LA-4523-MS

LOS ALAMOS SCIENTIFIC LABORATORY
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Nuclear Safeguards

Research and Development

Program Status Report

May-August, 1970

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

NUCLEAR SAFEGUARDS RESEARCH SERIES

G. Robert Keepin, Editor

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LOS ALAMOS SCIENTIFIC LABORATORY
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Nuclear Safeguards

Research and Development

Pulsed Neutron Research Group, A-1

G. Robert Keepin, Group Leader

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Distributed September 1970

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Nuclear Safeguards Research and Development

NEUTRON INTERROGATION AND NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS

Delayed-Neutron Assay of Fissionable Material in One-Gallon Recovery Cans - Computations

R. A. Forster and D. B. Smith

In the previous progress report,[1] computations were presented which showed the relative delayed-neutron response per gram of ^{239}Pu uniformly distributed in various CH_2 matrices in the active 4π can counter. It was found that normalizing the volume-averaged delayed-neutron response per gram to the fission counter response significantly reduced the effects of the matrix material. Although this normalization to fission chamber response does not completely eliminate matrix effects, it is possible to incorporate the measured fission-chamber response into an iterative technique (described below) which does effectively remove matrix material effects.

Consider Figs. 1 and 2 which show a series of calculated calibration curves for a Pu-CH_2 system (similar standard curves could be obtained experimentally using known mixtures of Pu-CH_2). The computed standard responses designated by the points in the figures were calculated using the DTF-IV regular and adjoint solutions as described previously.[1] Figure 1 shows the total volume-averaged delayed-neutron response vs ^{239}Pu mass for various densities of CH_2 matrices. As expected, the total response increases with increasing ^{239}Pu mass and with higher CH_2 densities because of increased multiplication producing more fission neutrons. Figure 2 depicts the response of the ^{239}Pu fission chamber per gram (chamber located 1.5 cm from the edge of the can) vs density of the CH_2 matrix for various ^{239}Pu masses. The fission chamber response, of course, also increases with increasing ^{239}Pu mass and CH_2 density.

Thus, when the total volume-averaged delayed-neutron response and fission-chamber response have been measured for an unknown sample, the curves in Figs. 1 and 2 (or similar experimental curves) can be used to determine the amount of ^{239}Pu present. To illustrate the iterative process used, first assume there is no matrix material: the mass of ^{239}Pu can then be approximated from Fig. 1 using the $\rho(CH_2) = 0.0$ curve. From this estimated mass of ^{239}Pu, an estimate of the unknown matrix in terms of the standard CH_2 matrices can be obtained from Fig. 2. Returning to Fig. 1 and using the curve whose CH_2 density corresponds to the value from Fig. 2 (this will probably require interpolation), one obtains a new estimate of the mass of ^{239}Pu. This iterative procedure is continued to convergence.

[1] D. B. Smith and R. A. Forster, LA-4457-MS, pp. 3-5 (1970).

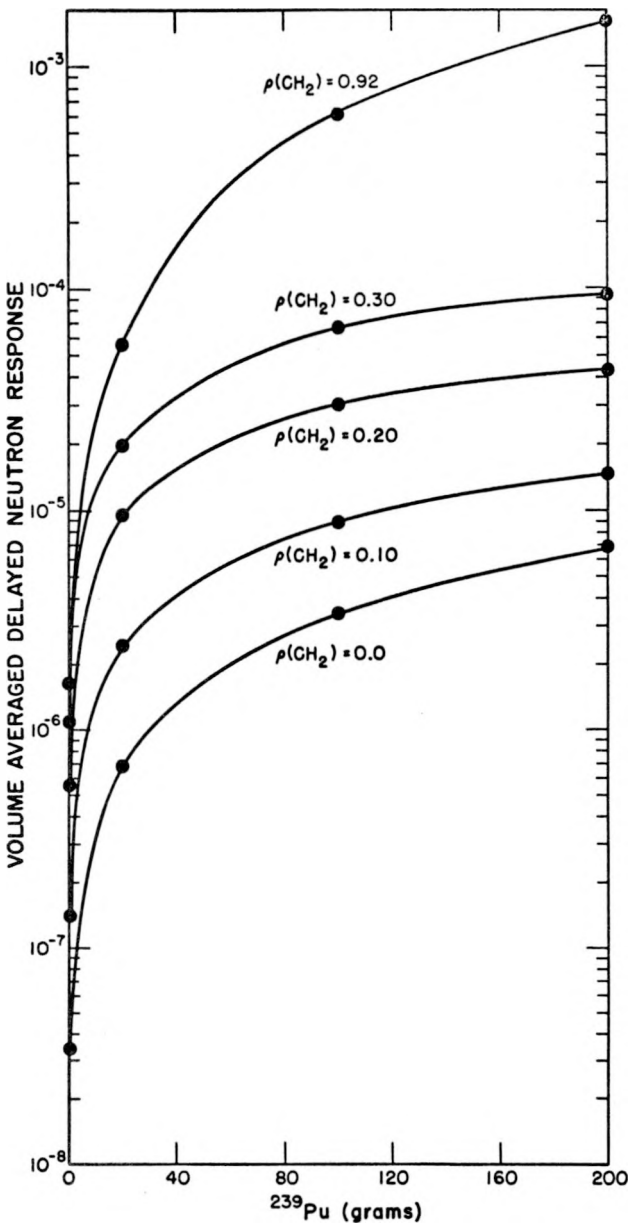

Fig. 1. Calculated total volume-averaged delayed-neutron response of one-gallon cans versus ^{239}Pu mass homogeneously distributed in various densities (g/cc) of CH_2 matrices.

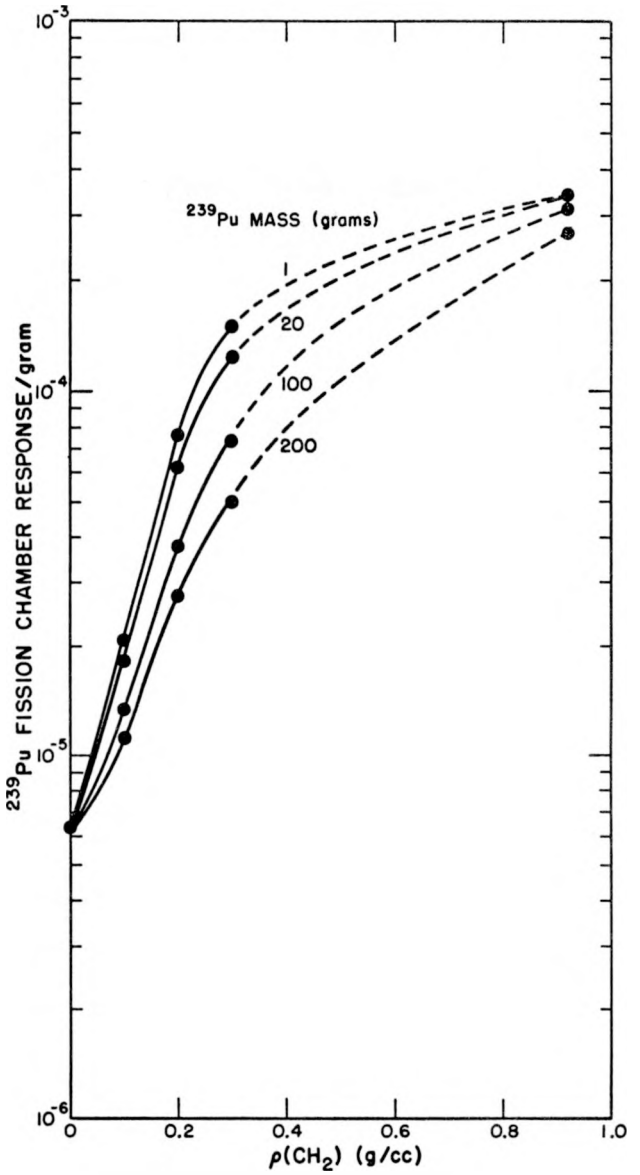

Fig. 2. Calculated ^{239}Pu fission chamber response per gram versus CH_2 matrix density homogeneously mixed with various amounts of ^{239}Pu (denoted by curves parametric in ^{239}Pu mass).

In order to check this method, several "unknown" cans were computationally analyzed which contained arbitrary amounts of ^{239}Pu (up to 200 g) homogeneously mixed with a variety of matrices such as C, CH_2, and Al and Si. The ^{239}Pu loadings predicted by this iterative method agreed to within 10% of the actual value for all cases and to within a few percent for the higher Z matrices. Thus for active assay of one-gallon (and smaller) containers, a measurement of the total volume-averaged delayed-neutron response and the response of an external fission chamber can be used with the iterative procedure described herein to yield quite accurate assay results. Clearly the iterative procedure requires good interpolation between parametric standard curves such as shown in Figures 1 and 2; accurate interpolation can be facilitated by

measuring a greater number of parametric standard curves.

If the unknown can is heterogeneous in character (which, incidentally, is unlikely for fuel in the oxide form), one would expect a larger error in the assay because total delayed-neutron response becomes a function of radius. Figure 3 serves as a guide to this additional error caused by lumping of the ^{239}Pu. The points on the curves are the volume-averaged responses. In general, the more ^{239}Pu and CH_2 matrix present, the more the response varies with radial position and hence the larger the error. For common matrices with $\rho(CH_2) < .1$ g/cc, errors of up to 15% can be expected. If large CH_2 densities such as 0.30 g/cc are encountered, then fuel lumping could cause errors as large as 50%. For non-hydrogenous matrices such as insulation, slag, and ash, errors due to fuel lumping should be of the order of a few percent or less.

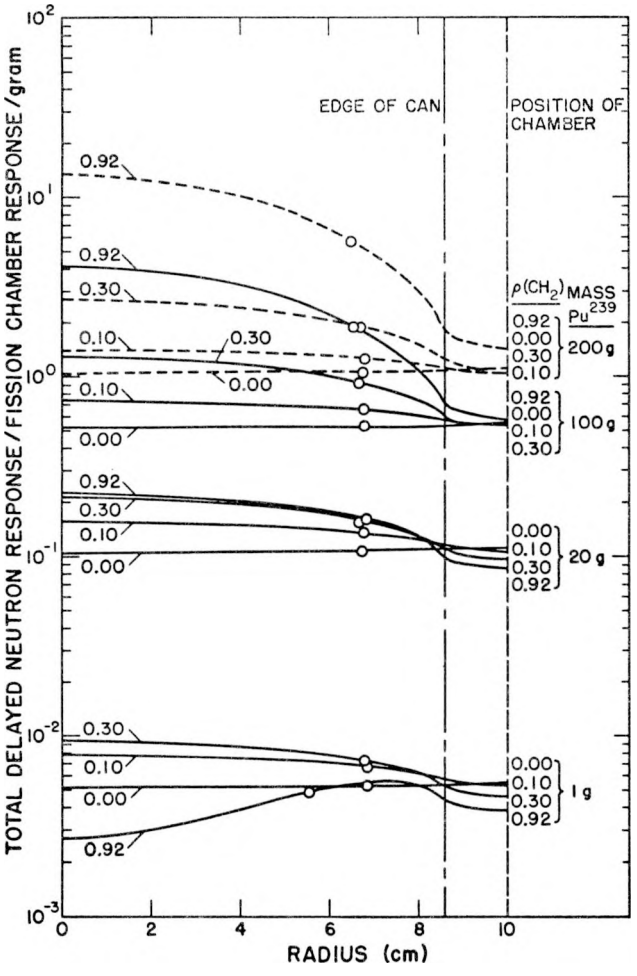

Fig. 3. Calculated total delayed-neutron response per fission-chamber response per gram versus radius in one-gallon cans for various ^{239}Pu fuel loadings homogeneously dispersed in several CH_2 densities (the points are the volume-averaged responses).

