^{239}Pu determination of ≤1.0% at the 1σ level for solutions with concentrations ≥0.5-mg Pu/mℓ will be possible, the complete analysis taking approximately one-half hour. For solutions with concentrations ≥2.0-mg Pu/mℓ the accuracy should be ~0.5% at the 1σ level. A transmissions measurement with an external source will guarantee a high degree of immunity to error introduced by varying matrix density. The ^{241}Pu determination will not be as accurate for the low-burnup plutonium expected due to the rather low intensity of the 148-keV gamma ray upon which the analysis must be based. Relative accuracies of a few percent can be expected. Americium-241 concentrations will also be available with accuracies of a few percent.

The initial analysis system is intended primarily for ^{239}Pu, ^{241}Pu, and ^{241}Am determinations for plutonium concentrations between 0.5-g Pu/ℓ and 10.0-g Pu/ℓ, though it will be capable of ^{239}Pu, ^{241}Pu, and ^{241}Am determinations for any concentration ≥0.5-g Pu/ℓ. Future modification will make it possible to measure the higher concentrations (≥ 100-g Pu/ℓ) by the transmission ratio method that measures total plutonium.[10]

F. A Simple Method for Neutron Coincidence Counting (J. E. Swansen, N. Ensslin, D. A. Close, and H. O. Menlove)

A method for neutron coincidence counting of plutonium based on a single one-shot time delay of fixed duration is being investigated. Such a circuit would be very useful as a portable instrument for passive assay of spontaneous fission neutron emitters, such as ^{240}Pu. A similar circuit has been developed by Birkhoff, et al., at EURATOM.[11,12]

Figure 12 illustrates the application of this circuit with a neutron well counter. The counter contains a ring of moderated ^{3}He thermal-neutron counters,

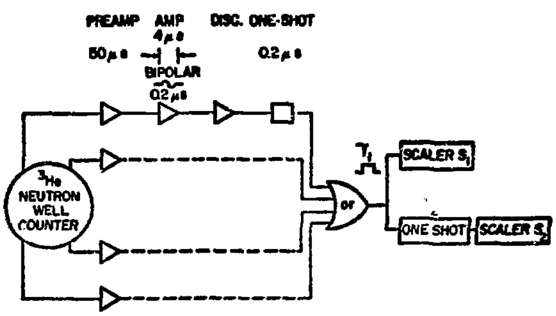

Fig. 12
Schematic diagram of neutron coincidence detector and associated electronic circuitry for simplified coincidence counting.

and has an absolute efficiency e of about 25% for a source placed inside it. Fission neutrons of multiplicity ν are detected in bursts of $e\nu$ neutrons that die away exponentially in time with a time constant of 30 μs. Superimposed on these fission events is a random background, which may be much larger than the spontaneous fission rate.

For a circuit similar to that in Fig. 12, but with only one input amplifier, the input deadtime T_1 to the scaling circuit is equal to the amplifier deadtime. One of the advantages of multiple input amplifiers, as illustrated in Fig. 12, is a substantial reduction of T_1, which enhances the counting efficiency for fission neutrons. The deadtime T_2 of the one-shot should be on the order of the die-away time of the detector. For nonupdating one-shots, and for a sample that has no correlated events, the total number of counts is

$$S_1/1 - S_1T_1 = S_2/1 - S_2T_2 \ , \qquad (1)$$

where S_1 is the count in scaler 1, and S_2 is the count in scaler 2.

If correlated fission events are present, the two scaler readings, corrected for background as above, will not be identical. To first order, the fission events are proportional to $S_1/1\text{-}S_1T_1 \ - S_2/1\text{-}S_2T_2$. This would be exact only if $T_1 = 0$ and if fission events were always counted in S_1 and always missed in S_2 because of the duration of T_2. Work is now in progress on the equations required for accurate analysis at high counting rates. The aim is to find a circuit and formula that will provide an assay result that is linear with the amount of fissile material and independent of background rate.

