60
23, 73

th. 230

LA-5291-PR

PROGRESS REPORT

32.00

Nuclear Analysis Research and Development

Program Status Report

January - April 1973

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7403-ENG. 36

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

This report presents the status of the Nuclear Analysis Research and Development program at LASL. The four most recent reports in this series, all unclassified, are:

LA-4883-PR
LA-4994-PR

LA-5091-PR
LA-5197-PR

Printed in the United States of America. Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

2/1/01

LA-5291-PR
Progress Report
UC-15

ISSUED: May 1973

Nuclear Analysis Research and Development

Program Status Report

January - April 1973

Nuclear Analysis Research Group, A-1

G. Robert Keepin, Group Leader

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASTER

ABSTRACT

This report presents the status of the Nondestructive Assay R&D program of the LASL Nuclear Analysis Research Group, A-1, covering the period January 1, 1973, through April 30, 1973. The topical content of this program status report is summarized in the table of contents.

CONTENTS

I. Neutron Interrogation (Active) Assay Applications and Results	1
A. Delayed-Neutron Assay of Inventory Verification Samples	1
B. Measurement of the LASL Enriched Uranium Inventory	1
C. Random Driver Measurements at the LASL Uranium Recovery Facility (CMB-8)	1
D. Nondestructive Assay of HTGR-Type Fuel Elements	1
II. Passive Assay Applications and Results	5
A. Passive Gamma-Ray Assays and Services	5
B. Alpha Waste Measurements at the 10 nCi/g Level	5
C. UF ₆ Enrichment Monitor for a Gaseous Diffusion Plant	7
D. Passive Gamma-Ray Assay of Fuel Materials Containing ²³² Th and ²³⁸ U	7
E. Application of the "Elephant Gun" for Assay of Uranium in Combustible Waste	9
III. Instrumentation Development, Calibration Standards, and Measurement Controls	14
A. Portable Neutron Probe, "SNAP"	14
B. The Design of a Multi-Energy Californium Assay System	14
C. NDA Instrument Calibration and Uncertainty Statements	16
D. Evaluation of a Gridded ³ He Spectrometer Tube for Safeguards and Other Nuclear Analysis Applications	17
E. PDP-9/L Computer-Based Data-Acquisition System	19
IV. Materials Identification and Analysis with Stopped Muons	20
References	22
Publications	22

I. NEUTRON INTERROGATION (ACTIVE) ASSAY APPLICATIONS AND RESULTS

A. Delayed-Neutron Assay of Inventory Verification Samples (A. E. Evans, R. H. Augustson, and J. J. Malanify)

Assay of 28 Rover inventory verification samples was completed in February 1973. Comparison of NDA data with preliminary chemical assay data indicates reasonable agreement, i.e., a mean and standard deviation in the 1 to 2% range. More definitive comparison of assay data must await final chemical analysis results on these samples.

The overall performance of the pneumatic small-sample assay system for use with the Van de Graaff¹ was tested by assaying the above-mentioned 28 samples both in the manually operated small-sample assay chamber² and in the pneumatic system. Results indicate that samples may be assayed using the pneumatic sample changer without affecting overall assay accuracy.

Preliminary results of the interlaboratory comparison program for assay of HTGR fuel materials have been received from the New Brunswick Laboratory. Preliminary comparison of the chemical assay results with LASL nondestructive assay indicates excellent agreement on uranium assay of uranium-thorium-carbide microspheres. More definitive analyses of the data on oxide powders are needed to resolve some apparent (few percent) biases in the uranium and thorium assay of the oxide materials.

To improve on counting statistics and more efficiently assay samples containing sub-gram quantities of fissile material, a new high-efficiency "wrap-around" delayed-neutron detector has been designed and constructed. This detector is expected to give a three- to five-fold increase in delayed-neutron counting rate and at the same time exhibit increased signal-to-background ratio due to better shielding. The new detector has been installed and is currently being tested at the small-sample assay station of Group A-1's Nondestructive Assay and Standards Laboratory (NASL).

B. Measurement of the LASL Enriched-Uranium Inventory (R. H. Augustson, J. H. Menzel, and T. L. Atwell)

During this reporting period approximately 300 recovery cans containing 250 kg of 93% enriched uranium have been measured using the delayed-neutron active

assay technique with 14-MeV neutron interrogation.^{3,4} To date, a total of 900 kg (~one ton) of enriched uranium in recovery-can geometry has been measured in the continuing effort to put the LASL inventory on a fully measured basis by the end of FY 73.

C. Random Driver Measurements at the LASL Uranium Recovery Facility (CMB-8) (J. E. Foley)

A Random Driver assay system has been installed at the CMB-8 uranium recovery facility for routine non-destructive assay of highly enriched uranium samples. The assay unit is interfaced to a HP 9100B electronic calculator to provide automatic data reduction capability and instant assay readout. A typical assay sequence consists of a 200-sec count to determine the induced fission rate, followed by a 200-sec ²⁵²Cf "source addition" matrix correction run, resulting in an assay rate of about six samples per hour. Using fast neutron interrogation, the sensitivity of the system (defined as signal equal to twice the standard deviation of the background) is ~12 g uranium for 100-sec assay of the standard 2-gal recovery cans.

Nineteen 2-gal cans of Rover scrap material that had been previously assayed with the 14-MeV neutron generator in Group A-1 were reassayed with the Random Driver to provide a comparison of the two techniques. The results of the comparison are shown in Fig. 1. The 45° slope straight line in Fig. 1 is the line of identical assays between the two techniques. The comparison is seen to be quite good. The estimated standard deviation of a Random Driver NDA measurement is ~50 g uranium.

D. Nondestructive Assay of HTGR-Type Fuel Elements (R. H. Augustson and J. J. Malanify)

With increasing interest in high-temperature gas-cooled reactors, there is commensurate interest in safeguarding the highly enriched uranium fuel used in this generic reactor type in its various fabricated forms. The Nuclear Analysis Research Group, A-1, has undertaken a development program exploring neutron interrogation techniques for NDA of the fissile and fertile contents of whole fuel

Fig. 1.

Comparison of random driver measurements on 2-gal Rover scrap samples with 14-MeV accelerator results.

elements. The results of preliminary studies in this program are reported below.

Figure 2 shows a typical HTGR-type hexagonal carbon block before loading with fuel. The striking features of the 31-in.-high hexagonal block (14 in. across the flats) are the three distinct types of penetrations: 200 1/2-in.-diam fuel holes, 100 5/8-in.-diam coolant holes, and one 2-in.-diam central hole to accommodate lifting gear. Of importance to the development of a neutron interrogation assay technique is the presence of the carbon, acting as a neutron moderator. This is contrasted with the PWR and BWR fuel elements which can be measured without the reactor moderator (H_2O) being present. This complication is partially compensated for by (1) the smaller axial length of the HTGR element allowing for easier handling and (2) accessibility, via reentrant holes, to the interior of the element.

The A-1 Van de Graaff (VDG) provided a subthreshold source of 100- to 600-keV neutrons for the initial measurements in the geometry shown in Fig. 3. Both prompt and delayed neutrons from induced fissions were counted using a bank of ^4He gas tubes for prompt-neutron detection and a standard slab detector for the delayed neutrons. A small ^{235}U fission chamber was inserted into various cooling channels and fuel holes to determine the fission response and cadmium ratio for a 100- to 600-keV neutron source. Since not enough HTGR fuel sticks were available to fill the graphite element (several thousand are needed), PWR fuel rods ($\sim 3\%$ enriched) were used to

Fig. 2.