Nondestructive Assay of Fissionable Material in One-Gallon Recovery Cans - Experimental

R. H. Augustson and H. O. Menlove

The preceding program-status report [2] described the nondestructive assay of six one-gallon containers of Pu scrap using three assay techniques: passive γ counting, coincidence neutron counting, and two delayed-neutron yield measurements (at two different interrogating energies). Results of the four different measurements agreed within the stated uncertainties of the data. Each of the six cans have now been completely recovered chemically, thereby obtaining independent Pu assay values for each can. Table I presents the comparison between chemical analysis, nondestructive assay, and the "by-difference" Pu values originally assigned to each can. The average % deviation between the chemical results and an average of the nondestructive assay values is 11%.

The extreme case of Sample XBL-45 in Table I is noteworthy in that the nondestructive assay and chemical analysis results agree very well ($\sim$ 5% deviation), but both of these results disagree markedly ($>$ 60% deviation) with the original by-difference mass estimate. The shortcomings of by-difference methods in materials accountability are well known and ubiquitous throughout the nuclear industry; it is indeed gratifying that the essential role of direct physical measurements in

[2] R. H. Augustson and H. O. Menlove, LA-4457-MS, pp. 6-8 and p. 31 (1970).

TABLE I

Pu ASSAY OF ONE-GALLON SCRAP CONTAINERS

Slag & Crucible Can No.	By-Difference Mass Assignment (g Pu)	Chemical Analysis (g Pu)	Nondestructive Assay (Average Value) (g Pu)
XBL 40-1	13	17.2	21.25
XBL 40-2	13	14.0	16.3
XBL 42	11	14.0	13.8
XBL 43	23	20.4	22.5
XBL 44	18	17.2	15.7
XBL 45	24	14.4	15.2

effective materials accountability is becoming more and more widely recognized throughout industry and government.

Delayed-Neutron Assay of ^{235}U Content in a Spent MTR Fuel Element

R. H. Augustson, C. N. Henry (A-2), and C. R. Weisbin

A direct measurement of the amount of ^{235}U contained in a spent MTR-type fuel element has been made using the delayed-neutron yield technique.[3,4] Approximately 35% burnup of the initial ^{235}U content had been achieved during irradiation in the core of the Los Alamos Omega West Reactor. At the time of the measurement, the fission product inventory was estimated to be 12,000 curies. The measurement was performed with the element inside a 5000-lb Pb transfer cask. The delayed neutron result agrees with the burnup calculation to within the stated uncertainties of each.

The MTR element, in the Pb transfer cask, was irradiated with D, T neutrons (suitably spectrum tailored, as described below) produced by a 300-keV Cockcroft-Walton accelerator. The accelerator was modulated at a 50% duty cycle, and the delayed neutrons resulting from fissions induced in the sample during the "accelerator-on" intervals were counted during the "accelerator-off" intervals. For the present measurements, the timing sequence used was a 50-millisecond irradiation followed by a 50-millisecond count interval. Figure 4 shows the physical arrangement of neutron source, sample, and detector as used for assay of the spent MTR fuel element. The tritium target of a Cockcroft-Walton accelerator was enclosed in a 2-in. thick Pb assembly which moderated and multiplied (via (n, 2n) reactions) the 14-MeV D, T neutrons. The fuel element in its Pb cask was positioned next to this Pb assembly (cf Figure 4); thus the 6-in. thick walls of the cask provided additional neutron moderation and multiplication. Figure 5 presents the calculated effective neutron-interrogation spectrum as seen through a total of 8 inches of lead. Mounted on the 2-in. Pb assembly was a ^{235}U fission chamber for monitoring the neutron source intensity. Delayed neutrons were counted by a high efficiency, flat-energy-response detector.[5] Because of non-uniform burnup along the longitudinal axis of the element, the cask plus element was

[3] C. F. Masters, M. M. Thorpe, and D. B. Smith, Nucl. Sci. and Eng., 36, 202 (1969).

[4] Proceedings of the AEC Symposium on Safeguards Research and Development, WASH 1147, p. 110 (1969).

[5] L. V. East and R. B. Walton, Nucl. Instr. and Methods, 72, 161 (1969).

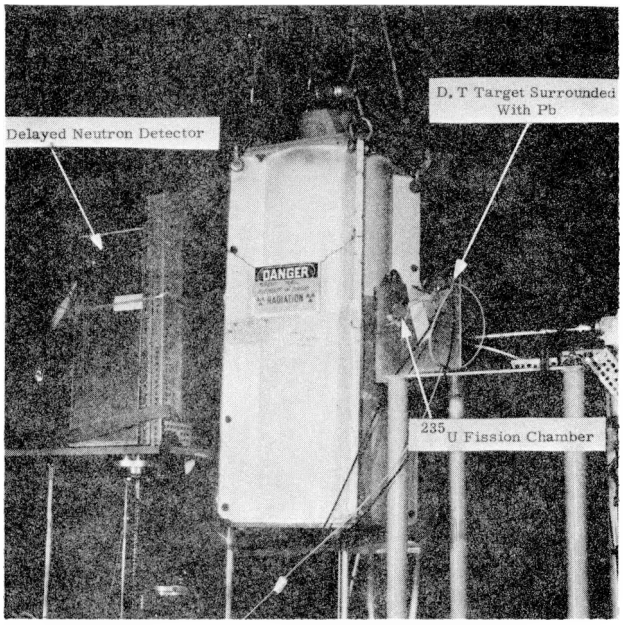

Fig. 4. Experimental arrangement for assay of "spent" MTR fuel elements. The spent element is encased within the large (5,000 lb) Pb cask labeled "DANGER, RADIATION". (This high (35%) burnup element contains ~ 12,000 curies of fission product activity.

scanned past the neutron source-detector arrangement to determine the axial dependence of ^{235}U concentration. A comparison of the scan measurements between a uniformly loaded element and the spent element is shown in Fig. 6.

An unirradiated fuel element containing a known amount of ^{235}U was measured under identical conditions to provide the standard by which the absolute amount of ^{235}U in the spent element could be determined. Delayed-neutron contributions from ^{238}U and ^{236}U were calculated using the one-dimensional DTF-IV code and in the case of ^{238}U, calculations were confirmed by measurement. The discrete ordinates calculations were made in the one-table (isotopic transport), S_8, 25-group, 35-spatial-points, approximation with cross

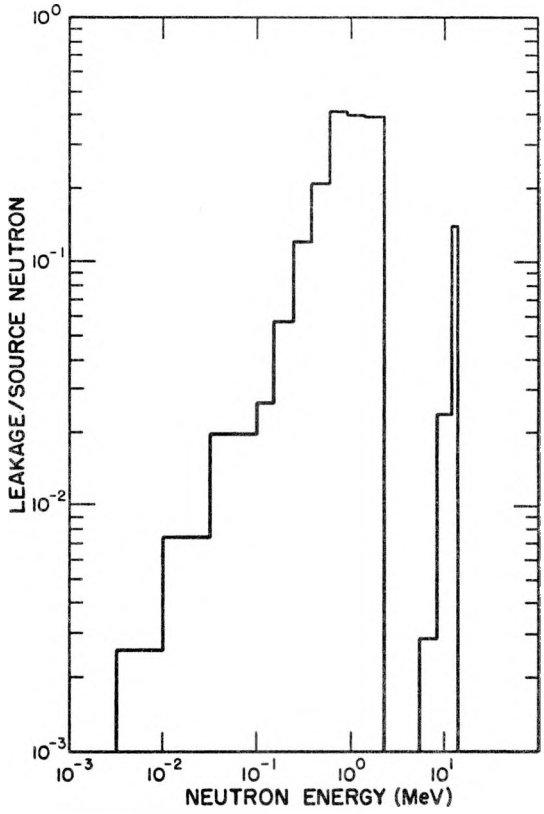

Fig. 5. Neutron-interrogating spectrum of 14 MeV neutrons through 8 in. of lead.

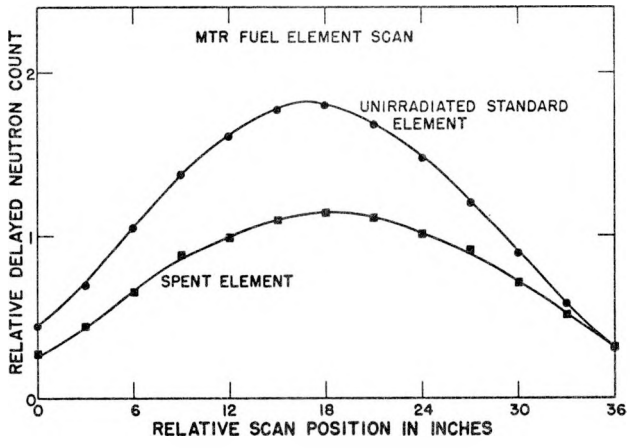

Fig. 6. Comparison of MTR fuel element scans; (uniformly loaded standard element vs. spent element).

sections obtained from the LASL library.[6] The calculated delayed-neutron yield ratio for $^{235}U/^{238}U$ on a per-gram basis was 4.33 compared to a measured ratio of 4.4. For the ^{236}U contribution, the corresponding ratio was 3.3. These contributions lead to an overall correction of 2.5% to be applied to the final ^{235}U result. It should be noted that calculations also predict a linear delayed-neutron response over a wide range of ^{235}U loadings (see Fig. 7). Table II presents the measured ^{235}U content of the fuel element and compares it with the value obtained from the reactor operator's burnup calculation (stated to be accurate to within ± 3%) for this particular element. It can be seen that the agreement is well within the uncertainties of the two values.