G. Delayed-Neutron Energy Spectra (A. E. Evans* and M. S. Krick)

Equilibrium delayed-neutron energy spectra have been measured from the fast fission of ^{235}U, ^{238}U, and ^{239}Pu. The equilibrium was established by irradiating and counting the samples in a pulsed mode with a 100-ms period. The beam was pulsed on for 35 ms and, after a 15-ms cooling period, the delayed neutrons were counted for 40 ms. The pulsing was accomplished by a sychronized electrostatic deflector and a mechanical shutter previously developed for small sample assay by delayed-neutron counting.[13] Since the longest-lived delayed-neutron group has approximately a 200-ms half-life and since this group contributes only a few percent to the total delayed-neutron yield, this pulsed mode essentially produced an equilibrium spectrum.

*LASL Group R-5.

The irradiating neutrons were produced with the LASL 3.75-MV Van de Graaff accelerator using the $^7Li(p,n)^7Be$ reaction. For the fissile-sample irradiations 25 μA of 2.15-MeV protons were used with a 2-mg/cm^2 lithium target to produce neutrons with energies ranging from 80 to 420 keV and with an intensity of approximately 10^9 n/sr-s in the forward direction. For the ^{238}U sample, the accelerator was operated at 3.5 MV to produce 1.6- to 1.8-MeV neutrons.

The samples were metal plates approximately 75-mm square covered with cadmium. The samples' masses and enrichments are shown in Table III.

The Shalev neutron spectrometer[14] used for these measurements was a gridded ^3He proportional counter filled with 5 atm of ^3He and 2 atm of argon. The sensitive volume was approximately 5-cm diameter by 20-cm long. The spectrometer tube was shielded by 1.9-cm lead, 0.5-mm cadmium, and a 5-mm-thick pressing of 50% aluminum and 50% boron (enriched to 85% in ^{10}B). An additional 3.8-cm lead was placed between the sample and detector to reduce detector pileup from early fission-product gamma radiation. The spectrometer was approximately 20 cm from the sample.

Pulse shape analysis was used to eliminate the continuum of ^3He recoil pulses from the spectrometer response. Since the track lengths of protons from the $^3He(n,p)$ reaction were longer than those of the recoiling ^3He nuclei, the rise times of the ^3He recoil pulses were shorter than those from the desired reaction and were discriminated against by the use of an ORTEC 458 pulse shape analyzer.

The energy dependence of the spectrometer response was measured using approximately monoenergetic neutrons between 50 keV and 2.0 MeV produced by the $^7Li(o,n)^7$ and $^{51}V(p,n)$ reactions.

TABLE III

SAMPLE DATA FOR DELAYED-NEUTRON SPECTRAL MEASUREMENTS

Sample	Total Mass (g)	Enrichment (%)
^{235}U	111	97
^{238}U	229	99.7
^{239}Pu	136	98

The calibration runs were normalized to the response of a modified ^{3}He long counter[15] that has an energy-independent response over the energy range of interest. A measured monoenergetic response function is shown in Fig. 13. The full-energy-peak response of the spectrometer as a function of neutron energy with shielding installed is shown in Fig. 14.

The uranium and plutonium delayed-neutron pulse-height distributions were accumulated for approximately 50 h per sample. The raw pulse-height distribution for ^{235}U is shown as the upper curve in Fig. 15. Two operations were performed to convert the pulse-height distributions to energy distributions. The first consisted of determining for each monoenergetic spectrum the fraction of the full-energy-peak counts that were contained in the region between the thermal peak and the full-energy peak; this energy-dependent fraction was used to approximately unfold the pulse-height distribution by the subtraction of an appropriate amount from each channel. The subtracted quantity is shown as the lower curve in Fig. 15 for ^{235}U. The second operation was the division of the subtracted pulse-height distribution by the efficiency curve of Fig. 14 to obtain the energy distribution of the delayed neutrons.