Photograph of a typical HTGR-type hexagonal carbon block before loading with fuel.

Fig. 3.

General experimental arrangement for active neutron interrogation and assay of HTGR-type fuel element.

simulate the HTGR fuel. In addition, graphite rods were inserted into some of the cooling channels to more closely approximate the graphite loading in an actual fueled HTGR element.

Because of the sensitivity of thermal neutron fission to bead size, it is imperative that the contribution of thermal neutrons to the fission rate be minimized. Measurements of the ^{235}U cadmium ratio were made as a function of position, amount of graphite, and amount of fuel. Figure 4 shows the cadmium ratio as a function of position with the element loaded with 100 PWR rods. The observed cadmium ratio varied from 1.1 to 1.2—this would likely be higher for an actual HTGR element because of the increased amount of graphite. This indicates that the thermal contribution could lead to significant errors in the assay if the bead size (or U/Th ratio) varied from the assay standard to the unknown element. However, because of the accessibility of the cooling holes, the thermal neutron flux can be greatly reduced by inserting poisons such as cadmium or boron rods into these holes. This approach has been used successfully.

The prompt-and delayed-neutron responses as a function of ^{235}U loading have been measured for the case of spatially uniform removal of fuel rods. Figure 5 shows the data for delayed-neutron response using Q monitoring* and internal fission chamber monitoring. The internal fission chamber was located in the central lifting hole of the element; it can be seen that this method of monitoring leads to a linear calibration curve and operationally is quite feasible. A similar curve holds for the prompt-neutron response.

Fig. 4.

Cadmium ratio as function of position in HTGR-type element loaded with 100 PWR rods.

Fig. 5.

Delayed-neutron response vs. loading (HTGR-type fuel element).

*Q monitoring is a normalization procedure based on neutron output of the VDG.

The calibration curve in Fig. 5 is somewhat deceiving in that a nonuniform distribution of fuel will give different results; i.e., the spatial response is not flat in the present configuration. The present spatial distribution of delayed-neutron response is shown in Fig. 6; this response can be flattened by combining different detector locations with rotation of the element, somewhat analogous to the rotation-collimation technique used in passive gamma analysis. Such flattening procedures will be examined in the near future.

Using what has been learned from these initial studies, the program is now concentrating on the utilization of both (D,T) and radioactive neutron sources. The (D,T) source appears promising for assay of thorium content. Measurements with an Am-Li source were encouraging but pointed out the need for decreasing the contribution from $^{18}\text{O}(\alpha,n)$ reactions. A more optimum source will be procured. The VDG measurements just described are providing valuable benchmarks for a recently initiated A-1 calculational program on neutron interrogation and assay of complete fuel elements.

Fig. 6.
Spatial distribution of delayed-neutron response
(HTGR-type fuel element).

II. PASSIVE ASSAY APPLICATIONS AND RESULTS

A. Passive Gamma-Ray Assays and Services (J. L. Parker, T. D. Reilly, T. L. Atwell, R. B. Walton, and J. H. Menzel)

The major non-R&D assays and services performed during this reporting period were:

1. Five recovery cans of miscellaneous uranium waste in support of the LASL inventory measurement program were assayed; each of these required a full transmission-corrected segmented gamma scan.

2. Several cans of resin which had been shipped from another facility to LASL for plutonium recovery were assayed. Besides doing a quantitative assay for ^{239}Pu , a rough check of the plutonium isotopic was also made showing that the ^{240}Pu content was not that assumed for purposes of doing a neutron coincidence assay. This finding resolved a rather vexing difference between shipper and receiver assays.

3. A portable SAM I gamma-ray unit was used to assay ^{235}U (in residual materials from the Rover project) contained in approximately 300 ft of ductwork in LASL Shops Department (Shop 15-SM 102).

4. A total of 58 HEPA secondary filters and 17 4-ft by 4-ft by 6-ft boxes of miscellaneous uranium-bearing waste materials were assayed using the SAM I unit.

B. Alpha Waste Measurements at the 10 nCi/g Level (C. J. Umbarger and R. A. Forster)

Work has begun on developing a detection system for assaying low-density solid alpha wastes (e.g. paper, plastics, rubber, incinerator ash, etc.) in bulk form at and below the recently established 10-nCi/g level. This is the presently accepted⁵ maximum activity level for alpha wastes to be placed in nonretrievable storage (e.g., burial). This work is in support of the alpha wastes repository program of the Division of Waste Management and Transportation, U.S. Atomic Energy Commission. Alpha wastes are defined here as wastes contaminated with certain transuranic elements that are alpha emitters and that are produced in significant quantities in wastes generated by AEC laboratories and production facilities. Table I lists

the transuranic isotopes of major interest in the program along with ^{233}U which is also being considered due to its presence in alpha wastes. The isotope ^{241}Pu is not included in this list because it is primarily a β emitter and, due to its relatively short half-life (14.3 y), soon decays to ^{241}Am which is then included.

To assay bulk wastes at the 10-nCi/g level, a different approach must be utilized than the usual neutron or gamma-ray detection schemes. This is due to the very low gamma-ray and spontaneous fission neutron yields from alpha wastes at the 10-nCi/g level. For example, at this activity level, a 2-ft³ cardboard box containing 4,000 g of typical waste material can contain only 640 μg of ^{239}Pu . For such a bulk sample, this amount of ^{239}Pu , with a 6% ^{240}Pu content, is at least several orders of magnitude below that measurable with presently available barrel and box neutron counters. In addition, for reasonably short counting times (~ 100 sec), passive detection of high-energy characteristic gamma rays (e.g., 414 keV for ^{239}Pu analysis) cannot meet this detectability limit for such bulk samples. High throughputs, hence short assay times, are important here, besides the low detectability limit, because of the large volume of alpha wastes presently being generated in the nuclear industry.

A more sensitive technique than neutron or high-energy ($E_\gamma \geq 100$ keV) gamma-ray detection schemes involves use of a thin NaI (Tl) detector—as in the FIDLER⁶ concept. The NaI (Tl) crystal, 5.0 in. diam by 0.06 in. thick, is calibrated to measure photon yields in the 10-to 100-keV region, thus monitoring primarily the daughter L x rays emitted following at least 4% of the alpha decays of the transuranics. Table I lists these elements along with their half-lives, specific α activities, spontaneous fission neutron yields, daughter L x-ray activities following alpha decay, and the most intense gamma-ray yields. Table I exhibits the large abundance of L x rays—exceeding the number of spontaneous fission neutrons or high-energy gamma rays by three orders of magnitude or more. For samples of low-density wastes (comprising a considerable fraction of the total generated alpha wastes) in low-density containers (e.g., cardboard, plastic, etc.), this 10-to 100-keV detection window provides the highest sensitivity for assay of alpha wastes. Attenuation problems for such low-energy photons are not as severe as commonly thought and can be corrected by attenuation calculations, transmission scans, etc.