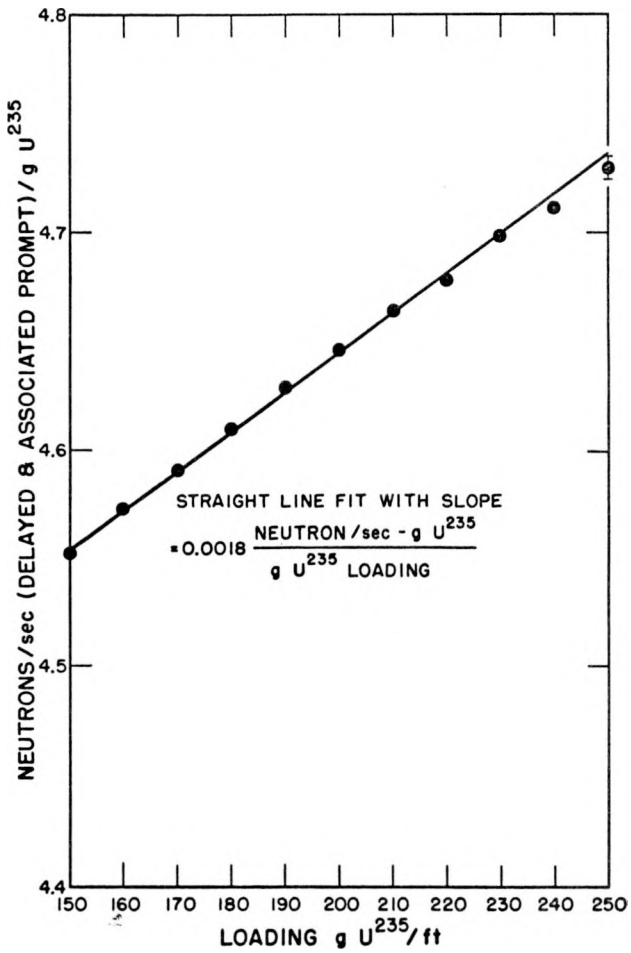

Fig. 7. Delayed neutron leakage/gram vs. loading.

The delayed-neutron yield technique can also be applied to irradiated power reactor fuel elements contained in Pb transfer casks. For these low-enrichment fuels, it is advantageous to enhance the discrimination between fissile and fertile materials by tailoring the source spectrum to produce a larger percentage of neutrons below the ^{238}U fission threshold. This increased discrimination can be obtained using such neutron sources as a Van de Graaff accelerator, radioactive sources (e.g., ^{238}PuLi, SbBe, RaBe), or a suitably moderated D, T source.

TABLE II

DELAYED-NEUTRON ASSAY OF ^{235}U CONTENT IN A HIGH-BURNUP MTR FUEL ELEMENT

Cold Element (Standard, g)	Delayed-Neutron Assay (g)	Burnup Calculation (g)
220	144 ± 2	142 ± 4

Non-Destructive Assay of Power Reactor Fuel Elements

R. H. Augustson and A. E. Evans

Techniques to measure the total ^{235}U content of low-enrichment power reactor fuel elements are under development. The initial approach to this formidable task has been to interrogate with completely subthreshold neutrons produced by the A-1 Van de Graaff accelerator and employ the well-established delayed-neutron yield technique. In the future, interrogation with subthreshold radioactive sources and prompt neutron detection will also be

6 R. Lazarus and L. Stewart, Collections of Cross-Section Evaluations From a Variety of Sources Including LRL, KAPL, UK and LASL, updated 1968.

examined as an alternative method of assaying power reactor fuel elements.

The experimental measurements to date have been made using a simulated fuel element patterned after the Dresden 1 BWR design. As shown in Fig. 8, the element consists of a 6x6 array of 5/8" diameter rods with each rod removable. Two 28" long segments have been fabricated and can be joined together to form a standard 56" fuel element. The rods are presently loaded with natural uranium oxide (.7% ^{235}U) powder packed to a density of 3.1 g/cc. In the future, it is planned to introduce enriched U pellets with densities of 10 g/cc. The experimental arrangement, shown in Figure 9, is similar to that used in previous delayed-neutron assay work.[7] The interrogating neutrons, produced by the Li(p, n) reaction using a thick target, have energies ranging from 100 to 400 keV. At these energies the large quantity of ^{238}U present in the element produces less than 0.05% of the fissions from interactions with the primary neutron beam.

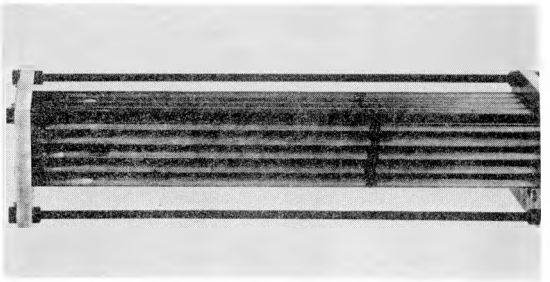

Fig. 8. Simulated BWR fuel element with removable fuel rods.

In the quantitative assay of a sample as dense, as large, and as geometrically complex as a complete power reactor element, it is important to flatten the spatial response across the width of the element; ideally, every rod should contribute equally to the observed delayed-neutron response independent of rod location in the element. The contribution of a given rod to delayed-neutron res-

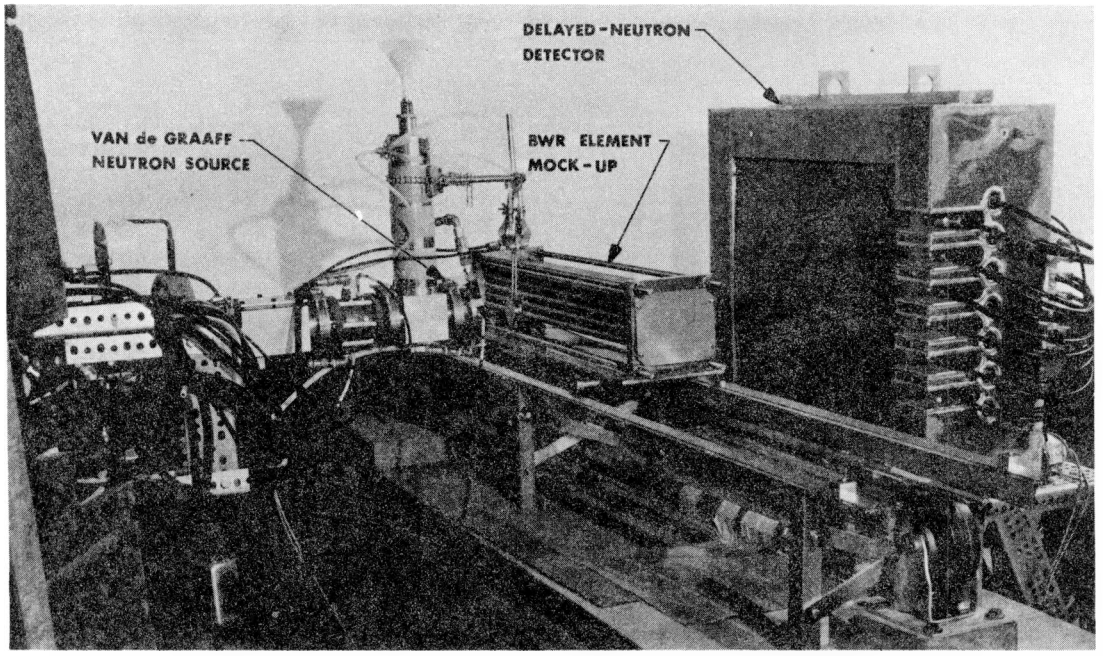

Fig. 9. Experimental arrangement for assay studies of low enrichment BWR power reactor fuel elements.

[7] R. H. Augustson, H. O. Menlove, R. B. Walton, L. V. East, A. E. Evans, and M. S. Krick, Paper #SM 133/51, IAEA Symposium on Progress in Safeguards Techniques, July 6-10, 1970, Karlsruhe, Germany.

ponse at the detector is determined by the product of at least three factors:

1. The first factor is the spatial dependence of the incident neutron flux from the interrogating source as modified by the presence of the other rods in the element and any other material present. Some fraction of the incident neutrons are scattered as they pass through the element, changing their direction and energy with each collision. In the present case of collisions with UO_2 the energy is not changed appreciably. Measurements with a ^{235}U fission chamber are underway to study this effect.
2. Prompt neutrons resulting from the fission of ^{235}U can cause additional fissions in the large quantity of ^{238}U present, leading to subsequent delayed-neutron emission. Using a fission spectrum averaged cross section of 0.3 barns, it is estimated that this effect can be the order of 5% of the delayed-neutron rate. The position dependence of prompt neutron multiplication is being studied calculationally.
3. The efficiency for counting delayed neutrons emitted from an individual rod is also position-dependent. The detection efficiency falls off with distance from the detector due to solid angle considerations. In addition, the delayed neutrons are scattered as they pass through the fuel element, some being scattered into, and others out of, the detector view. Figure 10 presents measurements taken with a PuLi source simulating delayed neutrons. Rows of fuel rods were added between the neutron source and the detector. The counting rate decreased as more rows were added indicating outscattering of the delayed neutrons. The experiment was then repeated, adding rows of rods behind the source. In this configuration the counting rate increased by $\leq 10\%$. For a given rod in the 6x6 BWR type array, both of these effects will contribute to the net delayed-neutron detection efficiency for that rod.

The total effect of the three factors just described can be measured directly without differentiating between each individual process. This was done by loading one rod with 93% enriched ^{235}U and measuring the delayed-neutron response as a function of position of this rod within the element array. The delayed-neutron response of the other 35 rods was measured and subtracted, thereby isolating the response of a single rod in the presence of the other rods. Following this procedure, a source-sample-detector arrangement was determined which had an essentially "flat" response

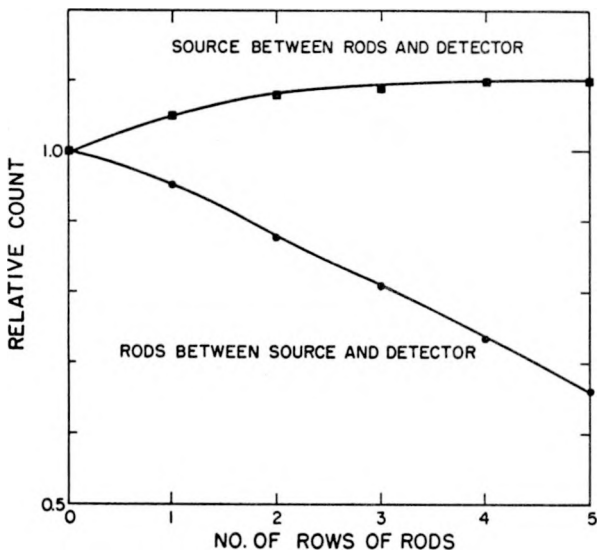

Fig. 10. Delayed neutron response studies in BWR-type fuel element assays.

across the width of the element. As shown in Fig. 11, the element is positioned 18" from the Van de Graaff target and an equal distance from the detector, with two inches of steel surrounding the element. Also shown in Fig. 11 are the measured response-vs-position dependences. The response is seen to be uniform to within 1% along the horizontal plane and to within 10% along the vertical plane.