The delayed-neutron spectra for ^{235}U, ^{238}U, and ^{239}Pu are shown in Figs. 16, 17, and 18, respectively. As a visual aid, the spectra were smoothed with the weighting function

$$x_m = \frac{1}{2.5}\left[x_m + \frac{1}{2}(x_{m-1} + x_{m+1}) + \frac{1}{x}(x_{m-2} + x_{m+2})\right], \qquad (2)$$

where x_m is the content of channel m. No calculation was performed to correct for the presence of fission neutrons resulting from multiplication in the sample; this correction would not affect the total counts by more than a few percent. The primary uncertainties in the final spectra arise from the simplifying assumptions made to facilitate the unfolding of the pulse-height distributions. Since the subtractions are large for low neutron energies, the possible errors were also large at low energies. It is not known at this time whether the statistical uncertainties in the original data will permit the application of formal unfolding procedures to extract the delayed-neutron energy spectra and the associated energy-dependent standard deviations.

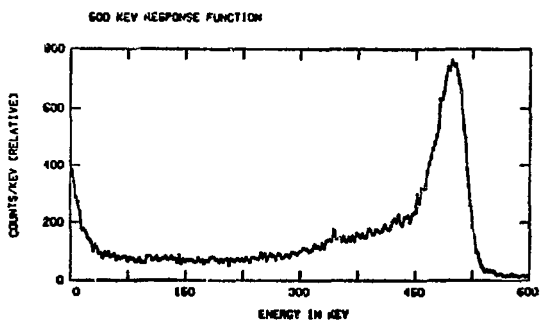

Fig. 13
Monoenergetic response function for 500-keV neutrons.

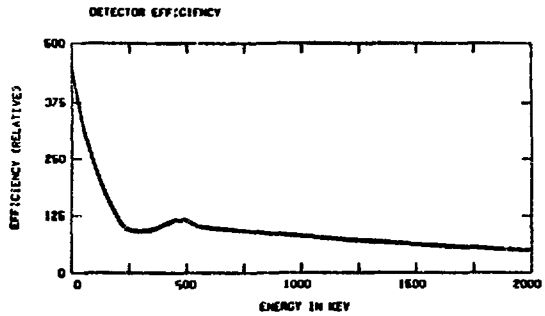

Fig. 14
Efficiency function for the shielded Shalev spectrometer.

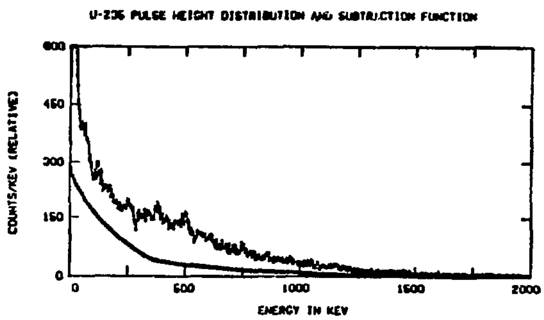

Fig. 15
Pulse-height distribution for delayed neutrons from ^{235}U (upper curve); subtraction distribution resulting from unfolding procedures.

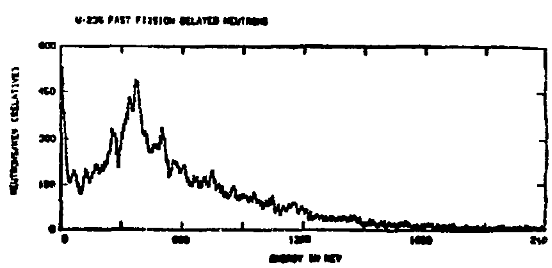

Fig. 16
Delayed-neutron response spectrum from the fission of ^{235}U.

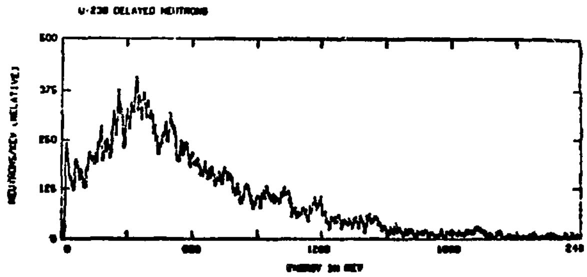

Fig. 17
Delayed-neutron spectrum from the fission of ^{238}U.

