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A PROGRESS REPORT

MASTER

Nuclear Analysis Research and Development

Program Status Report

January - April 1972

los alamos
scientific laboratory

of the University of California

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Nuclear Analysis Research and Development

Program Status Report

January - April 1972

Nuclear Analysis Research Group, A-1
G. Robert Keepin, Group Leader

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I. NEUTRON INTERROGATION AND NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS

A. Investigation of Sampling/Chemical Assay Using Non-destructive Assay Techniques—(J. H. Menzel, A. C. Dumrose (CMB-8), and R. H. Augustson)

The reported comparison studies^{1,2} on two categories of highly enriched (93% ²³⁵U) uranium scrap, namely "Rover dust" and organic ash, have resulted in the conclusion that nondestructive assay of whole containers should be used *in lieu* of sampling/chemical analysis for low-uranium-concentration scrap. Consequently, the neutron-interrogation/delayed-neutron-yield method has been used to measure the SS value of 250 cans of Rover dust and graphite pieces for accountability purposes (see Section I-C), and procedures are being established to assay all recoverable uranium scrap at LASL by this NDA method on a routine, current basis. Thus, the major objective of this investigation has been completed. What remains to be done is to study the mechanisms involved in blending and sampling procedures and to determine the limits of error of sampling/chemical analysis as well as nondestructive assay for scrap materials. With respect to the latter, recent results on the precision of sampling/chemical analysis obtained as part of the field program at Oak Ridge³ are particularly revealing.

Previous difficulties at Oak Ridge⁴ with analytical measurements of low-enriched incinerator ash (< 30%U, 1 < % ²³⁵U < 10) were traced to the use of auger rather than V-type blending. In conjunction with the evaluation of nondestructive techniques for assay of this material, the precision of analytical measurements on samples taken from sixteen cans after thorough blending was determined. The results of the analytic measurements are shown in Tables I and II for sampling/chemical analysis (g U) and sampling/mass-spectrometer analysis (% ²³⁵U), respectively. In each case, the average and the relative standard deviation of three measurements has been calculated. Based on these data, the average standard deviation of sampling/chemical analysis is 3.6% and that of sampling/mass-spectrometer analysis is 2.7%; further, the average standard deviation of the ²³⁵U value for each can is 5%, due to the propagation of errors of the uranium and % ²³⁵U analyses. The second and third columns of Table I also provide information on the precision of sampling/chemical analysis on duplicate samples from the same blend. The precision for this case is 2.2%, which is in

TABLE I
INCINERATOR ASH
PRECISION OF SAMPLING/CHEMICAL ANALYSIS

First Blend	Uranium Content (g)			Rel. Std. Dev. (%)
	Sample 1	Sample 2	Average	
597	569	565	577	3.0
1613	1586	1543	1581	2.2
1051	1062	1035	1049	1.3
651	674	689	671	2.9
1146	959	1011	1029	9.3
914	964	949	942	2.7
676	631	659	655	3.5
2448	2387	2358	2398	1.9
1296 ^a	518	491	505	3.8
1014	997	1007	1006	0.8
1423	1446	1460	1443	1.3
400	383	402	395	2.6
200	260	273	244	15.9
175	159	168	161	3.6
375	378	386	380	1.5
655	666	644	655	1.7
			Avg. Dev.:	3.6%

^aOmitted from calculations

agreement with the 2.5% result obtained at LASL on so-called organic ash.¹ Thus, it is clear that for this category of materials (e.g. incinerator ash) sampling errors—even after thorough blending—limit the accuracy of sampling/chemical analysis and sampling/mass-spectrometer analysis.

B. Assay of Small Samples by Subthreshold Neutron Interrogation—(A. E. Evans, M. M. Thorpe, and J. J. Malanify)

The small-sample fissile-assay system described in the previous progress report⁵ has been tested in two

TABLE II
INCINERATOR ASH
PRECISION OF SAMPLING/MASS SPEC. ANALYSIS

First Blend	²³⁵ U Enrichment (%)			Rel. Std. Dev. (%)
	Sample 1	Sample 2	Average	
3.67	3.23	3.25	3.38	7.3
4.02	4.17	4.04	4.08	2.0
4.38	4.22	4.08	4.23	3.6
2.51	2.66	2.43	2.54	4.6
4.05	4.15	4.15	4.12	1.4
2.40	2.33	2.41	2.38	1.9
2.59	2.64	2.75	2.66	3.2
2.92	2.74	2.81	2.82	3.1
8.41 ^a	4.14	4.08	4.11	1.1
5.68	5.88	5.71	5.76	1.9
5.10	5.30	5.35	5.25	2.5
5.51	5.82	5.98	5.77	4.1
6.86	6.71	6.77	6.78	1.1
8.12	8.00	7.87	8.00	1.6
9.03	9.51	9.08	9.21	2.9
5.06	5.03	4.98	5.02	0.8
			Avg. Dev:	2.7%

^aOmitted from calculations.

production-type inventory campaigns. This system is now operational and ready for carrying out routine assay runs.

A batch of 220 samples of uranium-thorium-carbide fuel material withdrawn from an AEC licensee plant during a recent DNMS inspection was sent to LASL for nondestructive assay before chemical analysis at the New Brunswick Laboratory. The samples had been withdrawn into previously tare-weighted vials. The gross weight of the samples was determined at Los Alamos. These samples contained an average of $\sim 1/8$ to $1/4$ g of fissile material per sample, and required 1000 sec of irradiation-counting time per sample to obtain counting statistics of 2% at the 1σ level for the lower-content samples. Furthermore, the fissile concentration of material in the samples was outside the range of concentrations studied during initial tests of the system, so that additional standards had to be prepared to test the validity of the assay results obtained. The tests indicated that, for powdered uranium-thorium-graphite samples in glass vials identical to those used in all previous work (Kimble No. 60910-L 15 by 45 mm,

1-dram with screw-on Bakelite cap), volume insensitivity is obtained for all vial fillings up to full volume. However, mockups of completed fuel pins, about the same size as the vials, were found to assay low by a factor of 1.042, and this factor was applied to the results for these items.

Assay of the complete batch of DNMS samples took 129 h of accelerator operating time. Two men operated the system: one to operate the accelerator and nuclear counting system, and another simply to change samples. The addition of automatic sample-changing equipment clearly could reduce the manpower requirements appreciably. The estimated cost of assaying these samples was approximately \$30.00 each, which compares favorably with an estimate of \$200 per sample for chemical assay of this particular type of material;⁶ this cost can be further reduced by the installation of automatic sample-changing equipment.

Results of chemical assay are being awaited to permit an evaluation of overall precision and bias of the system. One immediate conclusion of this exercise, however, is that a larger standard vial should be used for inventory samples with average fissile material concentrations as low as encountered in the above batch. A minimum of 1 g of fissile material per sample would enable 0.5% counting statistics to be obtained in 10 min counting time (while also permitting the added confidence of assaying each sample at least twice).