TABLE I
SPECIFIC ACTIVITIES OF ISOTOPES FOUND IN ALPHA WASTES

Isotope	Alpha Half-Life (yr)	Alpha Specific Activity (α /sec-g)	Spontaneous Fission Activity (f/sec-g)	Major γ -Ray Activities		L X-Ray Activity (X/sec-g)
				γ Energy (keV)	Activity (γ /sec-g)	
^{233}U	1.6×10^5	3.5×10^8	—	42	6.7×10^5	1.4×10^7
				55	1.4×10^5	
				97	2.1×10^5	
				146	6.4×10^4	
^{239}Pu	2.4×10^4	2.3×10^9	1×10^{-2}	52	3.2×10^5	9.2×10^7
				129	1.3×10^5	
				375	3.5×10^4	
				414	3.5×10^4	
^{240}Pu	6.6×10^3	8.4×10^9	4.1×10^2	45	3.8×10^6	8.8×10^8
				104	8.4×10^5	
				160	8.7×10^4	
^{242}Pu	3.8×10^5	1.4×10^8	7.7×10^2	—	—	1.4×10^7
^{241}Am	4.58×10^2	1.2×10^{11}	—	60	4.2×10^{10}	4.6×10^{10}
				99	2.6×10^7	
				103	2.3×10^7	

Preliminary measurements on shredded paper and rubber gloves doped with calibrated ^{241}Am , ^{239}Pu , and ^{233}U sources show that a detectability limit of less than 10 nCi/g can be obtained in a 10-sec count at the signal-equal-to-background level. Sample sizes were 18 in. diam by 3 in. high containing nearly 400 g of wastes with the NaI(Tl) detector viewing the flat sample from above. At an assay rate of one 400-g sample every 11 sec, over 1000 kg of wastes can be assayed in an 8-hr shift. Incinerator ash samples containing 170 g of ash in an 8-in.-diam by 1/2-in.-high configuration were similarly measured at the sub-10-nCi/g level in 10 sec. At this rate, 445 kg of ash can be measured in an 8-hr workday. Larger sizes (hence greater throughputs) can be accommodated with the system in this "flat waste" geometry, but detectabilities and counting times reported here are based on actually measured samples. For these measurements, the radioactive sources ("spikes") were placed underneath the sample with the detector above the sample. The reported detectability is for the spiked sources located under the outside edge of the sample, giving the worst possible case for activity location. For more uniformly placed activity

(e.g., throughout the sample), the detectability limits are considerably better. For example, Monte Carlo photon calculations using the MCP code⁷ for the 400-g sample of paper and rubber showed that the detection efficiency for 16-keV photons (neglecting the much more penetrating 60-keV ^{241}Am gamma rays found in all plutonium samples) increased by 40% for a volume-distributed source over a plane source under the sample. Other calculations showed that a sample 2.4 times (~ 960 g) the size of the original 400-g sample with a uniform volume-distributed source produced the same signal strength as a plane source (emitting 16-keV photons) placed underneath the 400-g sample. Additional calculations for dry ash and wet ash with varying amounts of water content showed equal promise for measuring larger samples at the 10-nCi/g level.

For more massive samples, 2-ft³ cardboard boxes were made up containing 4,000 g of cheesecloth contaminated with known amounts of plutonium. The boxes were rotated and scanned vertically past the NaI(Tl) detector described previously. For 100-sec counting times, detectability limits at the signal-equal-to-background level were

measured to be in the few tens-of- μg of plutonium range. For 4,000 g of waste, this is below the 10-nCi/g level. Additional measurements are presently underway to determine calibration curves, assay accuracy, etc. For accurate assays, gamma transmission scans of the larger boxes (e.g., using ^{57}Co 122 keV gamma rays) will probably be necessary; these tests are just beginning.

In summary, low-density alpha waste measurements are being performed at the 10-nCi/g level for bulk samples up to 2 ft³. This detectability limit is achieved in short counting times (≤ 100 sec) at the conservative signal-equal-to-background level.

C. UF_6 Enrichment Monitor for a Gaseous Diffusion Plant (T. D. Reilly, E. R. Martin, J. L. Parker, and R. B. Walton)

The UF_6 enrichment monitor for the Goodyear Atomic Gaseous Diffusion Plant was completed during this period.^{8,9} The system continuously measures % ^{235}U and % ^{234}U in liquid UF_6 at the product withdrawal

station. The ^{235}U measurement is based on counting the 185 keV gamma from this isotope, and the ^{234}U measurement is based on counting neutrons from $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ with ^{234}U being the major alpha source. Figure 7 shows the details of the detector and sample chamber assembly. The overall system (see Fig. 8) includes a LASL-designed arithmetic unit which scales the counter data and computes % ^{235}U and % ^{234}U ; assay results are displayed (LED) and recorded (teletype printer) directly in % ^{235}U and % ^{234}U . A preliminary operating manual has been written for the system. All materials have been received by Goodyear and will be installed by LASL in mid-May.

D. Passive Gamma-Ray Assay of Fuel Materials Containing ^{232}Th and ^{233}U (L. V. East)

Since fuels containing ^{232}Th and ^{233}U are beginning to play an ever more important role in the nuclear power industry, it was decided to re-examine the possibilities and limitations of passive gamma-ray assay of these materials. Fuel containing ^{233}U also contains small

Fig. 7.
 UF_6 neutron counter and gamma enrichment meter assembly.

Fig. 8.

UF₆ enrichment monitor system: detector and sample chamber assembly, electronics cabinet, and teletype.

concentrations (~5 to 50 ppm) of ²³²U. Both ²³²Th and ²³²U decay ultimately to ²⁰⁸Pb by a series of α and β emissions. The gamma-ray spectra resulting from these decay chains contain ~40 discrete gamma-ray lines. Through a series of α and β emissions, ²³³U also decays to ²⁰⁹Bi but produces a somewhat less complicated gamma-ray spectrum.

The gamma-ray spectrum obtained from a ²³²Th sample with a high resolution Ge(Li) detector is shown in Fig. 9. All of the gamma rays observed in Fig. 9 are from ²³²Th daughter products, most notably ²²⁸Ac, ²¹²Pb, ²¹²Bi, and ²⁰⁸Tl. The ²³²Th decay chain may be approximated by

since the daughters between ²²⁸Th and ²⁰⁸Pb have half-lives of a few days or less. The time required for the daughter activities to reach equilibrium is determined by the half-lives of ²²⁸Ra (6.7 years) and ²²⁸Th (1.91 years), and is on the order of 35 years.

During the chemical processing of ²³²Th for the fabrication of fuel material, all daughter products *except* ²²⁸Th will be removed. The activities as a function of time of ²²⁸Ac and daughters below ²²⁸Th (i.e., ²⁰⁸Tl) will be approximately as shown in Fig. 10. This difference in gamma-ray intensities as a function of time could be used, in principle, to determine the time elapsed since the chemical processing of fuel containing unirradiated ²³²Th. For example, one might use the intensity ratio of the 277.4-keV ²⁰⁸Tl gamma ray to the 270.2 ²²⁸Ac gamma ray as an indicator of the sample "age." Indeed, such an intensity ratio could provide a rather sensitive indication of the age in the range of a few weeks to three years or so;

Fig. 9.

Gamma-ray spectrum obtained from a thorium salt sample.

Fig. 10.
Relative activities of ^{228}Ac and ^{208}Tl as a function of time after chemical purification of ^{232}Th .

after about four years the ratio becomes rather constant, and after about eight years it begins to increase slightly.