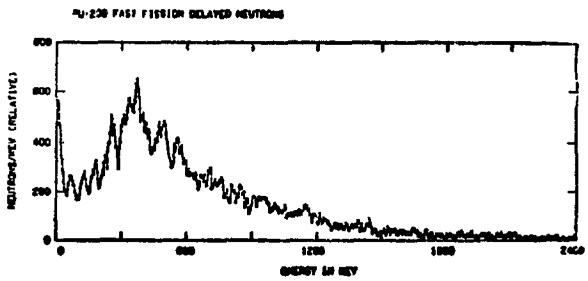

Fig. 18
Delayed-neutron spectrum from the fission of ^{239}Pu.

III. ERDA NONDESTRUCTIVE ASSAY TRAINING PROGRAM
(R. H. Augustson and T. R. Canada)

Twenty-seven people, representing government, government contractors, private industry, and the International Atomic Energy Agency (IAEA), attended the third U.S. ERDA nondestructive assay course on the use of portable instrumentation for safeguards. The course format was changed so that the individual laboratory groups were reduced to 2-3 persons per instrument. Although the class was large, this reorganization allowed each attendee greater opportunity to operate the instrumentation and to interact individually with the instructor. The following list of attendees emphasizes the wide representation at the course.

Winston Alston
IAEA, South Africa

Harvey C. Austin
Oak Ridge National Laboratory

Joan Beiriger
Lawrence Livermore Laboratory

Eugene E. Clark
Oak Ridge Gaseous Diffusion Plant

Steve Cocking
Los Alamos Scientific Laboratory

George T. Furner
Atlantic Richfield Hanford Company

Tom Gardiner
Los Alamos Scientific Laboratory

Herminio Gonzalez-Montes
IAEA, Spain

James Griggs
Goodyear Atomic Corp.

Neil Harms
IAEA, USA

A. T. Hirshfield
National Bureau of Standards

Ian Hutchinson
IAEA, United Kingdom

Donald R. Joy
Westinghouse Electric

Ahmed Keddar
IAEA, Algeria

Tony R. Lopez, Jr.
Sandia Labs

Gaston Martinez-Garcia
IAEA, Chile

Tsuyoshi Mishima
Power Reactor and Nuclear Fuel Development Corp.

Catherine S. Morimoto
U.S. ERDA, Albuquerque Operations Office

Leo Oudejans
IAEA, Netherlands

Donat Petrunin
IAEA, USSR

Vladimir Poroykov
IAEA, USSR

Harry T. Rook
National Bureau of Standards

Robert Schaer
IAEA, Switzerland

Don Sharpe
Babcock & Wilcox, NUMEC

Thomas Shaub
U.S. NRC

Willi Theis
IAEA, Germany

Stanley Turel
IAEA, USA

IV. IN-PLANT DYNAMIC MATERIALS CONTROL— DYMAC PROGRAM (R-Division, CMB-Division, and E-Division Staffs)

A description of DYMAC, the LASL R&D Program in real-time nuclear materials control, has been presented.[16] To achieve the DYMAC goals, the program is organized into three major areas of effort: (1) concepts and system development, (2) implementation, and (3) technology transfer. The concepts and system development effort provides an in-depth study of real-time accountability components. This effort includes the tasks listed in Table IV. In particular, the development of model simulation capability is being actively pursued. The immediate use for this valuable tool is the study of limits of error on net material flow through specific unit processes. The implementation effort has the long-range goal of installing a fully operational real-time accountability system at the LASL plutonium facility (TA-55) now under construction. Phase I of that effort is a test and evaluation program involving installation of in-line NDA instrumentation and interactive terminals at the present plutonium facility (DP-Site). In parallel, the preparation of specifications for the TA-55 DYMAC system (Phase II) has begun. Technology transfer is recognized as a vital and integral part of the overall program. The mechanisms for communication with government and industry are being formulated (Table V).