A second test of the assay system was made using 50 samples (~ 1 to 2 g each) of UAl_x (93% ²³⁵U) powder supplied by the Aerojet Nuclear Corporation. Chemical analysis by Aerojet verified that all samples contained 66.26 wt% ²³⁵U with a standard deviation of $\pm 0.032\%$. The mean fissile content of the samples, with reference to a LASL (Group CMB-1)-prepared standard, was determined to be 66.13 wt% ²³⁵U, an apparent relative bias of -0.19% . For 0.5% standard deviation on the measurement of each of fifty samples, the standard deviation of the mean is $\sim 0.5\%/\sqrt{50}$ or $\sim 0.07\%$; on this basis, then, a measured bias of 0.19% is not unreasonable. The distribution of assay values about the mean, shown in Fig. 1, together with a superimposed normal curve, is evidence of the reliability of the system.

The quantity of fissile material in these samples (roughly a gram) permitted the entire batch to be assayed in two working days, with 19 accelerator operating hours, for a total assay cost of approximately \$15.00 per sample. This indicates the improvement in assay efficiency that can be achieved when adequate material is provided.

Operational efficiency of the small-sample handling system has been aided by a data-handling procedure which completely eliminates hand transcription of data. Data from scalers, including manually entered sample numbers and sample weights, are read out onto a punched paper tape, which is then transferred to the A-1 PDP

Fig. 1.

Assay results on 50 samples of UAl_x powder by neutron interrogation and delayed-neutron counting. The dotted curve is a predicted normal distribution based upon counting statistics.

computer system for reduction. Computer output includes a compilation of background runs and standard calibrations, results of each assay run, ratio checks of paired counters, ordering of the results by sample number and averaging assays of the same sample, and, finally, print out of an assay-report sheet.

Capability for assay of fertile isotopes was achieved with the recent upgrading of the A-1 Van de Graaff to a maximum accelerating potential of 3.75 MV. This enables the production of neutrons of energies up to 2.07 MeV from the $^7\text{Li}(p,n)^7\text{Be}$ reaction and up to 2.97 MeV from the $^3\text{H}(p,n)^3\text{He}$ reaction. The capability for nondestructive assay of such breeder-reactor feed materials as ^{232}Th and ^{238}U is destined to be increasingly important in the future as fast-breeder fuels enter the power-production scene.

C. Measurement of the LASL Enriched-Uranium Inventory--(J. H. Menzel and R. H. Augustson)

Interfacing of the neutron interrogation data-acquisition system with Group A-1's PDP-9/L computer has been completed. Programs have been written to provide remote real-time output of various experimental parameters and results at the assay station. Computer presentation of the final results of a set of inventory measurements are incorporated directly into the LASL SS-material inventory system.

Using these automated procedures, the SS-material inventories of two highly enriched uranium-scrap categories were updated by neutron interrogation measurements. A total of 263 recovery cans (8 in. diam by 10 in. high) containing 129 kg uranium (93% ^{235}U) in 1,658 kg of bulk weight were transferred and measured in less than one work week.

D. Matrix-Material Effects on Fission-Neutron Counting Using Thermal- and Fast-Neutron Detectors--(H. O. Menlove)

For active assay techniques using neutron interrogation, it is often desirable to count the induced prompt neutrons using fast-neutron detectors such as ^4He gas tubes or ZnS counters. In addition, the less-abundant delayed neutrons can be counted using high-efficiency thermal-neutron detectors such as ^3He gas tubes. A series of measurements have been started to determine the relative effect of various matrix materials on these fast-neutron and thermal-neutron detectors.

For the measurements, 1-gal recovery cans were filled with the representative scrap materials listed in Table III. A constant source of fission neutrons (13 μg of ^{252}Cf) was then placed in the center of each can and the changes in the three detector responses were recorded. Since the matrix materials decreased the outgoing neutron energies, the response in the thermal-neutron detector (^3He) was generally enhanced by the added material; however, since the ^4He and ZnS detectors only count neutrons with energies above a low-energy bias (e.g. 400 keV), their response is decreased by the matrix material. Also, the fast-neutron detectors are much more sensitive than the thermal detector to the type and amount of matrix material as can be seen in Table III.

It should be noted that in actual neutron-assay measurements, the matrix material affects both the interrogation-neutron flux and the induced-fission neutrons. These combined matrix effects will be investigated in future work.

TABLE III
MATRIX MATERIAL EFFECTS ON FISSION-NEUTRON DETECTION
IN 1 GAL SCRAP RECOVERY CANS

Matrix Material	Material Mass (kg)	Normalized Detection Rate		
		³ He Detector (thermal neutrons)	⁴ He Detector (fast neutrons)	ZnS Detector (fast neutrons)
Empty 1-Gal Can		1.00	1.00	1.00
Metal Parts	3.60	1.04	0.83	0.75
Slag-Crucible	1.80	1.03	0.94	0.91
Concrete	3.24	1.05	0.84	0.79
String Filters	0.60	1.07	0.95	0.86
CH ₂ (ρ=0.65 g/cc)	0.27	1.06	0.96	0.92
CH ₂ (ρ=0.12 g/cc)	0.43	1.09	0.92	0.90
CH ₂ (ρ=0.27 g/cc)	0.97	1.19	0.71	0.67
H ₂ O (ρ=1.00 g/cc)	3.62	0.98	0.36	0.35

E. Technical Support of the Active-Assay Program at a Uranium-Scrap Recovery Facility--(R. H. Augustson and D. B. Smith)

LASL is providing technical support for the active-assay program at the United Nuclear Corporation Recovery Facility, Wood River Junction, Rhode Island. The measuring system utilizes a repetitively pulsed, 14-MeV sealed-tube neutron generator, fission-chamber monitoring, and a delayed-neutron detector similar in design to the LASL slab detector. This system is designed to measure the uranium content of incoming shipping containers without opening the outer container. Most of the incoming shipments consist of 55-gal barrels with a 5-in. diam central pipe in which the fissionable material is contained. The annular volume between the pipe and the outer barrel is usually filled with vermiculite. Initial measurements showed that some barrels contained wet vermiculite due to moisture leakage into the barrel. Since this moisture will strongly influence the neutron response, a ²³⁵U fission chamber is used to provide sensitive detection and monitoring of moisture content in these containers. United Nuclear Corporation is investigating the range of H₂O content over which useful assay results can be obtained. To support this effort, LASL has made a series of one-dimensional DTF-IV calculations providing guidance for the experimental program. Representative results are shown in Table IV for a range of H₂O and ²³⁵U densities. The fission-chamber response per source neutron results indicate the sensitivity of the fission chamber for detecting wet barrels. There is a factor of five increase in going from no water to 0.1 g/cc (~5 gal of H₂O per 55-gal barrel). An additional important result is that the fission-

TABLE IV
DTF-IV CALCULATIONS OF RESPONSE FOR
UNC SHIPPING CONTAINERS AS A FUNCTION
OF URANIUM AND H₂O CONTENT

ρ(²³⁵ U) (g/cc)	ρ(H ₂ O) (g/cc)	Normalized ²³⁵ U Fission Chamber Response/Source Neutron
0.009 (~100g)	0.0	1.0 ^a
	0.1	5.7
	0.2	10.8
0.1 (1000g)	0.0	1.0
	0.1	5.2
	0.2	10.9
0.2 (2000g)	0.0	1.0
	0.1	4.9
	0.2	10.9

^aNormalization point.

^aNormalization point.

chamber response depends only on the H₂O and not the uranium content.

The spatial effects of material distribution are difficult to calculate, and will be studied largely through experimental measurements.