The effect of varying the total amount of uranium in the element was studied using the "flat response" configuration just described. First the delayed-neutron response of the fully loaded element (36 rods of natural uranium) was measured, followed by measurements taken with certain rods removed. Figure 12 shows a comparison of these assay measurements with the "true" values obtained by weighing the rods. The comparison shows a systematic deviation of assayed value from weighed value--the assayed value being systematically low. This may be attributable to the "response enhancement" effect of prompt neutron multiplication. Further studies are in progress to investigate this deviation. A practical alternative to making theoretical corrections for the deviation is to use the data given in Fig. 12 as a calibration curve for direct fuel element assay. This curve is linear to within 1% over a 20% change in fuel element loading. Thus practical assay using the calibrated-response-curve approach is quite feasible since the present experimental sensitivity is such that a 2% change in mass can be easily detected.

Civilian power reactor fuel elements are typically eight to ten feet long, and in the course of routine nondestructive assay would be scanned along their length. Such scanning measurements with the present simulated fuel element segments indicate no particular difficulties with an axial scanning procedure.

The results obtained thus far in the LASL program to develop nondestructive assay methods for power reactor fuel elements are indeed encouraging. Systematic studies of the detailed effects of material density, isotopic enrichment, geometrical distribution, self shielding, etc., are underway or planned using both delayed and prompt neutron signatures.

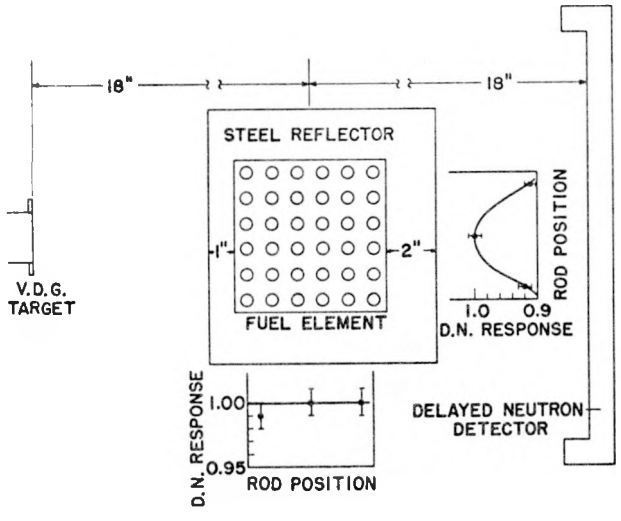

Fig. 11. Delayed neutron assay of BWR fuel element. (Spatial response dependence in flat-geometry configuration.)

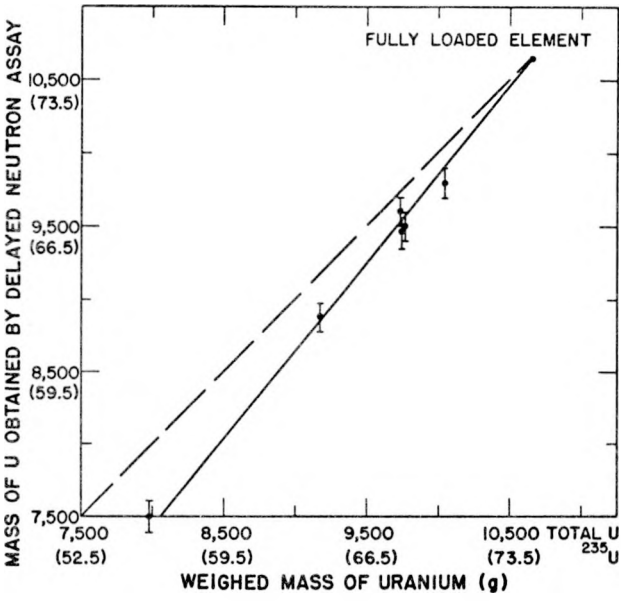

Fig. 12. Delayed neutron assay of BWR fuel element. (Assayed vs. weighed mass values.)

Criticality Calculations--4π Active Can Counter

D. B. Smith

In preparation for assay in the 4π active can counter[8] of ~ 100 cans (from Idaho Nuclear Corporation) each containing up to 4 kg of ^{235}U (93%) in an aluminum matrix, a series of calculations was carried out on the multiplication factor, k, for various amounts of uranium in the counter. As in the series described previously for cans containing plutonium,[9] the discrete-ordinates transport code, DTF-IV, and Hansen-Roach[10] 16-group cross sections were used. The can (6 3/16 in. diam by 8 3/4 in. high cylinder) was modelled by a sphere having the same volume. This was followed by spherical shells of void, Fe, Cd, B_4C, and carbon of the thickness contained in the actual counter. The spherical "can" contained varying masses of ^{235}U and sufficient Al to fill it. The resulting curve of multiplication factor, k, vs ^{235}U mass (Fig. 13) indicates that there is no possibility of criticality if the cans contain nothing but uranium and aluminum. To investigate the consequences of an hydrogenous matrix material (e. g. water, inadvertently introduced into the can), one case was run for 4 kg ^{235}U plus sufficient CH_2 to fill the can. This configuration produced a multiplication factor, k = 0. 9--still a non-critical situation.

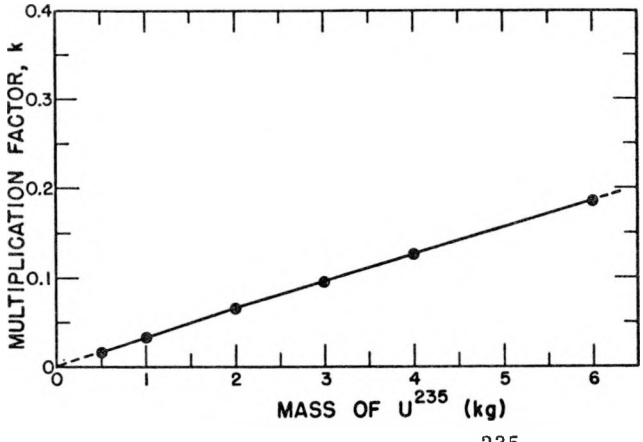

Fig. 13. Multiplication factor, k, vs ^{235}U mass; 4π active can counter.

Curves presented in Reference 11 indicate that for water-moderated, water-reflected ^{235}U (93%) systems, approximately 12 kg of ^{235}U would be required to reach criticality in a container of the size (~ 4. 3 ℓ) under consideration. Thus, there should be no possibliity of criticality with the amounts of ^{235}U and the matrix material expected.

The results of these calculations will be used to establish safe operating procedures for assaying the cans of uranium aluminide from Idaho Nuclear Corporation.

Delayed-Neutron Response of Small Amounts of Fissionable Material

D. B. Smith

The response of a detector to delayed neutrons produced by an external neutron source incident on a system containing a small amount of fissionable material is of interest in nuclear safeguards and fissionable scrap assay. The calculation of the detector response may be complicated by the necessity of including delayed neutrons, time dependence, and multi-dimensional geometry. A two-stage calculational technique has been devised for solving such problems utilizing the Monte Carlo method.

A direct simulation of the neutron transport is computationally inefficient since the source neutrons seldom enter the small volume of fissionable material and cause fission. This difficulty is circumvented by solving the problem in two stages. In the first stage, a source of first-generation fission neutrons is obtained by sampling from the adjoint transport equation using the adjoint Monte

[8] R. H. Augustson and H. O. Menlove, LA-4457-MS, pp. 6-8 (1970).

[9] D. B. Smith and R. A. Forster, LA-4457-MS, pp. 8-11 (1970).

[10] G. E. Hansen and W. H. Roach, LAMS-2543 (1961).

[11] H. C. Paxton, J. T. Thomas, A. D. Callihan, and E. B. Johnson, TID-7028, pp. 14-15 (1964).

Carlo code, MCNA.[12] The second stage of the calculation uses this first-generation neutron source as a starting point for a direct simulation of all subsequent neutron generations using the delayed-neutron Monte Carlo code, DDMC. The two codes are coupled through a special source subroutine which has been written for the delayed neutron code.

The advantage obtained by sampling from the adjoint transport equation in the first stage of the calculation is that the "pseudo-neutrons" begin their life histories in the small volume of fissionable material.

As a test problem, the delayed neutron response of a small sphere of ^{235}U surrounded by polyethylene (3 in. thick) to a 14 MeV pulsed neutron source at a distance of 30 cm was calculated. A direct simulation of the neutron transport in such a system would very likely require an hour or more of computer time to produce a useful result. The two-stage technique required approximately 15 min to produce a result with a statistical error of less than 10%.

RADIOACTIVE NEUTRON SOURCES FOR NONDESTRUCTIVE ASSAY APPLICATIONS

Neutron Source Moderator-Shield Assemblies

R. A. Forster and H. O. Menlove

Significant progress has been made in LASL's continuing program of calculations and measurements to investigate optimum moderator configurations for use with ^{252}Cf and other radioactive neutron sources. Calculations have been performed using the DTF-IV code[13] for the following moderator assemblies:

1) $Be/C/CH_2$-8.7/8.0/6.0-cm,[14]
2) $CH_2/C/CH_2$-8.7/8.0/6.0-cm,
3) $ZrH_2/ZrH_2/CH_2$-8.7/8.0/6.0-cm.

The choice of moderator materials and geometries was partially determined by fabrication considerations. All of the moderators were surrounded by a 10-cm thick $B_4C + CH_2$ shield.