A. Concepts Development and System Analysis

Unit Process Study by Model Simulation (D. B. Smith). For nuclear material accountability systems oriented around the unit process, the material balance is computed for each integral portion of a facility process. Material accountability based on the unit process provides more incisive and more timely detection of process anomalies whether they be due to holdup in process equipment, measurement biases, or diversion of material, and can reduce the need for periodic shutdown and cleanout of the facility for physical inventory.

Effective implementation of this kind of material control requires measurement of all material entering the unit. All product and residue leaving the process must also be measured. This complete measurement of all input and output permits a material balance to be computed for the unit. Because of measurement uncertainties, the value obtained for the material balance usually will not be zero, and the error associated with this value must be determined in order to judge whether or not a nonzero material balance is indicated.

To investigate the sensitivity of material control around the unit process, a Monte Carlo model of the

TABLE IV

CONCEPTS AND SYSTEMS DEVELOPMENT TASKS

Data Collection	Data Base Management	Real-Time Accountability/Control Parameters
Criteria for unit process definition	Available data base management systems	Develop modeling capability study sensitivity of various control parameters
Automatic item identification options	Hardware/software tradeoffs	Develop calculational algorithms for limits of error
Operator/ terminal interaction philosophy	Hardware/software security	Evaluate filtering techniques and use of estimation formalism

TABLE V

TECHNOLOGY TRANSFER FUNCTIONS

I. Documentation

 A. Progress Reports
 B. Topical Reports
 C. Operational Manuals
 D. Technical Specifications and Drawings

II. Presentations/Briefings to Government, Industry, and Public

III. Technical Training Courses

IV. Close Interaction with Facility Planning Program

V. Input to Regulations/Guides

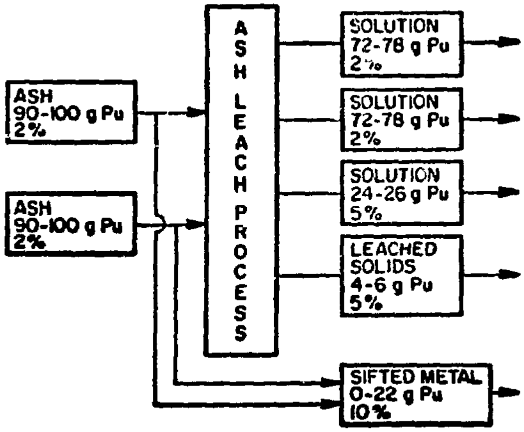

Fig. 19
Schematic diagram of ash-leach process Monte Carlo simulation. Percentage values represent relative precision (one standard deviation) of the associated measurement.

ash-leach process at DP-Site has been constructed. The model assumes that measurements of input, product, or residue are distributed normally about the true value with specified precision and accuracy. Provision is made for unmeasured holdup and diversion. A material balance is computed for each batch of material processed in the unit.

In typical operation of the ash-leach process, one or more containers of plutonium-bearing ash are sifted to remove metal pieces, then combined and dissolved in acid. The output from the process consists of several batches of solution and a small amount of leached solids. In the Monte Carlo model the amount of plutonium in each of the input and output channels is determined, and the measurement of these quantities is simulated using appropriate values for the imprecision of the various measurements. Figure 19 gives values used for the plutonium content and measurement uncertainty. Typical results for the processing of 100 batches of material under ideal conditions (independent and unbiased measurement of all input and output with no unmeasured holdup) are shown in Figs. 20 and 21 as cusum charts. The cusum chart is a graphical control scheme particularly suited to detecting changes in the operation of a process. On these charts the cumulative sum of batch-wise material balances is plotted as a function of batch. The vertical bar represents the uncertainty (one standard deviation) in the cumulative sum. Both figures show the same series of batches and measurements except Fig. 21 shows 1 g of plutonium removed from one of the output channels in each batch. For the quantities of material and measurements involved in this operation of the ash-leach process, 1 g is equal to approximately one-fourth of the uncertainty in each batch balance. Removal of this small amount of material would go undetected in a single batch balance; however, after only 20 g of plutonium have been diverted, the cusum chart has begun to look suspicious.