In general, it would appear that the ^{208}Tl gamma rays are well suited for ^{232}Th assay,* provided that the age of the sample is known (or is the same as that of a standard) and that no ^{232}U is present. In particular, the 2.614-MeV gamma ray from ^{208}Tl (not shown in Fig. 9) is very penetrating, and detectable either with NaI or Ge(Li). The 510.7-keV and 583.1-keV gamma rays should be used with some caution. The 583.1-keV gamma ray could be interfered with by a gamma ray of approximately the same energy resulting from the reaction $^{19}\text{F}(\alpha, n)^{22}\text{Na}$, and the 510.7-keV gamma ray could be interfered with by 511-keV annihilation radiation resulting from $^{22}\text{Na} \beta^+$ decay or the interaction of high-energy gamma rays. The presence of the $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ reaction could be determined by looking for the 1.275-MeV gamma ray from ^{22}Na decay.

If the presence of ^{232}U is suspected (as would be, for example, if the sample contains ^{233}U), then the ^{228}Ac gamma rays could be used to determine the ^{232}Th content, as pointed out previously,¹⁰ again provided that the sample age is known. The presence of ^{232}U would prevent an age determination as suggested above, or at least make it much more difficult.

The α decay of ^{232}U to ^{228}Th , gives rise to the same ^{212}Pb , ^{208}Tl , etc., gamma rays as the decay of ^{232}Th . Since

*Indeed, they have been used for such measurements for many years.

the α decay of ^{232}U proceeds predominantly to the ground state and first three excited states of ^{228}Th , none of the high-energy (greater than about 300 keV) gamma rays associated with the decay of ^{228}Ac are observed in the decay of ^{232}U . Figure 11 shows the gamma-ray spectrum obtained from a ^{233}U sample containing less than 50 ppm of ^{232}U . The gamma rays from the ^{232}U daughters are clearly visible, as are the weaker 57.6-keV and 129.1-keV gammas directly associated with the decay of ^{232}U .

Unlike ^{232}Th and ^{233}U , most of the gamma rays from ^{233}U are a result of the decay of ^{233}U itself rather than its daughters. Although the gamma rays from this decay are not particularly intense, many of them are clearly visible in Fig. 11. Gamma rays from the daughter products ^{229}Th and ^{223}Fr are also visible in Fig. 11, indicating that the sample was rather old.

In fuel material containing ^{232}Th and ^{233}U , the ^{208}Tl gamma rays will be predominantly from the decay of ^{232}U because of its relatively short half-life of 72 years. For example, in fuel material composed of 90% ^{232}Th and 10% ^{233}U containing 10 ppm ^{232}U , 95% of the ^{212}Pb , ^{212}Bi , and ^{208}Tl gamma rays would result from ^{232}U decay and only 5% from ^{232}Th .

It is planned to investigate in detail the use of various gamma rays and gamma-ray ratios to make a complete assay of ^{232}Th - ^{233}U fuel materials, including sample age and ^{232}U content determination.

E. Application of the "Elephant Gun" for Assay of Uranium in Combustible Waste (R. B. Walton and E. I. Whitted)

NDA methods for the assay of combustible uranium waste generated in the production of low-enriched uranium are needed for efficient screening for burial, for criticality safety in recovery, as well as materials accountability. Waste package volumes are typically 55 gal or larger.

The "elephant gun," which was used to evaluate hold-up in a gaseous diffusion enrichment plant,^{11,12} has been applied to the assay of 4-ft by 4-ft by 3½-ft plywood boxes of low-enriched uranium waste. This detection system consists of a 5-in. by 5-in. NaI detector enclosed in a 2-in.-thick lead collimator and linked to a portable electronic unit. The delineation of an assay method for these boxes, which are used by the General Electric Co., Wilmington, North Carolina, is an example of a method which is generally applicable to low-enriched uranium waste material.

The principle of the method is detection of the 767- and 1001-keV gamma rays from ^{234m}Pa (a daughter of ^{238}U), which provides quantitative assay of ^{238}U when the

Fig. 11.
Gamma-ray spectrum obtained from a ^{233}U sample.

uranium has aged three months or more after chemical processing. The mean free path for these gammas in combustible waste ($\rho \approx 0.2 \text{ g/cm}^3$) is approximately 30 in., and their mass absorption coefficients are essentially invariant over the range of medium and low-Z matrices. A large NaI detector is preferred because of the low specific activity, $\sim 100 \text{ g/g } ^{238}\text{U}$. An energy window from 680 to 1120 keV was selected for counting with the single channel analyzer (SCA) on the portable electronic unit. This window includes not only the uncollided gammas, but also some of those Compton-scattered by the matrix.

Figure 12 is a photograph of the elephant gun and the "standard" box used for calibration. To minimize gamma-ray self-absorption, measurements are made with the detector viewing across the thinnest dimension of the box, i.e., 3½ ft. Effects of inhomogeneities are minimized by: (a) locating the detector at a distance from the box which is large compared to its thin dimension, in this case a spacing of 80 in. between the NaI crystal and the near side of the box, and (b) averaging two measurements made with opposite sides of the box facing the detector, i.e., 180° rotation of the box.

Fig. 12.
Experimental setup for assay of 3½-ft by 4-ft by 4-ft boxes of low enriched uranium waste using the LASL elephant gun.

Development of the assay method included the following steps:

1. Calibration and measurement of the gamma-ray attenuation correction factor (CF) using a "standard" box containing a typical weight of combustible material.

2. Comparison of experimental results with Monte Carlo gamma-ray transport calculations.

3. Calculation of CF vs box weight to provide calibration for a wide range of box weights. For this development the boxes are assumed to be completely full of homogeneous combustibles with gamma-ray attenuation properties of CH_2 ; consequently, the linear gamma-ray coefficients used for the calculations could be derived from the net weight.

For uniform loadings of uranium less than 5 kg, the gamma-ray self-absorption of the uranium could be neglected.

The "standard" box was designed to simulate uniform distribution of matrix and uranium without the expense and handling problems of a large box filled with synthetic, uranium-contaminated combustible waste. A top view of the box, referred to as the line-source standard box, is shown in Fig. 13. Three aluminum cylinders, into which 7-in.-diam cans of synthetic waste standards may be inserted, are secured inside a plywood box with cables. Rags (650 lbs) packed in the box simulate the combustible waste. Five standard cans, containing a total of 750 g

of uranium thinly dispersed in shredded Kimwipes, were used for the calibration. Measurements were performed with the five cans stacked vertically in each of the five positions available with 180° rotation of the box, and the calibration was obtained by averaging these results. The CF was obtained by taking the ratio of the calibration for the empty box to that for the box filled with rags. Corrections for the self-absorption of the standard cans were applied to obtain CF for a line source standard (LS).

Gamma-ray calculations for this study were performed with the Los Alamos Monte Carlo Program, MCG.¹³ Results for a homogeneous standard (650-lb) box and a line (5 in number) source standard box, i.e., the actual standard, showed the flux from the line source standard to be higher by only 1%; hence, the LS standard box adequately simulates a homogeneous standard. Since the Monte Carlo code readily handles a point isotropic detector, the effects of the angular response (or collimation function) of the elephant gun were computed numerically. This effect reflects an increase of 7% in the flux from the LS standard relative to that from the homogeneous box. Thus, the total correction required when using the LS standard box instead of a homogeneous standard is 8%.

The complete formula for obtaining the ^{238}U mass in an unknown box, M_x , from the observed counting rate $R_x(\text{cpm})$ is:

$$M_x = 1.08 M_{LS} \frac{R_x}{R_{LS}} \frac{CF'_x}{CF'_{LS}}$$

where:

M_{LS} = the ^{238}U mass of the LS standard box

R_{LS} = measured counting rate for LS standard box

CF'_x = Monte Carlo result for the CF of the homogeneous unknown

CF'_{LS} = Monte Carlo result for the CF of a homogeneous box with the same weight as the LS standard

The measured value of R_{LS}/M_{LS} was 720 cpm/kg ^{238}U -sec compared with a background (at sea level) of ~ 300 cpm.