The results of these computations are presented in Fig. 14 which shows the sub-cadmium

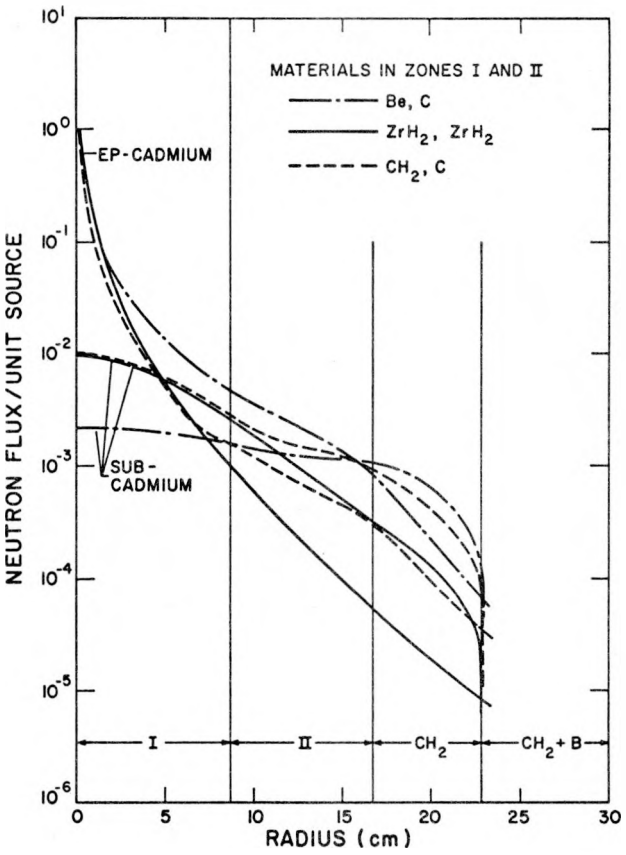

Fig. 14. Calculated neutron fluxes produced by a ^{252}Cf source in a moderating assembly with various materials in the first two regions.

12 Lee Carter, LASL, private communication (1970).

13 The Goad-Sandmeier 25-group cross-section set was used for the calculations.

14 Notation corresponds to a sphere of Be (8.7-cm radius) surrounded by a shell of C (8.0-cm thick) which is enclosed in a 6.0-cm thick shell of CH_2.

neutron fluxes vs radii of the moderating assemblies. Both ZrH_2 and CH_2 assemblies yield large sub-cadmium neutron fluxes at the center as expected. The relatively steep flux gradient would require that a sample be rotated to insure uniform irradiation. In Region II, however, the sub-cadmium neutron fluxes in the CH_2-C are larger and flatter than in the ZrH_2 because of the small absorption in the carbon. The Be-C assembly yields a relatively flat sub-cadmium flux in Region II whose magnitude is roughly the same as the other two cases. The only apparent advantage of the ZrH_2 system over a CH_2 (or H_2O) assembly is the added gamma shielding arising from the reasonably heavy Zr atom (atomic wt = 91.2). This gamma shielding could be important if a gamma-sensitive detector such as a liquid scintillator were used for neutron counting.

Using these calculations as guidance, the moderating assembly shown in Figs. 15 and 16 was fabricated for the experimental investigations. A schematic view of the moderator interior is shown in Fig. 17. The moderator contains two interior sample cavities as well as a beam port for extracting an exterior neutron beam. The $B_4C + CH_2$ in the outer region of the assembly acts as a neutron shield for personnel and detectors. The measured dose rate at 1 m from the shield for a ^{252}Cf source with a yield of $\sim 1.5 \times 10^9$ n/sec was 15 mr/h for neutrons and 20 mr/h for γ rays. The inner portion of the moderator was built with modular construction to permit interchanging of materials to give an interrogation spectrum that is appropriate for a given assay problem. For example, the configuration shown in Fig. 17 provides a high thermal neutron flux in the vicinity of the sample port in

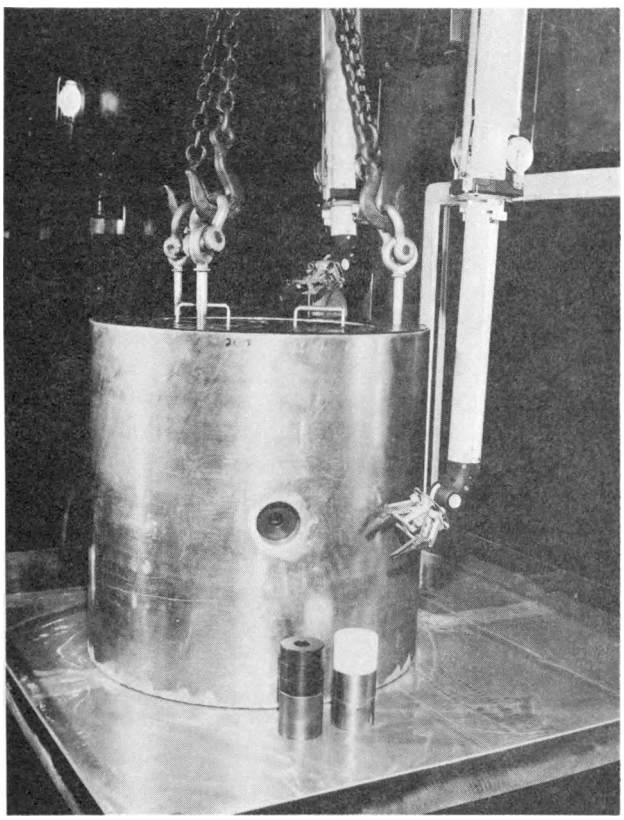

Fig. 15. Exterior view of neutron source moderating assembly with neutron beam port and collimator inserts. The remote control manipulators are part of the hot cell which facilitates the handling of high intensity sources.

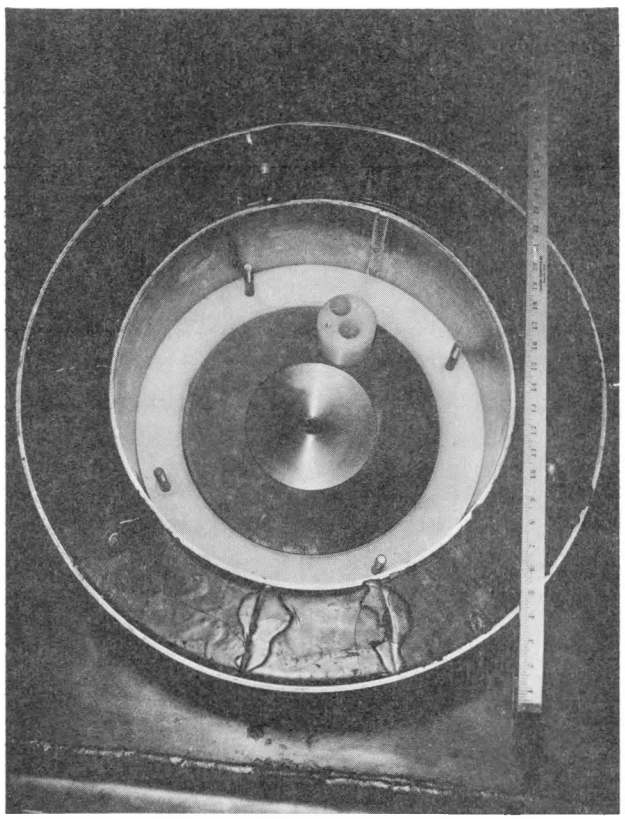

Fig. 16. Interior view of moderating assembly showing the central hole which normally contains the ^{252}Cf or (γ, n) neutron source and the sample-detector part at the intermediate radial position.

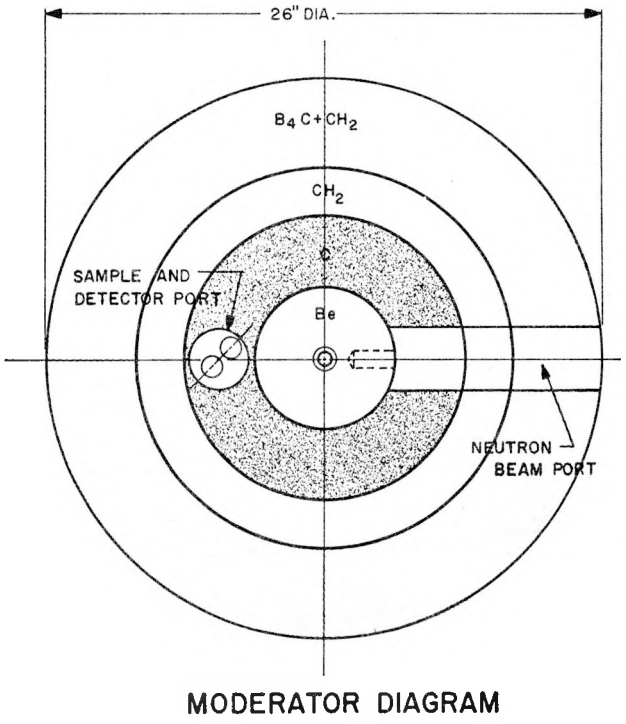

Fig. 17. Schematic diagram, interior of neutron source interrogation assembly. The modular interior rings can be replaced by D_2O, ZrH_2, or Pb materials if needed for a given assay application.

addition to a large number of epithermal neutrons in the beam port for self-indication[15] measurements. If a fast neutron irradiation is needed, the C and CH_2 regions can be replaced by Pb.

Self-Indication Measurements Using ^{252}Cf

H. O. Menlove and H. A. Walter

For self-indication measurements, it is desirable to extract from the moderating assembly a beam which has a large number of neutrons in the fission resonance energy region. The neutron beam port shown in Fig. 17 was designed for this purpose. When the neutron collimators (1-in. i.d.) shown at the bottom of Fig. 15 were inserted into the beam port, the neutron beam profile, measured at 2 in. beyond the shield wall, has a width (FWHM) of 1.3 inches. For assay applications, the size of the collimator can be chosen to fit the dimensions of the fissionable sample.

For the present measurements, a 3-mil thick Gd foil was placed over the beam hole to remove thermal neutrons, and ^{239}Pu disks with thicknesses ranging from 10-80 mil were placed in the neutron beam. The neutron beam then passed through fission chambers containing thin back-to-back foils of ^{239}Pu and ^{233}U. Figure 18 shows a curve of the measured $^{233}U/^{239}Pu$ fission ratio as a function of Pu sample thickness. The fission response variation is essentially the same as had been measured previously[15] in the neutron beam from a reactor. The counting rate in the present fission detector for the 1-mg ^{252}Cf source was ~ 3 cps, and this could be increased by about an order of magnitude by using a more efficient fission chamber.

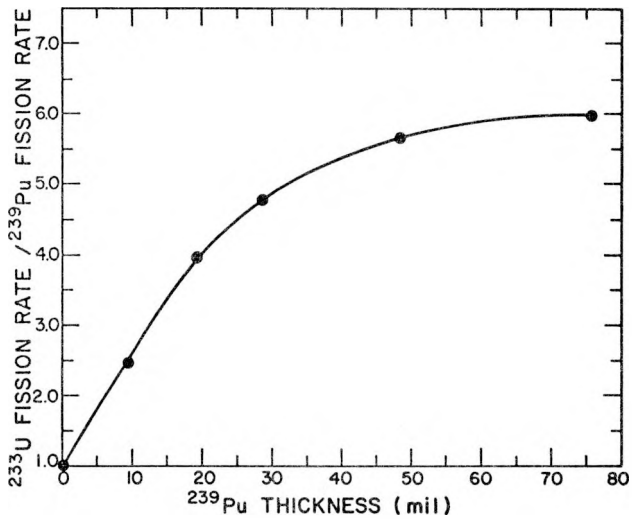

Fig. 18. The fission detector ratio $^{233}U/^{239}Pu$ vs ^{239}Pu sample thickness using a moderated ^{252}Cf neutron beam for the interrogation.