This model is one of the first building blocks in a system study that will link the various interacting unit processes in operating nuclear facilities. It is being used to investigate parameters of interest in the design and evaluation of safeguards systems.

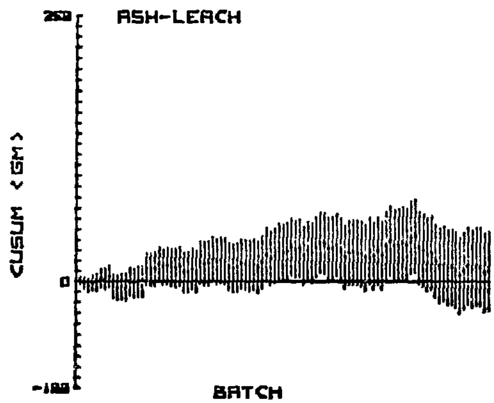

Fig. 20
Simulated results of processing 100 batches of material under ideal conditions (independent unbiased measurement of all input and output with no unmeasured holdup and no diversion).

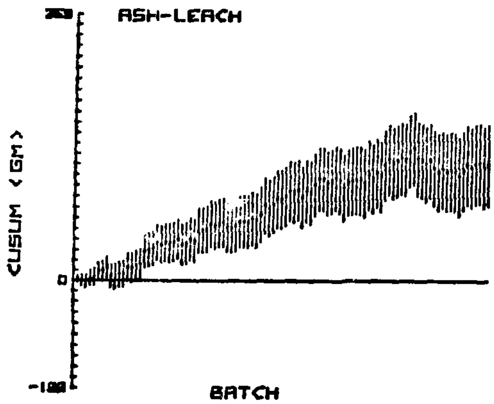

Fig. 21
Simulated results of processing the same 100 batches of material shown in Fig. 20 with 1 g of plutonium diverted from each batch.

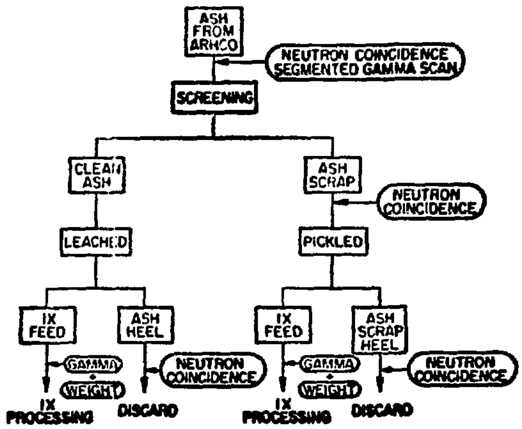

Fig. 22
Flow diagram of DYMAC Ash-Leach Process showing NDA measurement points.

Current investigations include the testing of various error estimation algorithms, the sensitivity of the system to measurement precision and accuracy and frequency of instrument calibration, and the effect of correlations between measurements using the same instrument.

B. DYMAC Implementation

1. DP-Site Test and Evaluation Phase

The present LASL plutonium processing facility (DP-Site) is being used to gain experience with various DYMAC components that will make up the real-time materials control system at the new site (TA-55). The test and evaluation program includes installation of in-line NDA instrumentation and a remote terminal prototype accounting system enclosing a portion of the recovery section, the ash-leach process.

a. Ash-Leach Unit Process (R. S. Marshall, N. Baron, and T. Gardiner)

The ash-leach process has been selected as a unit process around which DYMAC instrumentation will measure all plutonium inputs and outputs. Figure 22 schematically describes the ash-leach process. Material flow and types are identified by interconnected square boxes. DYMAC measurements are identified by round-cornered boxes with arrows showing process points where measurements will be made.