Figure 14 shows CF for the standard box weight vs the lower level of an energy counting window, the upper level being fixed at 1120 keV. The Monte Carlo results were obtained for a homogeneous standard and the experimental data were measured for the LS standard box. Since CF is a ratio of fluxes, the effect of the collimation

Fig. 13.

Standard box fabricated for calibration of the elephant gun method used in assaying large boxes of uranium waste. Seven-inch o.d. cans of synthetic uranium standards (not shown) are inserted in the holes. Rags are used to simulate combustible waste matrix materials.

Fig. 14.

Attenuation correction factor vs lower-level energy window for the standard weight. Results of Monte Carlo calculations are compared with values measured with the standard box.

function on the comparison of CF for line source and homogeneous configurations is negligible.

Values calculated for CF (i.e., CF') versus box weight (of CH_2) are shown in Fig. 15. Data for two energy counting windows, as well as for the uncollided flux, are presented. The reduction in CF' for the lower biases is caused by Compton scattering in the matrix material. With a high resolution Ge(Li) detector the uncollided flux could be measured directly.

To estimate the effect of matrix materials other than CH_2 , calculations were performed for SiO_2 and the results are compared in Fig. 16.

A semiempirical relationship sometimes used to estimate CF is $\mu x[1 - \exp(-\mu x)]^{-1}$, where μ is the linear attenuation coefficient and x is the sample thickness. Figure 17 compares the Monte Carlo results for CF (680 to 1120 keV counting window) vs weight and those obtained by normalizing the above expression to the Monte Carlo value at the standard weight. This extrapolation

Fig. 15.

A plot of attenuation correction factor vs net weight (CH_2) of box: for the uncollided flux and for two energy counting windows, both with an upper limit of 1120 keV.

procedure introduces significant errors for boxes with weights differing greatly from the standard weight.

In practice, the largest errors in this assay method are caused by heterogeneities in the distribution of uranium and matrix materials. Estimates of the error in the measured ^{238}U mass have been made for the following examples of heterogeneities:

1. -14%-If the uranium is completely surrounded by 1/8 in. of iron.
2. -10%-If all the uranium is located within a few inches of the bottom of the box.
3. -~55%-If the uranium is in the form of individual pellets.

Fig. 16.

A plot of correction factor vs net weight of box for both CH₂ and SiO₂. The energy counting window is 680 to 1120 keV.

A more detailed description of this assay method will be available in a separate LASL report. Additionally, a field test of the method, in which a large number of boxes were assayed, has been conducted by the General Electric Co., Wilmington, North Carolina.¹⁴

Fig. 17.

Comparison of Monte Carlo results for the attenuation correction factor (680 to 1120 keV counting window) and those obtained by normalizing a semiempirical expression to the Monte Carlo value at the standard weight. Box contents are CH₂.

III. INSTRUMENTATION DEVELOPMENT, CALIBRATION STANDARDS, AND MEASUREMENT CONTROLS

A. Portable Neutron Probe, "SNAP" (R. B. Walton and T. L. Atwell)

A shielded neutron assay probe (SNAP) has been designed for use with commercially available portable electronic units, e.g., Eberline SAM I and Ludlum Model 22. The SNAP detector consists of two (or one) 1-in.-diam by 5½-in.-long (active), 4-atm ^3He counters contained in a 3-in.-diam, 9-in.-long polyethylene cylinder which is surrounded by 0.030-in. cadmium sheet. This probe may be used without shielding, or placed inside a directional shield, as shown in Fig. 18. The weight of SNAP, including shield, is approximately 20 lbs, which is adequately portable. A photograph, Fig. 19, shows a SNAP system, complete with an Eberline electronic unit.

Fig. 18.

Assembly drawing of the portable shielded neutron assay probe (SNAP).

Fig. 19.

A portable shielded neutron assay probe (SNAP), complete with an Eberline SAM I electronic unit.

The detection efficiency of SNAP (with shield) for a point-neutron source at one meter is 9.7×10^{-5} , 8.3×10^{-5} , and 6.6×10^{-5} for $^{238}\text{PuLi}$ ($\bar{E}_n \cong 0.65 \text{ MeV}$), $^{238}\text{PuLiF}$ ($\bar{E}_n \cong 1.1 \text{ MeV}$), and ^{252}Cf ($\bar{E}_n \cong 2.2 \text{ MeV}$), respectively. As a function of radial distance r from the axis of the detector, the efficiency has been found to vary as $(r + 1)^{-2}$, where r is measured in inches and $r \geq 5 \text{ in.}$ When performing measurements in close proximity to surfaces that are highly reflective to fast neutrons, e.g., concrete, the efficiency will decrease less strongly than $(r + 1)^{-2}$. The directional sensitivity of SNAP, measured with a $^{238}\text{PuLi}$ source as a function of angle around the detector is presented in Fig. 20. SNAP provides approximately 120° of sensitivity in the forward direction and exhibits a shielding factor of greater than 4.2 for background neutrons impinging at angles between 80° and 280° . For source neutrons of higher mean energy than 650 keV, the shielding factor is somewhat less; e.g., it is about 2.25 for ^{252}Cf source neutrons.

Current applications research involving SNAP includes verification of UF_6 cylinder contents, UF_4 "green" salt, and metallic plutonium buttons.

B. The Design of a Multi-Energy Californium Assay System (R. A. Forster and H. O. Menlove)

The design of a neutron tailoring assembly for ^{252}Cf to produce various fast-neutron spectra for the interrogation

Fig. 20.

Directional sensitivity of SNAP as a function of angle, as measured in the midplane of the detector with a PuLi point source ($E_n \cong 0.65$ MeV).

of small samples, as well as fuel pellets and rods, has been completed. The design criteria for the neutronics of the system were:

1. Avoidance of thermal neutron interrogation to minimize neutron self-absorption effects (i.e., cadmium-covered irradiation channel).
2. Capability of both subthreshold irradiation (i.e., below the fission threshold of fertile materials) and superthreshold irradiation.
3. Subthreshold irradiation must be as "hard" (super-resonance) as possible with a $^{235}\text{U}/^{238}\text{U}$ fission ratio of ~ 1000 to permit assay of low-enrichment material.
4. Discrimination ratio between fertile and fissile components must be greater than 20.

Previous experience in development of the FFTF fuel rod assay system¹⁵ provided useful guidance in

determining the present system design. The FFTF system consisted of a ^{252}Cf source surrounded by a cylinder of tungsten (4 cm diam by 4 cm long) in the center of a beryllium cylinder with the irradiation channel in a nickel reflector. Beryllium has an intermediate slowing down power ($\xi\Sigma_s$) of 0.16 which is required for a system designed to reduce fission spectrum neutrons below ~ 1 MeV without thermalizing the majority of them. The $^{235}\text{U}/^{238}\text{U}$ fission ratio of the FFTF system is ~ 400 .

The MCN program¹⁶ was used for the present design calculations which examined neutron spectral shifts along the irradiation channel, flux gradients, and fissile/fertile fission ratios. In addition, various system parameters were investigated including moderator material, moderator size, source position, gamma-ray shielding material around the source, and nickel reflector size.