II. MATERIALS ANALYSIS USING ISOTOPIC SOURCES

A. Thermal ^{252}Cf Fuel-Rod Assay System with Pellet-to-Pellet Scan--(H. O. Menlove, R. A. Forster, J. L. Parker)

The design and fabrication of the thermal-neutron ^{252}Cf fuel-rod assay system,⁷ with modifications for pellet-to-pellet scan, has been completed. A diagram of the neutron-interrogation system and NaI detector for counting delayed gamma rays is shown in Fig. 2. As with the previous system,⁸ the total fissile content of the fuel rods can be measured using ^4He neutron detectors located in the graphite core of the moderator assembly. In addition, the pellet-to-pellet fissile content is measured using the small (2 by 2 by 3/4 in.) NaI detectors positioned near the fuel-rod exit channels. High-energy delayed gamma rays also can be used to determine the total fissile content if the gamma-ray background of the fuel rods is measured before irradiation. The ^{252}Cf moderator core was offset 3 in. axially from the center of the barrel

assembly to increase the delayed gamma-ray signal in the NaI detector and to improve the signal/background ratio in the ^4He detectors.

The central core of the moderator contains 3-cm-thick ZrH_2 surrounded by 2-cm-thick lead to give maximum neutron moderation and gamma-ray attenuation with minimum neutron attenuation. The $\text{D}_2\text{O} \cdot \text{WEP}$ (70% D_2O in extended polyester resin) provides the desired D_2O content with convenient handling and fabrication properties. This new material possesses the desirable moderation properties given in Table V which lists other common moderating materials for comparison.

Calculations have indicated that replacing the $\text{D}_2\text{O} \cdot \text{WEP}$ by carbon in the assembly shown in Fig. 2 reduces the induced signal by $\sim 12\%$, the $^{235}\text{U}/^{238}\text{U}$ fission ratio by 50%, the signal/background ratio in the ^4He detectors by 35%, and increased the external neutron dose by $\sim 30\%$.

Preliminary measurements using a prototype of the system shown in Fig. 2 indicate that a $500\text{ }\mu\text{g } ^{252}\text{Cf}$ source would yield a delayed-gamma-ray counting rate of $\sim 80,000$ cps for a 3% enriched PWR-type fuel rod. This would correspond to an assay accuracy of $\sim \pm 3\%$ (1 σ) on a pellet-to-pellet measurement for a rod feed rate of ~ 7 ft/min.

It is expected to have the prototype system shown in Fig. 2, with up to four assay channels, assembled and running in the near future to obtain more accurate data on its performance. This information is being made available to qualified instrument manufacturers on request.

Fig. 2.

Thermal neutron ^{252}Cf fuel-rod assay system with modifications for pellet-to-pellet scan. The ^4He neutron detectors in the carbon core count the prompt fission neutrons for total fissile determination, and the NaI detectors near the fuel-rod exit channel count the delayed gamma rays for pellet-to-pellet determination.

TABLE V

MODERATING PROPERTIES

Material	Slowing-Down Power ($\xi\Sigma_s$)	Moderating Ratio ($\xi\Sigma_s/\Sigma_a$)
C	0.063	170
CH_2	1.63	63
D_2O	0.18	2100
$\text{D}_2\text{O} \cdot \text{WEP}^a$	0.36	97

^aThe WEP resin contains $\sim 70\%$ D_2O by weight.

B. Fast Neutron ^{252}Cf Assay System for FFTF Fuel Rods—(H. O. Menlove, J. L. Parker, R. A. Forster, and Darryl B. Smith)

The fabrication of the ^{252}Cf assay system for FFTF fuel rods described in the previous progress report⁹ has been completed, and tests on the system components are in progress. Figure 3 shows the complete system including the neutron-irradiation assembly, two 5 by 5 in. NaI detectors, electronics rack, automation and data-reduction rack, and automated fuel-rod-handling system.

The two NaI through-hole detectors are used to count the delayed gamma rays induced by the $500\text{ }\mu\text{g}$ ^{252}Cf fast-neutron irradiation as well as the passive gamma rays from the plutonium for the pellet-to-pellet determination. The NaI detectors are shielded from the ^{252}Cf tank by ~ 6 in. of lead and tungsten to reduce the background in the detectors. Two NaI detectors, each with independent electronics, readout, etc., are used in the system to increase the statistical precision and to make the system operational even with the failure of one of the detector systems.

The fuel-rod transfer and data reduction is automated using a HP-9100B programmable calculator and extended memory unit (HP-9101A) together with a BEC 9507 scaler interface unit. After data reduction and fitting to a calibration curve, the results are printed out using the IBM typewriter shown in Fig. 3.

The assay system is undergoing tests at LASL to determine optimal operating parameters and statistical precision. In June 1972 the system will be set up at the Westinghouse HEDL facility in Richland, Washington, for in-plant evaluation and use.

C. Photoneutron Assay System Using ^{124}Sb or ^{88}Y —(H. O. Menlove and R. A. Forster)

The fabrication of the neutron reflector and gamma-ray shield for the Sb-Be assay system¹⁰ has been completed. Figure 4 shows the unit with the central core composed of beryllium, titanium, and nickel surrounded by ~ 3 in. of lead to shield the ^4He detectors from the ^{124}Sb gamma rays. Prompt neutrons resulting from fission

Fig. 3.

Fast neutron ^{252}Cf assay system for FFTF fuel rods. System includes $500\text{ }\mu\text{g}$ ^{252}Cf source and shield, two 5 by 5-in. NaI detectors to count the delayed gamma rays, automated fuel rod handling, and data reduction system.

Fig. 4.

Photoneutron assay system using either ^{124}Sb or ^{88}Y in the beryllium core surrounded by nickel and titanium neutron reflectors and lead gamma-ray shielding.

reactions in the central region are detected using the ^4He detectors which are biased above the energy of the interrogation neutrons. The ring of ^4He tubes is surrounded by 2 cm of nickel to reflect the fast neutrons back into the detector ring. The increase in the signal from the nickel reflector was calculated to be $\sim 40\%$. The same amount of fast-neutron reflection could be obtained with ~ 4 cm of iron with a corresponding increase in size and weight of the system.

Fast-neutron detectors using ^4He gas are being optimized for use in the system shown in Fig. 4. Detector gas pressures ranging from 9 to 20 atm are being tested for neutron efficiency and gamma sensitivity.

D. Random Source Interrogation System--(J. E. Foley)

A completely new random source-interrogation system (or "random driver") (Fig. 5) is undergoing final assembly and testing before its demonstration to begin in May 1972 at the Oak Ridge Y-12 plant. The new system is more versatile than the earlier system¹¹ and will accept samples up to 12 in. in diameter and 20-in. high. The two plastic scintillation detectors are mounted on ball bushings so they can be easily moved to accommodate various size samples and still retain as large a solid angle as possible. The front of the counter is fitted with doors for

easy access to the interior of the counter. The sensitivity of the system is $\sim 5 \text{ g } ^{235}\text{U}$ for 5-in. sample diameter and considerably less for larger volumes; i.e. sensitivity for a 12-in. diam container is $\sim 25 \text{ g } ^{235}\text{U}$. This reduction in sensitivity for large samples can be partially compensated for by an increase in the random neutron source strength.