Presently, most of the ash processed by LASL is generated at the Atlantic Richfield Hanford Co. (ARHCO), Richland, Washington, facility. The ash is a heterogeneous mixture averaging about 5-10 weight percent plutonium. Ash is shipped in small, sealed cans. Referring to the top of Fig. 22, each can is assayed for ^{239}Pu with a Segmented Gamma Scanner (SGS) and for the effective ^{240}Pu content with a Thermal Neutron Coincidence Counter. These instruments are located in the counting room at DP-Site.

The screening step involves removal of fragments of glass, steel, lead, and graphite, which are identified as ash scrap. The ash scrap will be assayed with an In-Line Neutron Coincidence Counter. Recovery of the plutonium retained by the ash scrap is accomplished by pickling, that is, contacting the ash scrap with a hot HNO_3-CaF_2 solution to dissolve the surface dust containing plutonium.

After pickling, the ash scrap is filtered. The filtrate, identified as ion exchange (IX) feed, will be assayed with an In-line Plutonium Solution Gamma Counting System. This system is described in Section II.E of this report. The small amount of plutonium retained by the ash scrap heel will be assayed in the In-line Neutron Coincidence Counter.

The amount of plutonium left in the clean ash will be calculated by subtracting the relatively small amount of plutonium retained by the ash scrap (about 2-5% of the total ash plutonium) from the total plutonium contained in the ash cans. Plutonium in the clean ash is recovered by leaching: selectively dissolving the plutonium from the matrix with a HNO_3-CaF_2 solution. After leaching, the slurry is filtered. The filtrate is identified as IX feed and the residue as ash heel. As with the ash-scrap recovery process, the filtrate will be assayed with the

In-line Plutonium Solution Gamma Counting System and the ash heel with the In-line Neutron Coincidence Counter.

A plutonium material balance will be calculated by subtracting measured outputs from measured inputs. The ash scrap is accumulated for a few weeks before being processed. In contrast, clean ash batches are processed on a two-three day basis and account for about 95% of the plutonium entering the ash-leach process. Material balances will be maintained on the clean ash portion of the ash-leach process on a two-three day basis.

The DYMAC system applied to the ash-leach process at the LASL DP-Site is expected to meet the following goals: (1) test and evaluate plutonium assaying instruments, (2) test and evaluate interactive terminals plus computer hardware and software for data handling and processing, (3) calculate material balances on the clean ash-leach process on a regular two-three day basis, (4) train both production supervisors and technicians to operate measurement instrumentation and interactive terminals, (5) generate a data base from which the accountancy system can be evaluated and improved as necessary, (6) increase process efficiency by eliminating bag-out operations for samples and incompletely processed materials, (7) decrease the time required for the IX feed analysis from one to three days to one to three hours, and (8) reduce uncertainties in the process material balance to ±1% at 1 σ (±60-g plutonium on a 6-kg plutonium monthly throughput).

Most of the goals listed apply directly toward installing many of the DYMAC systems at the future LASL TA-55 site. During the next six to nine months, the Ash-Leach Process DYMAC System will be tested and modified as necessary. This testing period will allow sufficient lead time to design and procure systems that will require minimal changes when installed at TA-55.

b. In-Line Neutron Coincidence Counter (N. Baron and R. S. Marshall)

A thermal-neutron coincidence well counter similar to one previously reported[17] is under design and construction for installation at DP-Site. This instrument, which measures the neutron coincidences from spontaneous fission of plutonium, will provide a passive in-line assay of the mass of plutonium in cans of various materials from the ash-leach process. The neutron counter will be positioned in the ash-leach process line so that the cans will not leave a glovebox environment. The cans will be transported on conveyor belts from various gloveboxes to the measuring station. The cans will be positioned on a platform in a glovebox and automatically raised into the well of the counter that is positioned on top of the glovebox. The atmosphere of the counter's well is open to the glovebox but sealed to the outside environment.