As a result of these studies, three major modifications were made to the FFTF system: (1) the source was moved radially off-center in the beryllium cylinder, (2) an axial segment of nickel was inserted in place of the beryllium in the immediate vicinity of the source, and (3) the beryllium cylinder was entirely surrounded by nickel reflector. The calculations confirmed quantitatively the following expected results: (1) moving the source produces a softer spectrum than the FFTF system when the irradiation channel is diametrically opposite the source; (2) conversely, the substitution of the nickel segment for beryllium, which displaces moderator in the vicinity of the source, hardens the neutron spectrum axially near the source, and (3) the nickel reflector significantly increases the epithermal flux in the system.

The final moderator design shown in Fig. 21 incorporates all of the above modifications. To reduce the amount of instrumentation required, the system will have only one irradiation channel. Thus, the central core assembly is rotated to produce the desired spectrum. This "dial-a-spectrum" technique is illustrated in Fig. 22, which shows the calculated neutron spectrum for $\theta = 0^\circ$ (hardest spectrum), $\theta = 90^\circ$, and $\theta = 180^\circ$ (softest spectrum) where θ is defined in Fig. 21. The spectra in Fig. 22 yield values for the $^{235}\text{U}/^{238}\text{U}$ fission ratio (for equal masses and neglecting ^{235}U self-shielding effects) of 36, 550, and 1100 for $\theta = 0^\circ$, $\theta = 90^\circ$ and $\theta = 180^\circ$, respectively. For $\theta = 180^\circ$, there is a factor of almost three in the improvement of the $^{235}\text{U}/^{238}\text{U}$ fission ratio over the FFTF system with a loss of fissile fissions per source neutron of $\sim 30\%$ because of the offset source. The maximum expected discrimination ratio between ^{235}U and ^{238}U indicated from calculations is ~ 30 for small samples (< 5 cm in length). The calculated average energy of fission for the three positions in the 5-cm central portion of the channel is 3 keV, 50 eV, and 25 eV, and the percentages of ^{235}U fissions occurring at energies greater than 0.1 MeV are 35%, 3.6%, and 1.2%.

Fig. 21.

Schematic diagram showing a cross section of the cylindrical moderating and shielding assembly for the multi-energy californium assay system. The beryllium cylinder containing the source and nickel segment are designed to rotate through $0^\circ \leq \theta \leq 180^\circ$ —in this view, $\theta = 180^\circ$.

Fig. 22.

Calculated average neutron spectra in the central 5 cm of the irradiation channel as a function of θ (see Fig. 21). The error bars represent 10% uncertainties.

The calculations have shown the magnitude of the flux gradient along the irradiation channel as a function of θ which will be important for the accurate assay of small samples. In addition, the neutron spectra do not change

appreciably along the channel for $\theta > 90^\circ$, but are fairly sensitive for $\theta \sim 0^\circ$. Thus, a scanning procedure might be required for some samples in lieu of a stationary irradiation.

This system, which will use delayed gamma rays plus possibly delayed neutrons as the signature, will have several important applications: (1) a two-spectrum irradiation (e.g., $\theta = 0^\circ$ and $\theta = 180^\circ$) would permit the measurement of the fissile and fertile content of unknowns using standards; (2) this technique could be extended to determine two fissile components and the fertile component by performing three irradiations—one irradiation at $\theta = 0^\circ$ and two softer irradiations (e.g., $\theta > 90^\circ$), either or both using resonance foil self-removal to change the response of the fissile components; (3) fissile measurements of highly enriched samples could be performed at $\theta \cong 0^\circ$ to reduce the self-shielding effects; and (4) the multi-spectra feature would permit more than one determination concerning differences between the standards and the unknowns (e.g., matrix activation, impurities, or isotopic compositions).

A complete NDA system incorporating the capabilities and features described herein is currently under construction at the NASL.

C. NDA Instrument Calibration and Uncertainty Statements (D. B. Smith and R. A. Forster)

The effect of the uncertainty in the calibration curve generated for an NDA instrument on the propagated uncertainty in a single assay has been discussed in previous progress reports.^{17,18} This unavoidable uncertainty in the calibration curve must also be included in any proper estimate of the uncertainty in the total quantity of material contained in an aggregate of samples assayed in the instrument using the same calibration curve, since all of the individual assay measurements are correlated through the calibration measurements. Thus, the expression for the variance of the total involves not only a summation of the variances (square of the standard deviation) of each assay but also a summation of terms containing the variances and covariances of the calibration parameters.

Consider a NDA instrument for which the calibration curve contains m parameters: the assayed quantity M_j of the j th sample is obtained from the relation

$$M_j = F(R_j; a_1, a_2, \dots, a_m) ,$$

where R_j is the measured response of this sample, and the a_i 's are the calibration parameters. The total of n samples

using this curve (i.e., the same set of values of the a_i 's) is given by

$$M_T = \sum_{j=1}^n M_j$$

with a statistically estimated variance of

$$S_T^2 = \sum_{j=1}^n S_{M_j}^2 + \sum_{j=1}^n \sum_{\substack{q=1 \\ j \neq q}}^n \sigma_{M_j M_q} \quad (1)$$

where $S_{M_j}^2$ is the statistically estimated variance of M_j ,

$$\sigma_{M_j M_q} = \sum_{i=1}^m \sum_{k=1}^m \frac{\partial F(R_j; a_1, a_2, \dots, a_m)}{\partial a_i} \sigma_{ik} \frac{\partial F(R_q; a_1, a_2, \dots, a_m)}{\partial a_k},$$

and

$$\sigma_{ii} = S_{a_i}^2 \quad \text{and} \quad \sigma_{ik} = \text{cov}(a_i, a_k)$$

are the variances and covariances respectively of the calibration parameters (estimated in the least-squares fitting procedure used to obtain the calibration curve).

Since the fractional uncertainty in the total (S_T/M_T) due to the uncertainty in each individual assay decreases as the number of samples included in the total becomes large, the additional uncertainty* caused by the correlation of the measurements through the calibration curve [i.e., the double sum term in Eq. (1)] places a lower limit on the fractional uncertainty in the total and thus will dominate any statement of LE MUF that is made concerning the inventory.

While Eq. (1) appears rather formidable, the necessary double summation can easily be performed on a small computer. Furthermore, if the R_j 's in Eq. (1) are all approximately equal, the expression simplifies to

$$S_T^2 = \sum_{j=1}^n S_{M_j}^2 + n(n-1)F_0,$$

where F_0 is a constant given by

$$F_0 = \sum_{i=1}^m \left[\left(\frac{\partial F}{\partial a_i} \right)^2 S_{a_i}^2 + \sum_{\substack{k=1 \\ k \neq i}}^m \text{cov}(a_i, a_k) \left(\frac{\partial F}{\partial a_i} \right) \left(\frac{\partial F}{\partial a_k} \right) \right].$$

*i.e., over and above the uncertainty as usually expressed by the first term on the right-hand side of Eq. (1).

For example, if $m = 2$ (i.e., only two calibration parameters),

$$F_0 = \left(\frac{\partial F}{\partial a_1} \right)^2 \sigma_{a_1}^2 + \left(\frac{\partial F}{\partial a_2} \right)^2 \sigma_{a_2}^2 + 2 \text{cov}(a_1, a_2) \left(\frac{\partial F}{\partial a_1} \right) \left(\frac{\partial F}{\partial a_2} \right),$$

and the computations necessary to obtain S_T^2 can be performed on a desk calculator.