The AmLi random source is mounted on a lead screw to produce linear vertical motion with the vertical travel of the source variable from zero to 20 in. The photomultiplier tubes have been mounted in the base of the instrument to protect them from accidental damage. The total height of the system is 42 in.

E. Calibration of ^{252}Cf Fuel-Rod Assay System--(D. B. Smith and R. A. Forster)

Since the last reporting period, G. E. (Wilmington) has fabricated four new BWR fuel-rod standards at each of the four nominal enrichments represented by the previous set of standards (0.71, 1.70, 2.10, and 2.79% ^{235}U enrichments). All sixteen of these new standards contain undished pellets. Because the current mode of operation of the assay system involves the assay of groups of production rods all having the same nominal enrichment and similar pellets (either dishd or undished), it was decided to completely recalibrate the system.

The response of each of the sixteen new standard rods, plus the two previous standard rods containing undished pellets, was measured repeatedly in each assay channel to determine precisely the response of each rod in each channel. Since a general non-linear regression program for reduction of the calibration data was not readily available at G. E., the data were analyzed at LASL using PACKAGE¹² to provide the calibration constants.

The regression procedure used previously¹³ with the programmable calculator was amended to derive additional information from the data; i.e., the channel normalization factors (formerly obtained by hand normalization of the data) were determined by the fitting procedure. Thus, the system of equations

$$R_{ij} = \frac{A}{k_i} (1 - e^{-B U_j}) \quad , \quad i = 1, 4; \quad j = 1, J$$

where R_{ij} is the response of the j^{th} standard in channel i , and U_j is the ^{235}U content of the j^{th} standard; the total number of standards, J , is 18 in the case at hand. A and B are the calibration constants, and k_i , the normalization factor for channel i , was fitted to the unnormalized data in order to obtain simultaneously A , B , k_2 , k_3 and k_4 ($k_1 \equiv 1.0$). The resulting calibration parameters are given in Table VI along with those from the previous calibration.

Fig. 5.

The random source-interrogation system used to determine the ^{235}U content in containers of up to 5-gal capacity.

TABLE VI
COMPARISON OF CALIBRATION PARAMETERS

	Current (2/72)	Previous (8/71)
k_1	1.00000	1.00257
k_2	1.10306	1.10242
k_3	1.15538	1.13449
k_4	1.09952	1.06951
A	1983.70	2195.50
B	0.00906769	0.00938350

Table VI shows that the channel normalization factors have not changed significantly relative to each other, indicating no gross relative change (channel-to-channel) in the electronics during the approximately six months the system has been in use. It should be emphasized that this change in the normalization factors is not reflected as a change in the assayed value for the ^{235}U content of any particular rod since the system is effectively "recalibrated" for each assay using the measured current background.¹³ The parameter A has decreased as expected due to decay of the ^{252}Cf source. The initial slope of the calibration curve is given by the product $A \times B$ and is not expected to change significantly from one calibration to

the next when corrected for source decay. The product $A \times B$ from the previous calibration is 20.60, and when corrected for source decay (187 days) the value 18.00 is predicted for the initial slope of the new calibration curve. The value of the initial slope from the current calibration is 17.99, which is in excellent agreement with the expected value.

After calibration, the new standards were assayed repeatedly by G. E. using the new calibration curve to check its precision and accuracy. Since the mean assayed mass at each enrichment was within 0.2% or better (2σ) of the "known" mass, no statistically significant bias in the calibration curve was evident.

The new standards were also assayed six times in each channel using the old calibration curve to check the reliability of this curve after more than six months of use since calibration. It is important to note that no statistically significant difference was observed between the assayed and "known" ^{235}U content of the standards at any enrichment.

Although it was necessary, in this case, to reduce the calibration data at LASL, a relatively simple FORTRAN-IV computer code has been written and sent to G. E. to enable G. E. to perform the weighted, non-linear least-squares regression necessary to obtain the calibration curve. This, coupled with the fact that G. E. personnel were closely involved with the entire calibration procedure, should give G. E. complete calibration capability in the future.

The question has been raised concerning the possibility of using, as a calibration standard, a fuel rod that contains pellets of a different diameter than the pellets in the other standards. The effect of variation in rod diameter was investigated using neutron-transport codes. The number of fission neutrons born per thermal source neutron was calculated for varying diameters of an infinitely long fuel rod inside a fixed-size channel in CH_2 moderator. The UO_2 fuel density was assumed to be 10 g/cm^3 , and the ^{235}U content was computed by assuming the ratio ^{235}U mass/ ^{235}U enrichment = 40, for a fuel diameter of 1.237 cm.

The results of the calculations are shown in Fig. 6 for fuel diameters of 0.9, 1.1, and 1.3 cm, which span the range of LWR fuel rods. For ^{235}U masses below $\sim 80 \text{ g}$, the differences are rather small. At small masses ($< 40 \text{ g}$ of ^{235}U), the smallest diameter rod has the largest response per unit mass because of its higher relative enrichment, hence less ^{238}U absorption. As the ^{235}U mass increases, this effect becomes relatively small and the geometry effects of the rod dominate; i.e., the larger the surface area of the rod, the greater the number of neutrons interrogating the rod per unit mass of ^{235}U . Thus the response at higher mass is proportional to the diameter of

Fig. 6.

Calculated fission-neutron response of typical LWR fuel rods from a thermal-neutron source as a function of ^{235}U mass content of the fuel rod for various fuel pellet diameters.

the fuel pellets. For example, a 3% reduction in the nominal diameter of BWR fuel pellets would produce an error of $\sim 0.5\%$ in the assay. Thus, it is clear that, in general, a standard rod containing pellets of a different nominal diameter than those of the other standards should not be used for calibration, particularly at ^{235}U masses greater than $\sim 80 \text{ g}$, because errors of the order of one to several percent would be introduced into the calibration curve.

F. The Response Function of the ^{252}Cf Fuel-Rod Assay System--(R. A. Forster)

The mathematical expression used to convert the response R of the thermal ^{252}Cf fuel-rod assay system to the ^{235}U content (in grams) of an LWR fuel rod is¹⁴

$$R = A (1 - e^{-BU}) \quad (1)$$

This function provides a simple semi-empirical means for accounting for thermal-neutron self-shielding by the fuel rod. The theoretical basis for this function has been investigated to determine its validity for interpolation between, and extrapolation from, calibration points obtained with standard rods and to search for an improved closed-form function.

A single channel of the assay system was modeled as a two-region, infinitely long cylinder. The inner region, defined by radius r_f , contains fuel and is surrounded by the second region, an infinite moderator. This model can be used to estimate the neutron absorption in the rod assuming the following:¹⁵

- one-group diffusion theory is valid in the moderator,

- the angular flux distribution incident on the rod is isotropic, and

- the first-flight transport model is applicable in the absorber.

The last assumption is modified using the work of Stuart¹⁶ to include the effects of scattering in the rod.