[15] H. O. Menlove, C. D. Tesche, M. M. Thorpe, and R. B. Walton, Nuclear Applications 6, 401 (1969).

Assay of Hot UHTREX Fuel Rods Using ^{252}Cf

H. O. Menlove, J. L. Parker, and H. A. Walter

The sample and detector port shown in Fig. 17 has been used together with a 1-mg ^{252}Cf neutron source to assay the fissile content of several UHTREX (Ultra High Temperature Reactor Experiment) reactor fuel rods. These rods are 6 in. long with an outside diameter of about 1 in. The rods are made of enriched uranium (93% ^{235}U) beads coated with graphite. The rods varied in mass from ~6-14 g U and four of the rods had been irradiated in the UHTREX reactor for 107 MWD giving a measured dose rate of ~ 1000 r/h at contact when the assay measurements were made.

Source neutrons for the interrogation were obtained from the ^{252}Cf source located in the center of the assembly shown in Fig. 17, and the fuel rods were passed through the hole in the CH_2 sample port. A small ^{238}U spiral fission chamber was placed in the detector hole (see Fig. 17) to count the prompt neutrons from induced fissions in the fuel rod.

When using a ^{252}Cf source emitting fission spectrum neutrons for the interrogation of an assay sample, it is necessary to reduce the source neutron spectrum to energies below the neutron bias level of the prompt neutron detector. For the present measurements the neutron detector was a ^{238}U fission chamber with its natural threshold bias level at ~ 1 MeV. The Be and CH_2 in the assembly in Fig. 17 reduced the ^{252}Cf energy spectrum such that the sample-in/sample-out ratio in the fission chamber was ~ 2.3 for the UHTREX fuel rods.

For the measurement of hot fuel elements, it is necessary to use a neutron detector that is insensitive to the high gamma-radiation levels, or to shield the detector with a substantial amount of heavy material. Ionization fission chambers are very insensitive to gamma radiation, and it proved possible to place the ^{238}U fission chamber next to the hot fuel elements without any increase in the background rate.

The results of the prompt-neutron assay of hot UHTREX rods are given in Table III. In order to obtain a calibration curve, three cold fuel rods with known uranium loadings were scanned through the interrogation assembly, and the calibration curve is shown in Fig. 19. The non-linearity in

TABLE III

ACTIVE ASSAY OF HOT UHTREX FUEL RODS USING A ^{252}Cf NEUTRON SOURCE

UHTREX Rod No.	Nominal Loading* (gU)	Prompt-Neutron Assay (gU)
603	6.6	6.55
4238	7.6	7.59
4523	8.9	8.75
5115	10.5	10.40

* The nominal loadings were measured during fabrication using a γ-ray scan.

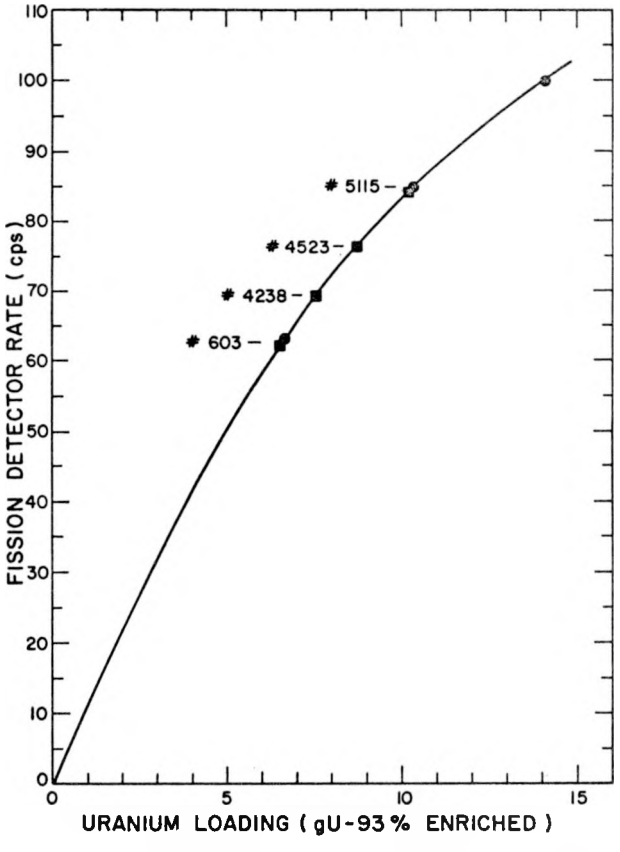

Fig. 19. Calibration curve showing detector-response vs uranium loading for the assay of UHTREX fuel rods. The serial numbers identify the unknown hot fuel rods.

the calibration curve is a result of thermal neutron absorption in the fuel material.

The assay results in Table III are seen to agree very well with the nominal loadings supplied by the fuel fabricator. The small amount of fuel burnup (~ 1-2%) in the reactor could account for the low bias (~ 1%) in the present measurements. It should be noted that if the fuel had been fully "spent" then the ^{235}U burnup would be ~ 30%, and the build-up of fission product poisons would make assay by thermal or resonance region neutron interrogation very difficult to interpret.

DELAYED-NEUTRON YIELD VS ENERGY MEASUREMENTS

M. S. Krick and A. E. Evans

During this reporting period, measurements of total delayed-neutron yields as a function of the energy of the neutron inducing fission have been extended to neutron energies over the range 4 to 7 MeV for ^{238}U, ^{235}U, and ^{233}U.

The experimental technique was as outlined previously.[16] The higher energy neutrons from the D, D reaction were produced by a 25-μamp deuteron beam from the LASL 3-MeV Van de Graaff accelerator impinging on a 1 mg/cm^2 TiD target. Because of the lesser durability of the (water-cooled) deuterium targets, beam current had to be lowered from that used with the $Li^7(p,n)Be^7$ reaction for this experiment, and a greater energy spread (~ 250 keV) and somewhat poorer counting statistics were accepted. A further limitation in statistics occurred because accelerator-produced backgrounds between beam pulses were roughly equal to the delayed-neutron signal from the samples, whereas this signal-to-background ratio was approximately 10:1 for the $Li^7(p,n)Be^7$ neutrons.

Originally, the accelerator-produced neutron background in the experimental area had been approximately fifty times greater during the acceleration of deuterons than when protons were accelerated. As part of a program to improve shielding of the experimental area, a two-ft thick concrete wall between the accelerator and the target area was increased in height from 10 to 19 ft. This reduced the accelerator-produced background in the Van de Graaff target area by a factor of 20, making the delayed-neutron yield measurements with $D(d,n)He^3$ neutrons feasible.

The measured yield-vs-energy data are presented in Figs. 20, 21, and 22, together with prior yield data at other energies.[16, 17] It has previously been shown that for all fissionable isotopes studied, the delayed-neutron yield from fission is independent (in the range from thermal to 1.8 MeV) of the energy of the neutron causing fission,[16] but that the delayed-neutron yield for 14.9-MeV neutrons is from 38 to 48% less than the yield for 3.1-MeV neutrons.[17] It is now established that, for the three isotopes measured, the delayed-neutron yield is independent of energy

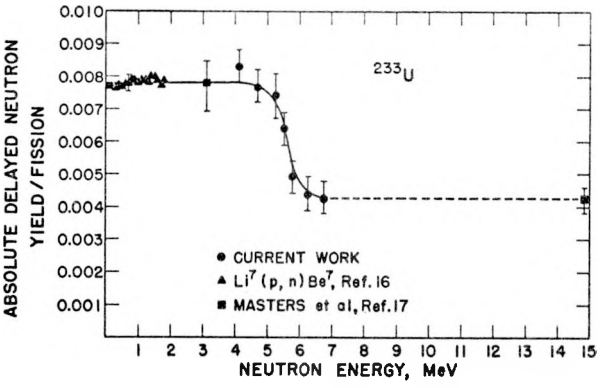

Fig. 20. Absolute total delayed-neutron yield from neutron induced fission of ^{233}U.

[16] M. S. Krick and A. E. Evans, LA-4368-MS, pp. 11-12 (1969).

[17] C. F. Masters, M. M. Thorpe, and D. B. Smith, Nuclear Sci. and Eng. 36, 202 (1969).

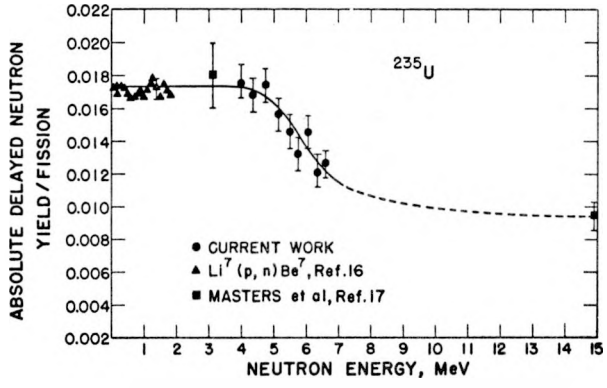

Fig. 21. Absolute total delayed-neutron yield from neutron induced fission of ^{235}U.

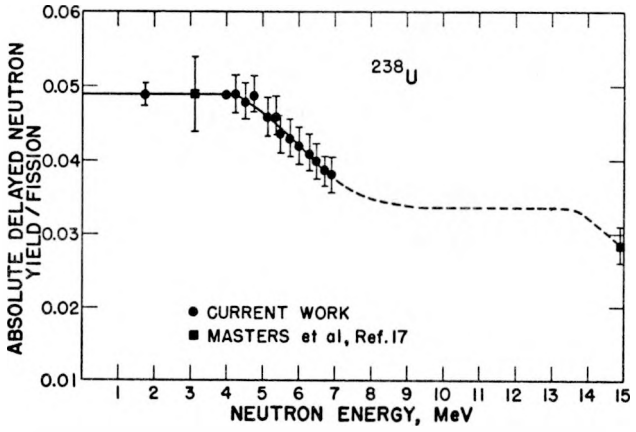

Fig. 22. Absolute total delayed-neutron yield from neutron induced fission of ^{238}U; the indicated extrapolation of the curve to 14 MeV takes into account a second inelastic scattering threshold level associated with the step rise in the ^{238}U fission cross section around 14 MeV.

from 0 to 4 or 5 MeV; the yield then decreases rapidly over a ~ 2 MeV interval to near the yield value at 14.9 MeV (see Figs. 20, 21, and 22). This yield-vs-energy behavior strongly suggests that the energy dependence of the delayed-neutron yield is associated with the threshold for fission after inelastic neutron scattering. Since the fissioning nucleus has in this case one less neutron to distribute among fission products, it is reasonable to expect fewer neutron-rich precursors of delayed neutrons than in the case of conventional fission induced by the capture of a neutron. These results are generally consistent with our understanding of the delayed-neutron emission process and with known systematics of total delayed-neutron yields.[18]

PASSIVE NEUTRON AND GAMMA-RAY ASSAY OF PROTOTYPE LMFBR FUELS

L. V. East and J. O. Barner (CMB-11)

Under the AEC's LMFBR fuels program at LASL, prototype fuel capsules are irradiated in the EBR-II reactor. The fuel in these capsules is in the form of $(U_{0.8}Pu_{0.2})C$ pellets, approximately 0.25 in. diam and 0.25 in. long. Fuel loadings usually consist of about 100 g U 93% enriched in ^{235}U and 28 g Pu (approximately 94% ^{239}Pu). In some high-heat-flux experiments, the ^{235}U is replaced with ^{233}U. Fuel loadings are then about 38 g ^{233}U and 10 g Pu. Five such prototype fuel capsules were gamma-scanned and neutron counted at the Nuclear Safeguards Research Laboratory, LASL. Two of these capsules, Nos. K-45 and K-46, contained ^{235}U and ^{239}Pu; the other three, Nos. K-49, K-50, and K-51, contained ^{233}U and ^{239}Pu.