The above estimate of the variance of the assay results assumes, of course, that the standards used for calibration do indeed represent the unknown samples in all physical characteristics, which can influence the assay results as measured by the particular NDA instrument.

D. Evaluation of a Gridded ^3He Spectrometer Tube for Safeguards and Other Nuclear Analysis Applications (A. E. Evans and L. V. East)

In the continuing development of new instrumentation for nuclear safeguards and materials accountability, a gridded ^3He ionization chamber neutron spectrometer tube of the type developed by Shalev¹⁹ was purchased from the Technion Research and Development Foundation Ltd., Israel. This device was set up and tested using monoenergetic $^7\text{Li}(p,n)$ neutrons from the A-1 Van de Graaff accelerator.

The spectrometer tube is a stainless steel cylinder ~5 cm diam by 20 cm long filled with a mixture of 5.1 atm of ^3He , 2.5 atm argon, and 0.4 atm of CH_4 . A grid assembly consisting of 25 wires at a radius of 0.5 cm from the central anode is maintained at 550 V, while the anode potential is set at 3,000 V. The ends of the spectrometer tube are capped with rather massive demountable ultra-high-vacuum flanges, and a heavy steel drying tube and valve are left attached to the filling tube. These fixtures may bear upon the use of the device, as explained below. The entire spectrometer is enclosed in cadmium and 1/4-in. Boral shields to reduce the thermal neutron response of the detector.

The pulse-height distribution from a ^3He spectrometer has three neutron-induced components, as illustrated in Fig. 23: a peak due to the reaction $n + ^3\text{He} \rightarrow ^3\text{H} + p + 770 \text{ keV}$, causing ionization equivalent to the neutron energy + 770 keV within the spectrometer tube; a continuum at pulse heights less than the peak due to wall effect, i.e., loss of energy due to ion collisions with the wall of the chamber; and a continuum from zero pulse height up to three-fourths of the energy of the neutron due to elastic scattering of neutrons by ^3He . In addition, there is the ever present thermal neutron peak due to the relatively high thermal cross section of the $^3\text{He}(n,p)$ reaction, as well as a continuum of < 300-keV pulses due to gamma radiation. Cuttler et al²⁰ showed that the ^3He

Fig. 23.

Pulse-height distribution from the Shalev ^3He spectrometer for 1.0-MeV neutrons. The upper curve is the ungated spectrum showing relative contributions of $^3\text{He}(n,p)^3\text{H}$ reaction peak, wall effect, ^3He recoil continuum, and gamma radiation. The lower curve is the same distribution gated with a pulse-shape analyzer to eliminate the ^3He recoils, gamma radiation, and some of the wall effect and pileup. The small peak in the vicinity of channel 500 is due to 499-keV neutrons from the $^7\text{Li}(p,n)^7\text{Be}^*$ reaction.

recoils have a faster risetime than the $^3\text{He}(n,p)$ pulses and thus can be separated from the $^3\text{He}(n,p)$ pulses by pulse-risetime analysis. Pulse-risetime analyses can also reduce peak broadening due to differences in ionization-track orientation and can reduce wall and pileup effects. In Fig. 23 the lower curve is a pulse-height distribution for 1-MeV neutrons gated with a pulse-shape analyzer signal to eliminate ^3He recoils and other extraneous effects. Such a system of pulse-shape discrimination is particularly necessary for the study of complex or continuous spectra containing any neutrons above 1.2 MeV, at which energy ^3He recoil pulses equal the pulse height of (n,p) reactions for thermal neutrons.

The response of the detector to monoenergetic neutrons of energies from 0.2 to 1.5 MeV is shown in Fig. 24. Each peak represents a fluence of 0.96×10^6 neutrons/cm² through the detector.

The use of the spectrometer for measurements below 200 keV appears somewhat hindered by the presence of low-energy peaks in the spectrum induced apparently by the scattering of higher energy neutrons. These peaks have been observed both in continuous neutron spectra such as those shown in Fig. 25 for $^{238}\text{Pu-Li}$ and $^{238}\text{Pu-Li-F}$

Fig. 24.

Response of the Shalev spectrometer to monoenergetic neutrons of energies from 0.2 to 1.5 MeV.

Fig. 25.

Pulse-height distributions from $^{238}\text{Pu-Li}$ and $^{238}\text{Pu-Li-F}$ neutron sources. The three peaks just above the thermal peak, corresponding to neutron energies of 71, 126, and 168 keV, appear also in pulse-height distributions from 400-keV monoenergetic neutrons.

neutron sources, and also in the pulse-height distributions for monoenergetic neutrons. The phenomenon appears to be resonant for 400-keV incident neutrons. Placing two inches of iron between the source and the spectrometer greatly enhances the effect. It has tentatively been concluded that these peaks are due to scattering in the walls and/or flanges of the spectrometer.

The peak efficiency of the spectrometer is shown in Fig. 26. The $^3\text{He}(n,p)^3\text{H}$ cross section increases above 0.75 MeV, and one might therefore expect the spectrometer efficiency to do likewise. However, the wall effect is also increasing with increasing neutron energy, and the net result is a peak efficiency decreasing with increasing neutron energy. The dip at 400 keV is doubtlessly due to resonant scattering in the walls of the detector.

The Shalev ^3He spectrometer must be used with considerable care; it is highly microphonic, it requires a low-noise preamplifier with FET input and a low-noise, low-ripple, high-voltage source. Amplifier time constant requirements (6 to 10 μsec for best resolution) may somewhat limit the rate of data acquisition. Also gamma-ray pileup effects and related considerations regarding thermal neutron shielding require further investigation.

Fig. 26.

Peak efficiency of the spectrometer as a function of neutron energy.

Properly used, the ^3He spectrometer promises to be a powerful new tool for the solution of many problems in neutron spectrometry. The high-resolution, nearly monotonic response functions will enable the study of complex neutron line spectra such as those from delayed neutrons^{21,22,23} in cases where time-of-flight spectrometry is not applicable. This instrument can also be used to study continuous neutron spectra, as for example reactor-shield leakage spectra. The first task to be undertaken with the spectrometer will be a study of isotopic neutron sources that are used, or planned for use, in nuclear nondestructive assay systems.

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

The programming system based on the use of an expanded FOCAL* interpreter has now been in routine use for over eight months, and appears to fulfill quite well Group A-1's need for high-level programming language for real-time data acquisition and reduction. A FOCAL program has been written to perform rather extensive analyses on gamma-ray spectra, including least-square fitting of multiple Gaussian functions to photo peaks. An additional 8192 words of core memory have been ordered to provide expanded data and program storage capability.

During this reporting period, the system has been used extensively for neutron spectra measurements, x-ray fluorescence studies, passive gamma-ray assay development, and ^{12}C and $^{13}\text{C}(p,\gamma)$ studies.

*FOCAL is a registered trademark of the Digital Equipment Corp.

IV. MATERIALS IDENTIFICATION AND ANALYSIS WITH STOPPED MUONS

(J. J. Malanify, C. J. Umbarger, R. H. Augustson,
L. V. East, and D. A. Close)

The primary goal of the NASA Space Radiation Effects Laboratory (SREL) measurements²⁴ was to demonstrate the feasibility of a new NDA technique based on the characteristic elemental and isotopic differences in muonic x-ray spectra.