The resulting solution using the above model for neutron absorption in the ^{235}U in the rod leads to the following expression for R:

$$R = AF\beta (1 - 1/[1 + S(2 - \beta)/\beta]) , \quad (2)$$

where

$$F = \Sigma_{a25} / \Sigma_{af} ,$$

$$\beta = \frac{\Sigma_{af}}{\Sigma_{Tf}} \beta_{PA} / \left(1 - \frac{\Sigma_{sf}}{\Sigma_{Tf}} [1 - \beta_{PA} / 2 \Sigma_{Tf} r_f] \right) ,$$

$$\beta_{PA} = 1 - \frac{2}{p^2} [1 - (p + 1) e^{-p}] ,$$

$$p = 3 \Sigma_{af} r_f ,$$

$$S = 2 D_M k K_1 (kr_f) / K_0 (kr_f) ,$$

and

$$k = \sqrt{\Sigma_{aM} / D_M} .$$

D_M is the diffusion coefficient of the moderator, β_{PA} is the fraction of neutrons absorbed by a pure absorber, F is the fraction of neutrons in the fuel absorbed by the ^{235}U , and A is the proportionality constant incorporating source strength, rod surface area, multiplication effects, and absolute detection efficiency. In Eq. (2), β is the probability that a neutron incident on the fuel rod will be absorbed, while the term in parentheses is the flux depression in the moderator caused by the fuel.

Equation (2) may be simplified, using some reasonable assumptions, to

$$R = AF\beta_{PA} . \quad (3)$$

By expanding β_{PA} into a series and collecting terms, R can be further simplified to

$$R \approx AF(1 - e^{-BU} e^{-\Sigma_{a28} N_{28}}) \equiv \frac{AU}{E + U} (1 - Ce^{-BU}) , \quad (4)$$

where

$$BU + \Sigma_{a28} N_{28} \approx 2p/3 .$$

Fitting Eq. (4) to latest G. E. standards data using the general least-squares code PACKAGE¹⁷ showed that $E \approx 1$ and $C \approx 0.994$. Thus, Eq. (4) can be reduced to the original function, Eq. (1).

Goodness-of-fit checks of Eqs. (4) and (1) showed that including either C or E in the model significantly improves the fit over Eq. (1), while including the second parameter gains little. Therefore, the following calibration curve is suggested as an alternate for Eq. (1):

$$R = A(1 - Ce^{-BU}) . \quad (5)$$

The fact that $R \neq 0$ when $U = 0$ is not disturbing since a rod of pure ^{238}U would yield a small response from the high-energy source neutrons (> 1 MeV) reaching the rod.

To estimate the ability of Eqs. (1) and (5) to interpolate between, and extrapolate from, the standard points, several weighted fits to the latest G. E. standards data were performed, omitting one or more of the enrichments. The resulting curves for each equation were compared to the same curve fit to all of the data. Table VII shows the results for Eq. (1). The nominal enrichments of 0.71, 1.70, 2.10, and 2.79% are denoted by the numbers 1, 2, 3, and 4, respectively. The four responses represent measured responses of the four sets of standards in channel one; thus, the reference curve obtained from a fit to all four sets (1-2-3-4) is quite accurate in predicting masses of ^{235}U at these responses. When either of the two middle enrichments (sets 2 and 3) is deleted, Eq. (1) interpolates to within 0.2% of the reference curve. In comparison, Eq. (5) interpolates to within 0.04% of its reference curve.

The errors caused by extrapolation are expected to be larger than those by interpolation, as verified by Table VII. If the trend for extrapolation exhibited in Table VII is typical, then Eq. (1) can be expected to overestimate a mass falling outside the range of the standard masses; e.g., extrapolating from 83.1 to 109.6 g of ^{235}U leads to an assay error of 0.71%; the error is 1.30% for extrapolation from 66.7 g. For comparison, the extrapolation errors for Eq. (5) were 0.14% and -0.30%, respectively--a significant improvement.

In conclusion, both Eqs. (1) and (5) fit the standard mass points satisfactorily and provide accurate means for

TABLE VII
ERRORS IN INTERPOLATION AND EXTRAPOLATION BY
 $R = A(1 - e^{-BU})$

Response (cps)	Standard Enrichment Sets Used in Fit								
	²³⁵ U Mass (g)	% Error in ²³⁵ U Mass Compared to 1-2-3-4 Fit ^a							
		1-2-3-4	1-2-4	1-3-4	1-4	1-2-3	1-2	2-3-4	3-4 2-3
450	28.408		-0.12	-0.18	-0.49	-0.37	-0.50	0.62	0.78 0.36
900	66.696		-0.11	-0.12	-0.37	0.07	0.22	0.32	0.41 0.28
1050	83.093		-0.11	-0.08	-0.31	0.29	0.60	0.17	0.22 0.24
1250	109.579		-0.11	-0.01	-0.19	0.71	1.30	-0.11	-0.12 0.17
Interpolation					Extrapolation				

^a % Error = $100(\text{Mass}_{x-x-x} - \text{Mass}_{1-2-3-4}) / \text{Mass}_{1-2-3-4}$

interpolation and extrapolation. Although the two-parameter function, Eq. (1), accounts for thermal-neutron absorption remarkably well, when three or more

enrichment standards are available, Eq. (5) can be used to provide a still more accurate response function.

III. GAMMA-RAY AND X-RAY ANALYSIS APPLICATIONS

A. Passive Gamma-Ray Assays--(T. D. Reilly and J. L. Parker)

A considerable amount of passive gamma-assay work has been performed during this reporting period, these being largely "service assays" as part of the LASL showcase effort (i.e., to demonstrate model safeguards and accountability for AEC contractors and licensees). A listing of major items assayed follows:

- 1) Ninety-five drums (30-gal) of ²³⁸Pu wastes and scrap.
- 2) Twenty-seven cans of plutonium-bearing residues from electro-refining procedures.
- 3) Some 36 packages of low-level residues from plutonium recovery processes.
- 4) Twenty-two mixed carbide fuel pins as a quality assurance check on ²³⁵U and ²³⁹Pu content before irradiation.

- 5) Thirty small inventory samples of (U,Th)C from DNMS inspections. This effort was undertaken to complement the study of small-sample assay methods using active neutron interrogation and delayed neutron response.

B. Training of CMB-11 Personnel in Passive Gamma-Ray Assay Methods--(J. L. Parker and T. D. Reilly)

As part of LASL's "showcase" program of demonstrating exemplary safeguards and materials accountability, the CMB-11 plutonium facility at DP Site has acquired a Ge(Li) detector, multichannel analyzer, and a general-purpose scanning table. A-1 staff members have conducted a training course for appropriate CMB-11 personnel in the use of this equipment for assay of ²³⁸Pu and ²³⁹Pu scrap and waste at the DP Site plutonium facility. Having completed about 20 hours of training sessions, CMB-11 personnel are now using gamma assay instrumentation (in conjunction with previously installed neutron-coincidence assay systems¹⁸) for routine assays of

currently generated plutonium scrap and wastes. Consultation and assistance, when and as needed, is provided by A-1 on a continuing basis.