Each fuel capsule was gamma-scanned on a pellet-by-pellet basis using a 30-cm^3 Ge(Li) spectrometer system in order to determine if the capsules were loaded with the proper types of pellets, i.e., ^{235}U or ^{233}U. Separated ^{233}U normally has associated with it a small amount of ^{232}U, which decays with a half-life of 72 years to ^{228}Th. The decay chain associated with ^{228}Th gives rise to many relatively intense γ rays, including several from ^{208}Tl decay. During the scans, these gamma rays were looked for in order to determine the presence of ^{233}U. The presence of three weak gamma-ray lines associated with the decay of ^{233}U were also monitored (see spectrum shown in Fig. 23). The presence or absence of ^{235}U was

[18] G. R. Keepin, Physics of Nuclear Kinetics, p. 101, Addison, Wesley, Reading, Mass. (1965).

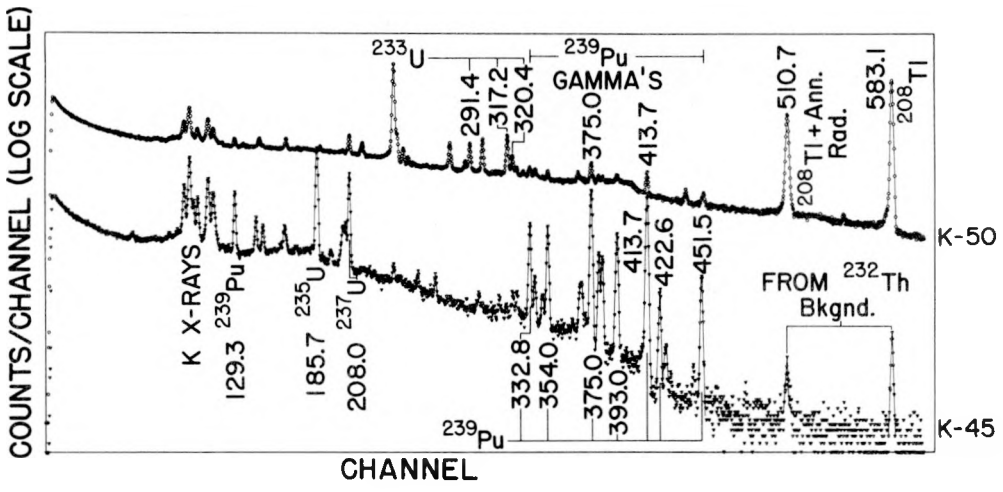

Fig. 23. Gamma-ray spectra from $^{233}U + ^{239}Pu$ (upper curve) and $^{235}U + ^{239}Pu$ (lower curve) fuel capsules. Gamma-ray energies are given in keV. Counting time for each spectrum was 1000 sec.

determined by monitoring the 186-keV gamma ray associated with ^{235}U alpha decay. Two characteristic lines from ^{239}Pu at 375 keV and 414 KeV were monitored for the Pu content.

The fuel pellets in Capsules K-49, K-50, and K-51 were all found to contain ^{233}U and ^{239}Pu in approximately the proper ratio; counting statistics were such that the variation in loading from pellet to pellet could not be determined to better than ~20%. It was definitely determined that there were no total substitutions of ^{235}U or ^{238}U for ^{233}U in any pellet; however, a partial substitution by as much as ~25% in a single pellet would probably not have been detected.

For Capsules K-45 and K-46, the absence of any detectable γ rays above background from ^{232}U allows a very conservative upper limit of 100 mg for the total amount of ^{233}U contained in these capsules. Since there was much less background from extraneous gamma rays associated with these capsules, the pellet-to-pellet variation in ^{235}U and ^{239}Pu could be determined to about 10% for counting times of 1 to 2 min per pellet.

A total gamma-count was also obtained for each capsule in order to determine the accuracy that could be obtained for the relative U and Pu content between capsules. It should be noted that such measurements require the assumption that the U and Pu are uniformly distributed in each pellet. In order to obtain the absolute content by this method, a standard capsule containing a precisely known amount of material would be required. The results of these measurements are presented in Table IV. Each capsule was counted for 17 min.

TABLE IV

RELATIVE LOADINGS OF PROTOTYPE FUEL CAPSULES DETERMINED FROM GAMMA-RAY & NEUTRON COUNTING

Capsule/Reference	Relative Loading			
	Gamma-Ray Counting			Neutron Counting
	^{233}U	^{235}U	^{239}Pu	^{240}Pu
K-45/K-50	-	-	2.84 ± 0.15	2.492 ± 0.014
K-46/K-50	-	-	2.78 ± 0.15	2.454 ± 0.014
K-49/K-50	1.024 ± 0.038	-	1.147 ± 0.067	1.036 ± 0.007
K-51/K-50	1.015 ± 0.055	-	1.118 ± 0.057	1.037 ± 0.007
K-45/K-46	-	1.012 ± 0.011	1.022 ± 0.010	1.015 ± 0.005

For Capsules K-49, K-50, and K-51, the relative ^{233}U and ^{239}Pu contents were determined to ~ 5% accuracy in this counting time. The ^{235}U and ^{239}Pu relative loadings of K-45 and K-46 were determined to ~ 1% accuracy.

The capsules were also neutron-counted using a 4π neutron coincidence counter.[19] This allowed the ^{240}Pu content of each capsule to be absolutely determined with an accuracy of about 2%. The relative ^{240}Pu content could be determined to better than 1%. Neutron-counting results are shown in Tables IV and V. From neutron counting, it would appear that Capsule K-50 contained about 0.5 g less Pu than Capsules K-49 and K-51. Both the neutron and gamma-ray counting results shown in Table IV indicate that the Pu loading of K-46 is less than K-45 by 1.5 to 2%, or about 0.5 g also.

This work has shown that gamma-ray scanning of mixed U-Pu-carbide fuel capsules using a Ge(Li) spectrometer can very quickly determine if any ^{233}U has been improperly placed in a ^{235}U capsule; however the determination of the presence of ^{235}U in a ^{233}U capsule is greatly hampered by the background of gamma rays from ^{232}U decay products. Neutron coincidence counting can be used to determine the Pu loading provided that the ^{240}Pu abundance is known for the Pu used in the fuel. It should be noted that neutron counting is much less sensitive to fuel nonuniformities and the presence of extraneous materials than is gamma counting.

TABLE V

TOTAL Pu CONTENT OF PROTOTYPE FUEL CAPSULES OBTAINED FROM NEUTRON COINCIDENCE COUNTING

Capsule No.	^{240}Pu (g)	Total Pu * (g)
K-45	1.56 ± 0.02	28.9 ± 0.7
K-46	1.54 ± 0.02	28.5 ± 0.7
K-49	0.629 ± 0.009	11.6 ± 0.3
K-50	0.607 ± 0.009	11.2 ± 0.3
K-51	0.630 ± 0.009	11.7 ± 0.3

* Assuming (5.40 ± 0.05)% ^{240}Pu

DETECTOR AND INSTRUMENTATION DEVELOPMENT

4π Neutron Counter for 55-Gallon Barrels

J. E. Foley and R. B. Walton

The mechanical construction of the 4π barrel coincidence counter[20] has been completed and the counter is presently undergoing preliminary testing and evaluation.

Description

The sides of the counter consist of a four-inch thick annulus of polyethylene containing 36 2-in. diameter BF_3 detectors; the top and bottom of the counter consist of 4-inch thick slabs of polyethylene, each containing nine 2-inch diameter BF_3 detectors. The annulus separates into two parts to allow introduction of a 55-gallon barrel. The top and sides of the counter are surrounded by a one-foot thick water shield. Figure 24 (upper photo) shows a general view of the 4π barrel counter in its "open" (separated) configuration, with a 55-gallon barrel inserted; the lower photo shows the barrel enclosed in full 4π shield.

Operating Parameters

Initially the counter has been operated without a cadmium sleeve on the inside of the polyethylene annulus--this is to keep the neutron counting

[19] LA-3921-MS, p. 20 (1968).

[20] J. E. Foley, D. B. Smith, and R. B. Walton, LA-4457-MS, p. 31 (1970).