Figure 27 is a schematic of the geometry employed. The incident muon beam is made to stop in the target (T) and the resulting x rays are observed in a Ge(Li) detector (D). The beam counters, S_1 and S_2 , define charged particles in the beam. The absorber (A) is chosen so as to maximize the number of muons that stop in the target. The counter S_3 identifies a muon entering the target and S_4 is used in anti-coincidence to eliminate those events when the charged particle continues on through the target.

Figure 28 shows a complete spectrum for ^{208}Pb . The scale here is approximately 1 keV per channel. Therefore the figure displays x rays up to about 8 MeV. The sharp single peak in channel 6680 is a calibration pulser. The strong peaks between channels 4450 and 5600 are the muonic K_α transitions. The muonic L x-ray transitions are between channels 2275 and 2450.

Figure 29 displays the details of the K_α region for the five metal targets: ^{208}Pb , ^{232}Th , ^{235}U , ^{238}U , and ^{239}Pu . These spectra were recorded with a large-volume Ge(Li)

Fig. 27.

Schematic diagram of the stopped-muon detector geometry.

Fig. 28.

Muonic x-ray spectrum for ^{208}Pb . The energy scale is about 1 keV/channel.

detector. Therefore, the figures display the characteristic full energy (FE) as well as the single (SE) and double escape (DE) peaks. Since ^{208}Pb is a "doubly magic" spherical nucleus of ground state spin zero, its spectrum is very simple. The other nuclei, because of their nuclear deformation, display appreciable hyperfine splitting. Table II lists the centroid energies for the K_α ($2P_{3/2}$ to $1S_{1/2}$ and $2P_{1/2}$ to $1S_{1/2}$) transitions displayed in Fig. 29. This table shows that various fissionable elements display energy shifts of approximately 90 keV, while the uranium isotopes display an energy shift of nearly 30 keV. Both of these energy shifts are clearly observable with the Ge(Li) detector which possesses about 7-keV resolution.

TABLE II

K_α CENTROID ENERGIES (keV)

Isotope	$2P_{1/2}$ to $1S_{1/2}$	$2P_{3/2}$ to $1S_{1/2}$
^{208}Pb	5779	5963
^{232}Th	6059	6356
^{235}U	6159	6482
^{238}U	6130	6455
^{239}Pu	6244	6595

Fig. 29.
Muonic K x-ray spectra for ^{206}Pb , ^{232}Th , ^{235}U , ^{238}U ,
and ^{239}Pu .

Figure 30 displays the muonic L x-ray spectral region for the same five metal targets: ^{206}Pb , ^{232}Th , ^{235}U , ^{238}U , and ^{239}Pu . The elemental and isotopic shifts are also observed for these x rays. The simple spectrum for ^{206}Pb is again evidence of its value as a calibration standard.

Fig. 30.
Muonic L x-ray spectra for ^{206}Pb , ^{232}Th , ^{235}U , ^{238}U ,
and ^{239}Pu .

The data presented above clearly demonstrate the elemental and isotopic discrimination aspects of this technique. It now remains to develop this technique at the high-intensity LAMPF facility and to extend the technique to meaningful quantitative assays.

REFERENCES

1. A. E. Evans and R. H. Augustson, "Nuclear Analysis Research and Development. Program Status Report, September-December 1972," Los Alamos Scientific Laboratory report LA-5197-PR (1973), pp. 1-2.
2. A. E. Evans, M. M. Thorpe, and J. J. Malanify, "Nuclear Analysis Research and Development. Program Status Report, January-April 1972," Los Alamos Scientific Laboratory report LA-4994-PR (1972), pp. 1-3.
3. R. H. Augustson and E. I. Whitted, "Nuclear Safeguards Research and Development. Program Status Report, May-August 1971," Los Alamos Scientific Laboratory report LA-4794-MS (1971), p. 7.
4. J. H. Menzel and R. H. Augustson, Los Alamos Scientific Laboratory report LA-4994-PR (1972), p. 3.
5. E. B. Fowler, J. L. Warren, K. A. Pashman, J. W. Healy, Los Alamos Scientific Laboratory report LA-5281-MS.
6. J. F. Tinney and J. J. Koch, UCRL-5007-67-3, Hazards Control Progress Report 29, p. 15.
7. E. D. Cashwell et al., Los Alamos Scientific Laboratory report LA-5157-MS.
8. T. D. Reilly and R. B. Walton, Los Alamos Scientific Laboratory report LA-5091-PR (1972), pp. 19-20.
9. T. D. Reilly, R. B. Walton, E. R. Martin, and J. L. Parker, Los Alamos Scientific Laboratory report LA-5197-PR (1972), p. 11.
10. L. V. East, J. H. Menzel, and R. B. Walton, Los Alamos Scientific Laboratory report LA-4070-MS (1968), p. 10.
11. R. B. Walton, Los Alamos Scientific Laboratory report LA-5091-PR (1972), pp. 20-22.
12. Los Alamos Scientific Laboratory report LA-4994-PR (1972), pp. 15-19.
13. E. D. Cashwell et al., Los Alamos Scientific Laboratory report LA-5157-MS (1973).
14. C. M. Vaughn, L. D. Anstine, J. P. Stewart, and J. S. Delap, "Development, Testing, and Utilization of the Elephant Gun for Measurement of Heterogeneous Radioactive Processing Residuals," Fourteenth Annual Meeting of the INMM, June 20-22, 1973, San Diego, California.
15. R. A. Forster, H. O. Menlove, and J. L. Parker, Los Alamos Scientific Laboratory report LA-4883-PR (1971) pp. 7-9.
16. E. D. Cashwell et al., "MCN: A Neutron Monte Carlo Code," Los Alamos Scientific Laboratory report LA-4751 (1972).
17. H. O. Menlove, J. L. Parker, D. B. Smith, and R. A. Forster, Los Alamos Scientific Laboratory report LA-5091-PR (1972), p. 6.
18. D. B. Smith and R. A. Forster, Los Alamos Scientific Laboratory report LA-5197-PR (1973), p. 6.
19. J. M. Cuttler, S. Shalev, and Y. Dagan, Trans. Am. Nucl. Soc. 12 No. 1, 63 (1969).
20. J. Cuttler, Y. Dagan, and S. Shalev, Trans. Am. Nucl. Soc. 14, 417 (1971).
21. S. Shalev and G. Rudstam, Phys. Rev. Lett. 28, 687 (1972).
22. S. Shalev and J. Cuttler, Trans. Am. Nucl. Soc. 14, No. 1, 373 (1971).
23. G. L. Woodruff, "The Measurement of Delayed Neutron Energy Spectra," USAEC report RLO-2225-T17-4 (July 1972).
24. Los Alamos Scientific Laboratory report LA-5197-PR, pp. 26-27 (1972).

PUBLICATIONS

1. D. A. Close, R. C. Barse, J. J. Malanify, and C. J. Umbarger, "X-Ray Intensity Ratios, $L\alpha/L\beta$ and $L\alpha/L\gamma$, Produced by 1.0 to 3.7 MeV Proton Bombardment," Bull. Am. Phys. Soc. 18, 558 (1973).
2. D. A. Close and R. C. Barse, "Average Resonance Spectroscopy in $^{93,95}\text{Tc}$," Nucl. Phys. A201, 337 (1973).
3. R. A. Forster and H. O. Menlove, "Neutron Coincidence Detector for FBR Fuel Pins: Description and Operating Procedures Manual," LA-5156 (1973).