C. Demonstration of the SAFE Effluent-Water Monitor at K-25--(C. J. Umbarger)

As follow-up to an earlier demonstration of a waste-water activity monitor at Oak Ridge, K-25, a sewer-water system mock-up, including holding tanks, valves, pumps, control console, etc., and incorporating a low-energy radiation monitor based on the FIDLER¹⁹ concept, was demonstrated for three different groups at K-25. The demonstrator, known as SAFE (Safeguards Analysis For Effluents), is shown in Fig. 7. The radiation monitor, a thin (0.063 in.) NaI (Tl) scintillator with a 0.010-in. beryllium window, is located directly under the sample tank within the graded shield shown in Fig. 7. The system operates on a continuous basis and records radiation yields as a function of time, printing out the yields at the end of each preset count period. If the radiation levels should rise above a preset value, the system automatically opens a valve which diverts the contaminated water into a "Dump Tank" representing a holding pond (see Fig. 7), sounds an audio alarm, and actuates flashing red warning lights, while continuing to recycle automatically on its counting and printing cycle giving a running record of the contamination levels in the water being diverted. When the radiation level has returned to an acceptable range the system can be reset (by two push buttons on the front console).

SAFE demonstrations at Oak Ridge were performed for 1) operations personnel of the K-25, Paducah, and Portsmouth uranium enrichment facilities, 2) the analytical staff of K-25 and representatives from ORNL, Y-12,

and AEC Oak Ridge Operations Office, and 3) representatives from Ortec, Canberra, and Victoreen--who are vendors interested in supplying such monitoring systems for the commercial market.

The SAFE system, which is similar to a sewer-water monitor installed at LRL,²⁰ has sensitivities of the order of 0.1 MPC or better. Table VIII contains representative sensitivities for such a system derived from the results of Ref. 19. The SAFE system is attractive for the following reasons.

- It has sensitivity at the sub-MPC level.
- Is relatively inexpensive.
- Is reliable and needs no constant supervision.
- It has completely automatic recycling and fast protective action should alarm levels be exceeded.
- It provides printed (or plotted) records for later reference and documentation.

TABLE VIII

SENSITIVITIES FOR THE DETECTION OF SELECTED ISOTOPES IN WATER^a

Isotope	MPC (Public Water)		Sensitivity (MPC)
	($\mu\text{Ci/cc}$)	($\mu\text{g/cc}$)	
²³⁵ U	3×10^{-5}	14	0.02 ^b
²³⁸ U	4×10^{-5}	120	0.04
U(Natural)	2×10^{-5}	29	0.03 ^b
²³⁹ Pu	5×10^{-6}	8×10^{-5}	0.4
²⁴¹ Am	4×10^{-6}	1×10^{-6}	0.04

^aFor a 200-sec count at the 3 σ level.

^bEstimated sensitivity.

Fig. 7.

SAFE demonstrator, including a sewer-water system mockup, recording scaler and printer, recycling electronics, and safety interlocks.

IV. DETECTOR AND INSTRUMENTATION DEVELOPMENT

A. Thermal Neutron Coincidence Detector for the Assay of FFTF Fuel Rods—(H. O. Menlove, J. E. Foley, R. A. Forster)

The design and fabrication of the thermal-neutron coincidence counter for the measurement of FFTF fuel rods has been completed. The design parameters were optimized to assay the ^{240}Pu content of the fuel rods which contain ~ 36 g plutonium (12% ^{240}Pu). The detector shown in Fig. 8 contains 32 ^3He tubes (1-in. diam by 20-in. long, 4 atm. gas pressure) in a CH_2 matrix which gives an absolute efficiency of $\sim 50\%$. However, a cadmium liner was added to the fuel-rod channel to reduce thermal neutron multiplication in the fissile isotopes in the fuel rod, and additional cadmium was placed between the detector tubes and the exterior CH_2 shield (6-in. thick) to reduce the neutron lifetime in the detector as well as to shield against external room neutrons. The

Fig. 8.

Thermal-neutron coincidence ^3He detector designed for ^{240}Pu measurement in FFTF fuel rods and plutonium recycle rods. The fuel rod is positioned in the central hole, and the external shield contains a 6-in.-thick layer of paraffin.

resulting efficiency with the cadmium in place is $\sim 36\%$ and the neutron lifetime in the detector was measured to be 28 μsec .

DTF-IV neutronics calculations were performed to determine the multiplication effects of FFTF-type rods in the counter. With cadmium lining the fuel-rod channel, the 22% plutonium-enriched rod yielded a net leakage of 1.00782 neutrons per fission neutron born in the rod while the 26% plutonium-enriched rod yielded a net leakage value of 1.00901. Most of this multiplication effect is due to fast fissions caused by the neutrons before leaving the rod; the remainder is due to neutrons reflected by the detector assembly. If the cadmium is removed, the multiplication effect more than doubles because of thermal fissions.

Figure 9 shows the net coincidence-counting rate as a function of coincidence gate width for an FFTF fuel

Fig. 9.

Net coincidence counting rate from an FFTF fuel rod as a function of coincidence gate time. Also shown is the assay precision (1σ) for 100 sec counting intervals for various gate times.

rod in the detector. Also shown is the assay precision as a function of gate width for 100 sec counting intervals. It can be seen that the best accuracy is obtained for a gate width of $\sim 50 \mu\text{sec}$, and that a counting period of a few minutes results in a statistical precision of better than 1% (1 σ).

The complete assay system includes automated fuel-rod loading and data reduction which is accomplished using a HP-9100B programable calculator together with a BEC-9507 interface module.

In June, 1972, the complete assay system will be taken to HEDL at Richland, Washington for in-plant evaluation.

B. Sealed-Tube Neutron Generator Evaluation--(R. H. Augustson)

The sealed-tube neutron generator has continued to perform well during the routine assay of LASL enriched-uranium inventory, as described earlier in this report. The sealed-tube system, including power supply and control chassis, which had been returned to LASL for testing and diagnostics, was shipped back to United Nuclear Corporation (UNC), Wood River Junction, R.I., and reinstalled in their active assay system. The system is currently being used both in routine assay of incoming shipping containers and in developing techniques for screening and measuring difficult assay configurations, e.g., wet barrels. The system at UNC is now meeting all performance specifications. As part of the continuing evaluation of the Philipps 18602 sealed tube at LASL, delivery is expected soon of an upgraded, pulsed high-voltage power supply for the 18602 sealed tube; this power supply is designed for tube operation at twice the rated target current and 50% duty cycle. This mode of operation is most useful for in-plant pulsed-neutron interrogation and delayed-neutron assay applications.

C. Coincidence Assay Systems for LASL Plutonium Processing Facility--(J. E. Foley)

Final installation and testing of both the 30-gal. and 2-liter neutron coincidence counters²¹ were completed in

February, 1972. Both systems are presently being used at LASL's plutonium processing facility (DP-Site) for routine assay of plutonium-bearing scrap and for assay of plutonium samples to meet AEC inventory measurement requirements. Many types of samples, such as contaminated rags and incinerator ash, are now routinely assayed at LASL with these neutron coincidence counters rather than being estimated or assayed by lengthy radio-chemical techniques. The 30-gal. barrel counter can detect $\sim 5 \text{ mg } ^{240}\text{Pu}$ in 1000 sec at 7200 ft (LASL altitude) or about 2 mg ^{240}Pu at sea level. The sensitivity of the 2-liter counter is $< 1 \text{ mg } ^{240}\text{Pu}$ at LASL. Several hundred samples have been assayed with these counters and good agreement has been obtained with both nondestructive gamma-ray and chemical-assay results when they were available. Typical counting times for samples containing $> 10 \text{ g}$ of plutonium are 200 to 400 sec with assay precision better than 10%.