The neutron counter's well diameter is 16 cm and a ring of 15 ^3He gas proportional counters to detect thermal neutrons is positioned in a polyethylene moderator, symmetrically about the well on a circular radius of 13 cm. The ash cans, when raised into the counting position in the well, will be surrounded by cadmium in order to decouple thermal neutrons in the moderator from the material in the can. This will reduce the probability of induced fission due to thermalized neutrons in the moderator backscattering into the sample. The moderator's outer radius is 18 cm, and it has an outer liner of cadmium. This cadmium liner, plus an outer shield of 10-cm thick polyethylene, insures low background count rates and improves the instrument's sensitivity. A mockup of this instrument was tested in the area where it will be installed and the instrument observed a background of about 1 count/s. The instrument should be capable of assaying material quantities as low as a few tenths of a gram. The data will be automatically processed by an on-line computer.

c. Computer Installation at DP-Site (J. H. Menzel and R. F. Ford)*

To implement the limited terminal-based accounting system, the Nova 840 computer has been installed at DP-Site. Four terminals have been delivered, but installation in the plant is awaiting checkout of the communications software. The present LASL accounting system, which is unit process based, is being used as the model for the real-time system. Significant effort has gone into understanding this accounting system to enable programming it on the Nova 840. This learning phase has involved close communication between CMB-11, the plutonium processing group, and ADASF, the LASL organization responsible for the entire laboratory's nuclear materials accountability. While still in the learning phase, a preliminary version of the program has been written using only one terminal for input. This version is being debugged in cooperation with CMB-11 and will soon be ready for expansion to multiple terminals. A number of diagnostics have been added to insure a high degree of accuracy in the information entered into the system. The kinds of output information to be assembled from the data

*LASL Group E-5.

base are still being formulated, but the minimum number of reports will be those available under the present accounting system.

2. DYMAC for the New LASL Plutonium Facility (TA-55) (T. Gardiner and R. H. Augustson)

The planning for the new LASL Plutonium Facility (TA-55) is being conducted in parallel with the test and evaluation efforts at the existing plutonium plant. One of the fundamental assumptions in the design of the safeguards system for TA-55 is that all of the basic hardware will be proven under actual plant conditions at the existing plutonium facility (DP-Site). Prototype NDA instruments, weighing devices, terminals, and a computer system are now undergoing evaluation in preparation for acceptance as part of the TA-55 project.

A scheduling plan is being developed utilizing PERT techniques. The major boundary target dates are known and the detailed schedule for instrument installation, building modifications, and other parts of the project has been tentatively established. The first PERT computer run has been made. This run showed the need for further refinements in the schedule. These will be made and the PERT program will be run again in the near future.

One of the most important tasks that must be performed on a project of this nature is the issuance of a formal specification. The past six months have been devoted to concept development, and it is now felt that there is adequate background to formalize the specification. An outline has been developed that divides the specification into two parts. An external or functional specification will state the goals and quantify the parameters that will define the overall effectiveness of the plant materials control system. An internal specification will be prepared that will detail the philosophy and the implementation for the facility. The implementation portion of the specification will identify equipment, equipment location, communication facilities, computer requirements, and all other elements that are presently believed to be essential to the facility.

It is anticipated that the specification and scheduling phase of the project will be completed in two to three months.

C. Technology Transfer (R. H. Augustson)

Several presentations have been given on various aspects of the DYMAC Program, including an invited paper[18] at the IAEA International Symposium on the Safeguarding of Nuclear Materials held in Vienna, Austria, on October 20-24, 1975. LASL personnel have visited the Rocky Flats and GE-Wilmington facilities, exchanging technical information on the implementation and basic concepts of real-time accountability. Further visits to other sites are being planned.

Documentation is being assembled on in-line NDA techniques as experience is gained during the DP-Site test and evaluation phase. This documentation will be published during calendar year 1976. Publications covering other aspects of real-time accounting are presently being planned.

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