D. PDP-9/L Computer-Based Data-Acquisition System--(L. V. East)

Work on the expanded version of the FOCAL* interpreter was essentially completed during this reporting period. In addition to the capability of supporting up to four users in a time-share mode, this expanded FOCAL contains such features as double array subscripts, free-format input including the use of expressions, extensive graphics, and additional library file manipulation commands. The writing of a foreground/background routine will begin in early summer to allow real-time analysis of data, acquired through the computer's ADC interface, to be performed by FOCAL programs.

The display control hardware was modified to reset the display digital-to-analog converters 200 msec after the last point is displayed. This modification eliminated an overheating problem in the main display deflection circuits that occurred if the display spot was left at a coordinate other than (0,0) for an extended period of time. The display software was also modified to improve the quality of alphanumeric displays.

*FOCAL is a registered trademark of Digital Equipment Corporation.

V. A-1 FIELD ASSAY OPERATIONS AT OAK RIDGE

Since October, 1971, LASL's Mobile Nondestructive Assay Laboratory (MONAL) has been operating at the USAEC facilities in Oak Ridge, Tennessee. These activities can be divided into three major categories: scrap measurements, plant effluent measurements (see Sec. III-C), and cascade process measurements.

A. Scrap Measurements

A wide variety of scrap materials were measured at Oak Ridge. LASL personnel measured over 500 items in more than 30 different sizes of containers; this points up the versatility of the array of measurement techniques available in the MONAL laboratory. Much of this material is difficult, if not impossible, to sample properly, as required for reliable assay by chemical or mass spectrometer methods; hence such material is not routinely measured, and some not at all. One of the goals of this work is to acquaint operations personnel with the unique capabilities of nondestructive assay in handling such measurement problems.

1. **UF₆ Cylinders.** A collection of miscellaneous UF₆ cylinders comprising the total inventory of a small storage warehouse at K-25 is a good example of the routine measurement service that can be provided by NDA instrumentation. These cylinders were quite old, in poor physical condition, and incompletely characterized because their condition precluded both normal sampling and destructive analytic techniques. K-25 management wished to clean out this building and dispose of the contents through recovery or burial but required better information in order to proceed. LASL personnel moved a Ge(Li) spectrometer system to the warehouse to perform a rapid, on-the-spot survey-type measurement of each item. The items were of about 20 different sizes ranging from small sample bottles to larger feed cylinders with several hundred pounds of material. These measurements were performed under adverse conditions of light, heat, space, and electrical power; yet, in a little over a week the desired survey measurements were completed. With this information the plant was able to begin clearing out this warehouse.

2. **Incinerator Ash.** Incinerator ash is generally of low uranium concentration (< 30%), and is presently sent to other AEC facilities for final recovery. It is blended, sampled, and analyzed for uranium concentration and ²³⁵U enrichment. The material is in sealed 6-in.-diam by

15-in.-high cans. A study was made on 16 of these cans to compare the reproducibility of the nondestructive assay with that of the conventional analytic assay (blending, sampling, and analysis). These cans were subjected to a NaI segmented scan for ²³⁸U content. They were then carefully reblended and sampled twice for analytic assay, and (under a different label) were finally remeasured for ²³⁸U and given a Ge(Li) segmented scan for ²³⁵U content and/or ²³⁵U enrichment.

The average and standard deviation of the two measurements were computed for each technique. A summary of the comparison of the sampling/analytic assay and the nondestructive assay is given in Table IX. The average deviation in percent for an individual measurement is computed from the two measurements made except for the NDA-Gamma, ²³⁵U; this is based on counting statistics since only one measurement of ²³⁵U was made by this technique. The total ²³⁵U assay is the same for both methods with the nondestructive assay showing a higher precision for individual measurements. Table I (cf. section I.A., "Investigation of Sampling/Chemical Assay Using Nondestructive Assay Techniques," of this report) shows the detailed comparison of ²³⁵U measurements for all 16 cans. On total uranium, the gamma assay is 5% lower than the analytic assay. The gamma total uranium assay is based on a NaI scan for ²³⁸U plus a Ge(Li) scan for ²³⁵U and a small correction for the minor isotopes (1% assumed). The calibration is based on a single standard. If this 5% difference is shown to be an actual bias, it can be calibrated out with a more detailed study involving a wider range of standards.

3. **Compressor Blades and Barrier Material.** Samples of used barrier material and compressor blades were measured in the mobile lab using an active delayed-neutron assay. The barrier material is processed to remove most of

TABLE IX

INCINERATOR-ASH ASSAY SUMMARY SAMPLING/ANALYTIC-NDA COMPARISON

	Uranium (g)	Ave. Dev. (%)	²³⁵ U (g)	Ave. Dev. (%)
Sampling/Analytic	13643	2.2	569	3.2
NDA-gamma	13011	2.3	566	2

the residual uranium. It is contained in a variety of large drums, mostly 20 gal and 55 gal. The compressor blades are washed and loaded into large drums. Since there is no good method for sampling and analyzing this material at present, it is not routinely measured. Both of these items are quite heavy and dense and require the penetrability of neutron interrogation and assay. The first item is a 55-gal drum of used, scrap barrier material that had been sampled and assayed to contain 415 g uranium. A neutron assay gave the value of 200 ± 50 and subsequent samples gave analytic assays of 90 g and 45 g. Clearly, it is difficult or impossible to obtain representative samples of this material. The other items listed are two large garbage cans of washed compressor blades. The quoted analytic measurement is obtained by dissolving several sample blades taken from each can and chemically analyzing the solution. Since the active nondestructive assay revealed over 60 times as much material as sampling/chemical assay, it is again evident that sampling is grossly inadequate.

4. Acid-Leached Residues. This material has been heavily leached and is of low uranium concentration (about 1% uranium, 93% ^{235}U). It is contained in 9-in.-diam by 14-in.-high "cream cans." These residues are being shipped off site for recovery, so they require good accountability data. The present procedure for measuring this material is to blend and sample each can, and then use x-ray fluorescence techniques to determine the uranium content to within $\pm 5\%$. This procedure is referred to in following paragraphs as the sampling/analytic measurement. The whole container is also measured on a NaI drum counter.²² If these two measurements do not agree, the can is resampled and analyzed.

Thirty-nine of these cans were measured in MONAL with the Ge(Li) segmented scan, and ten cans were also measured with the active delayed neutron system. A summary of the results on these ten cans is given in Table X.

TABLE X

ACID-LEACHED RESIDUES—COMPARISON
OF ASSAY TECHNIQUES (APPROX. 1% U, 93% ^{235}U)

Technique	Mean	Std. Dev.
Active Delayed Neutron ^a	1.00	0.00
Ge(Li) Segmented Gamma Scan	0.92	0.04
Sampling/Analytic (OR)	1.02	0.16
NAI Drum Counter (OR)	1.05	0.16

^aActive assay taken as best value

There is evidence of inhomogeneity in this material, so the sampling/analytic measurement cannot be taken as the true assay. On the basis of results on several test cans, delayed-neutron assay proved the most accurate and precise of the various measurement methods; hence, it was chosen as the standard of comparison. The numbers given in the table are the mean and standard deviation for the ratios of each of the other three assay values to the delayed-neutron assay.

The mean for the segmented gamma measurements is 0.92, indicating that they were 8% lower on the average, than the active. A single 10-g standard was used to calibrate the system for these measurements. Later measurements on other standards showed that this difference vanished with a more careful calibration and a better range of standards. With this in mind, consider the standard deviation of the various measurements. The neutron and segmented scans correlate very well; in view of the completely different physical basis of these techniques and the essential agreement of their assay results, it seems clear that the active assay should be used as the benchmark assay value. The sampling/analytic assay shows a standard deviation of 15% on individual measurements versus 5% for the nondestructive assay. The Oak Ridge Drum-Counter²² results are nearly as accurate and precise as the sampling/analytic measurement. The drum-counter measurement consists of a NaI scan with no direct correction for gamma attenuation, which is large in these cans. Accordingly, a very careful set of standards has been fabricated for this type of material. These standards were shown to represent accurately the average gamma absorption in the material.

5. Process Residues. This material is similar to the leached residues just described, and is stored in the same cans. It includes material such as crushed firebrick and incinerator ash and is generally of higher uranium concentration (up to 8%). Some 20 cans were analyzed, providing the same set of measurements as above. The results are summarized in Table XI; here again, delayed-neutron assay is taken as the standard of comparison.

6. Air Filters. These are 24-in. by 24-in. by 12-in. absolute filters contaminated with 93% enriched ^{235}U . Although not typical, these filters can contain up to a kg or more of uranium and are difficult to measure other than nondestructively. They can be gamma-scanned but this must be done carefully because of the possibility of high absorption and unevenly distributed material. This can best be done with a segmented gamma scan set up to cover a square grid (e.g., as with present medical scanners).

TABLE XI

**PROCESS RESIDUES—COMPARISON OF
ASSAY TECHNIQUES (0.3 TO 8.0% U, 93% ²³⁵U)**

Technique	Mean	Std. Dev.
Active Delayed Neutron ^a	1.00	0.00
GE(LI) Segmented Gamma Scan	0.98	0.07
Sampling/Analytic (OR)	1.03	0.22
NAI Drum Counter (OR)	1.01	0.17 ^b

^a Active assay taken as best value.

^b Neglects 3 cans with large differences;
neglects original results on 4 remeasured cans.

B. Cascade Measurements

1. Shut-Down Cascade. Since about 1964 the original enrichment cascade at the K-25 site has been shut down. Oak Ridge expressed interest in a measurement of the residual uranium holdup in the equipment of this facility. It was agreed to attempt measurements in two of the building sections near the top of the original cascade.

LASL constructed several instrumented carts for this work; one such cart is shown in Fig. 10. It was built to contain a portable NAI gamma spectrometer. The detector is shielded and collimated and can be telescoped over a fairly wide range of elevations to facilitate the measurement of this large equipment. Both the detector and the electronics are housed in freon evaporative coolers to enable them to be used in the 120°F environment of an operating cascade also.

Preliminary work indicated that most of the activity was concentrated in the seals, pumps, and converters, so these comprised the bulk of the measurements. It also proved that ²³⁵U gamma measurements on the pumps and converters were feasible. A combination of transmission measurements and calculations was used to correct the data for gamma absorption. The detector collimation permits focusing the measurement on specific pieces of equipment. The seal structures were behind too much metal to make reliable gamma measurements so neutron measurements were made instead. A polyethylene-moderated ³He probe was attached to the counter to make these measurements. The (α,n) neutrons from ²³⁴UF₆ were counted and a relatively constant ²³⁴U/²³⁵U ratio was assumed to interpret the data. Table XII summarizes the number of pieces of equipment measured and the percentage of the total equipment represented by this sample. The measurements were completed in about one week.

Fig. 10.
Cooled, portable NAI gamma spectrometer for assay
of ²³⁵U inside an operating gaseous-diffusion plant.

TABLE XII

**SUMMARY OF NUMBER OF COMPONENTS
AND MEASUREMENTS IN BLDGS. 306 AND 305**

	Seals	Pumps	Converters
Bldg. 306			
Total Comp.	1152	1152	576
Comp. Measured	114	156	78
% Of Total Meas.	10	14	14
Bldg. 305			
Total Comp.	1200	1200	600
Comp. Measures	108	36	18
% Of Total Meas.	9	3	3

Based on this sampling, the total uranium holdup in these two buildings was predicted by LASL assayists; this predicted uranium holdup agreed to within a factor of two with an independent Oak Ridge estimate based on laboratory experiments and decontamination data obtained when equipment was removed from the cascade for servicing. This work demonstrated the feasibility of making direct measurements on the residual uranium in a shutdown cascade.

2. Operating Cascade. A portable NaI gamma-assay unit was set up with a strip chart recorder to demonstrate the feasibility of dynamic monitoring of UF_6 retention in NaF traps. Several of these traps are used in series between the cascade and the effluent stack to filter UF_6 from the gaseous effluent of the cascade. NaF is used since it will adsorb UF_6 at one temperature and release it later under heating. The traps are regularly desorbed and rotated. The detector is set to measure the 186-keV activity as a function of time in one of these traps. Figure 11 is a graph of the strip chart data taken over a two-day period. There is no apparent uranium retention during the period it was operated as the second trap from the input; however, the signal shows a steady rise after it was moved to the head of the string. The total rise observed in the second 24-h period was about 2 mg/cm^3 of uranium. The sensitivity of the measurement was $\sim 0.2 \text{ mg/cm}^3$. Additional measurements with this instrument proved that it

Fig. 11.

Uranium concentration as a function of time for a NaF trap (at ORGDP). The trap was operated as the second trap in a series for 24 h then switched to first position in the series. The counting rate was obtained with a NaI system counting 186-KeV gammas from ^{235}U .

could spot lumps of retained uranium. It could be used as a continuous monitor to check trap operation and tell the operator when to switch or desorb a given trap. When a trap is being recharged it could provide a uranium assay of the material being removed. Since these traps occur just ahead of the effluent stack, the instrument could provide information on gaseous effluents and possibly a warning of system failure, excessive stack releases, etc.

Measurements were made to test the feasibility of using portable instrumentation to detect and assay material holdup in the operating cascade. The instruments used were a small NaI detector system under evaluation by operations personnel, the ^3He neutron detector system described earlier, and a collimated 5-in. by 5-in. NaI crystal coupled to a portable gamma-assay system. This device, nicknamed the "elephant gun," is pictured in Fig. 12. As a test ORGDP personnel hid nine 12A UF_6 cylinders within a portion of the operating cascade. These cylinders contained from 150 to 450 pounds of UF_6 . The

Fig. 12.

The "Elephant Gun"—a collimated, 5-in. by 5-in. NaI detection system for the assay of ^{238}U by detection of ^{234m}Pa gamma rays.

large NaI detector was pushed around the plant looking for gamma rays from the ^{238}U daughter $^{234\text{m}}\text{Pa}$. A majority of the cylinders were located in a one-hour hunt. Subsequent measurements showed that the device could unambiguously detect a 440-pound cylinder at a distance of 130 ft with a 30-sec count.

The detector was also used to look for uranium plating or holdup in the main gas pipes. The viewing cone of the detector sees a 5-ft-diam circle at 8 ft; at this distance it can detect 166 g ^{238}U in a 30-sec count. No large holdups were found except in one area where ORDGP plant personnel expected to find significant material.

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