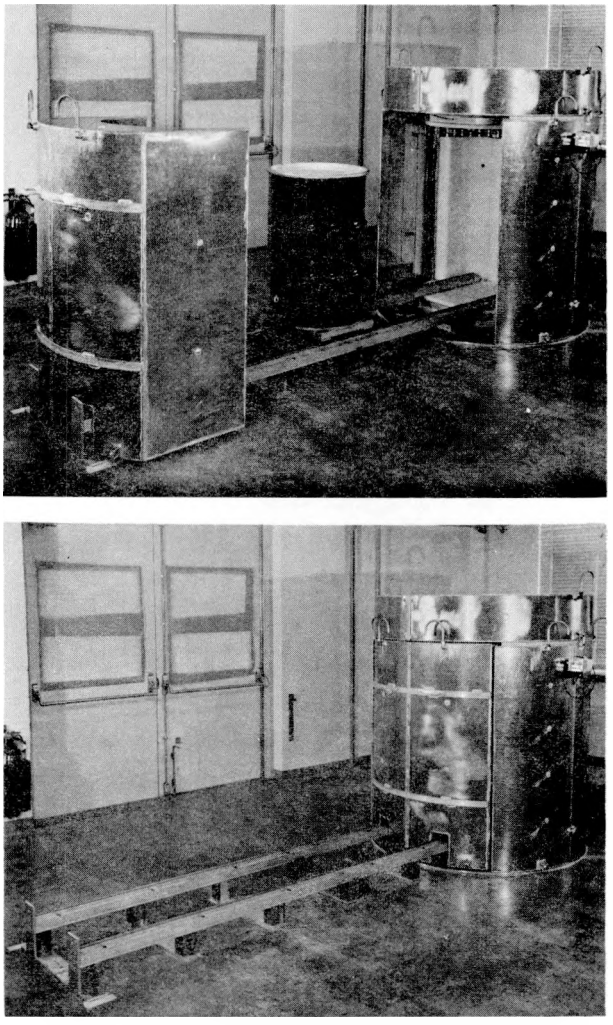

Fig. 24. Neutron coincidence counting system for assay of 55-gallon barrels. Upper photo shows the barrel ready for insertion into counter with shield section withdrawn. Lower photo shows the barrel enclosed in full 4π shield.

efficiency as high as possible and results in a long neutron lifetime (measured to be 125 μsec). This configuration is useful for low level counting, i.e., less than a few grams of plutonium in a barrel. The single-neutron counting efficiency was measured to be 12 percent, and the coincidence efficiency was measured to be 1.5 percent. The lifetime of 125 μsec will decrease to ~30 μsec when the cadmium layer is inserted inside the polyethylene annulus and of course the single-neutron and coincidence counting efficiencies will also decrease.

Coincidence Background

It has been found that neutrons generated by cosmic rays produce a considerable coincidence background in the counter. At Los Alamos (Elev. ~7500 ft) this background amounts to 0.250 ± 0.002 coincidence counts per second--the same rate as from a 0.75 g sample of plutonium (enriched in ^{240}Pu to five percent). This coincidence background limits the sensitivity of the counter to about one gram of plutonium. At altitudes near sea level, where most commercial plutonium processing facilities are located, the cosmic ray coincidence background for a detector of this type should not pose a significant background problem. The one-foot thick water shield reduces the single neutron background rate by a factor of four, but does not reduce the coincidence background appreciably. The coincidence background in a small slab detector was reduced by a factor of two when surrounded by three feet of concrete. Many feet of concrete or earth shielding would therefore reduce the coincidence background significantly; such shielding is not required for few-gram sensitivity of the 4π barrel counter, but would be necessary if sensitivities of less than one gram Pu were desired.

Six 2 x 4 x 8 in. lead bricks (~ 170 lbs.) were placed in the barrel counter to measure the production of neutrons by cosmic rays in high-Z materials. The observed coincidence counting rate (above background) was 0.91 ± 0.02 counts per second, which is equivalent to about 2.7 g of plutonium. It is interesting to note that the 4π-gallon can counter exhibits the same cosmic ray background effect.

Analytical Study

An analytical model has been developed to predict the counting rates of the various scaler channels and to predict the time required to assay a sample to within a given experimental precision. Assumptions used in the model are:

1) the neutron decay in the counter is described by a single lifetime,
2) the electrical pulses from the detector are of zero width (dead times associated with pulses are negligible with respect to dead times associated with gate openings),
3) the dead time associated with the counting process is equal to the gate width of the coincidence channel plus any delay time associated with gate openings.

A comparison was made between the values predicted by the analytical model and the actual scaler readings from the 4π-barrel counter with a 8.097 gram plutonium sample (0.422 g ^{240}Pu). The measured and predicted scaler readings are shown in Table VI. Since the random neutron production rate of the sample was not known it was necessary to vary this rate in the analytical model until the predicted total rate equaled the measured total rate. Similar results were obtained when the analytical model was applied to measurements obtained from the 4π-gallon can counter.

An assay of two known samples of plutonium was made as a check of the operation of the entire 4π-barrel counter system. A sample of 8.097 gm Pu (0.442 g ^{240}Pu) was used as a standard and known samples of 15.9 g and 41.0 g of plutonium were used as the "unknown" samples to be assayed. The magnitudes of the unknown samples were determined from the scaler readings by the relationship:

$$S_2 = S_1 \frac{\dot{C}_2 - \dot{R}_2}{\dot{C}_1 - \dot{R}_1}\left[\left(\frac{\dot{T}_2}{\dot{T}_1}\right)\left(\frac{\dot{T}_1 t_g - \dot{R}_1 t_d}{\dot{T}_2 t_g - \dot{R}_2 t_d}\right)\right]$$

where the index (1) refers to the standard sample and the index (2) refers to the unknown sample.

S is the amount of plutonium,
$\dot{C}$ is the count rate in the coincidence scaler,
$\dot{R}$ is the count rate in the random scaler,
$\dot{T}$ is the count rate in the total scaler,
t_g is the coincidence gate width,
t_d is the dead time associated with each gate.

The terms in the square brackets are the dead time corrections. This assay equation is valid assuming that

1) the efficiency of the counter is the same for both the standard sample and the unknown sample, and
2) the neutron lifetime of the counter is the same for both the standard sample and the unknown sample.

The resulting assayed values of the samples are shown in Table VII.

TABLE VI

COUNTS OBSERVED IN 1000 SECONDS FROM 0.442 gm ^{240}Pu SAMPLE; 100 μs GATE WIDTH

	Coinc. Scaler	Random Scaler	Total Scaler
Measured	3600	550	72,000
Predicted	3430	500	72,000

TABLE VII

4π-BARREL COUNTER; TEST ASSAY OF "UNKNOWN" Pu SAMPLES

Actual	Measured*
15.9 gm	16.2 ± 0.3 gm
41.0 gm	40.6 ± 0.3 gm

*Using 8.019 gm Pu sample as standard

Calculation of Detector Lifetime and Efficiency

D. B. Smith

In the design of a pulsed source-detector experiment, the fraction of counts collected in the detector in a given time is of considerable interest. The time-dependent Monte Carlo code TDMC has

been modified to tally the number of neutron captures in any designated time region of interest. This modification permits easy calculation of the fraction of counts collected in a detector as a function of time.

The response to thermal neutrons of a 1-in. diam by 24-in. long, 4-atm ^{3}He counter in a 2-in. diam polyethylene sleeve has been calculated. The resulting curve of fraction of captures vs time is shown in Fig. 25. The calculations indicate that 40% of the counts are collected within 10 μsec and 98% within 50 μsec.

The capture tally option in the Monte Carlo code also permits the direct calculation (with no normalization necessary) of the neutron detection efficiency of any counter assembly. The technique has been applied to the "thin slab" neutron detector.[21] This detector consists of 19 ^{3}He counters in a 3 x 20 x 24 in. slab of polyethylene covered with cadmium. The response of the detector to a point neutron source one meter from the detector face was calculated as a function of incident neutron energy. The resulting curve of detector efficiency is shown in Fig. 26. The individual points shown are experimental measurements[22] using calibrated neutron sources. The measured values have been arbitrarily increased by 10% to correct for counts lost because of the detector bias setting. The experimental point at 14.9 MeV lies significantly above the calculated curve primarily because of room return neutrons incident on the detector.

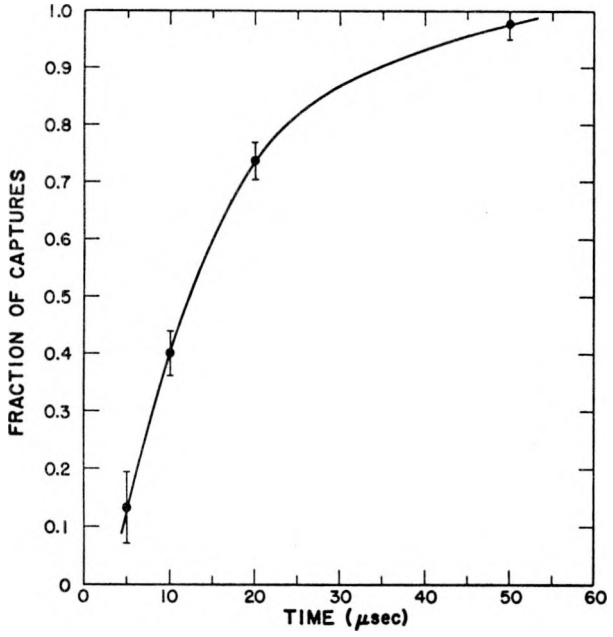

Fig. 25. Neutron capture as a function of time in a 1-in. diam ^{3}He counter surrounded by a 1/2-in. thick polyethylene sleeve.

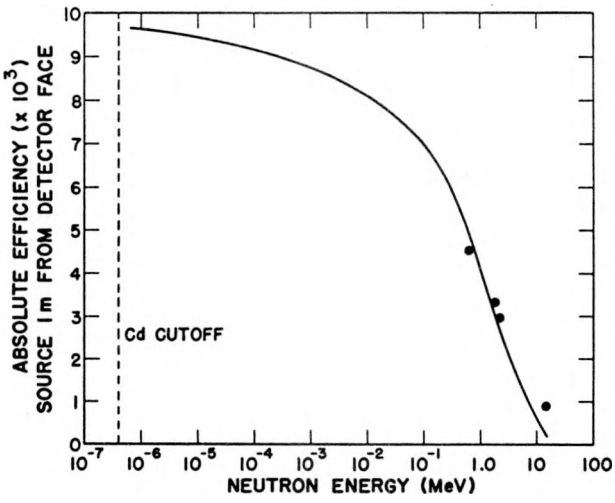

Fig. 26. Efficiency of "thin slab" neutron detector. The solid line is the result of a Monte Carlo calculation; the individual points are experimental measurements.

It is seen in Fig. 26 that the efficiency increases rapidly with decreasing incident neutron energy to about 100 keV and then rises more slowly for several decades of energy to the cadmium cut-off energy.

Multiplication Effects in 4π Neutron Counter Assay of One-Gallon Cans

R. A. Forster

A problem of possible concern in passive neutron assay of Pu (based on the spontaneous fis-

[21] H. O. Menlove and R. H. Augustson, LA-4368-MS, p. 13 (1969).

[22] ibid., p. 14.

sion of ^{240}Pu) is the error introduced by multiplication which would indicate that more ^{239}Pu is present than is actually the case. As noted previously[23] the actual count rate, CR, in the 4π counter is proportional to

$$CR \sim SL/(1-k)$$

where, in this case, the source strength S is the number of ^{240}Pu atoms present, and L and k are the epi-cadmium leakage and multiplication factor respectively of the sample in the counter. For the most accurate assay, the count rate should be a linear response with increasing ^{239}Pu content. This is not the case because of changing leakage and multiplication for each sample. The error introduced is given by

$$(CR_M - M \cdot CR_{1\,g})/(M \cdot CR_{1\,g})$$

where M is the mass of ^{239}Pu (g) being assayed.

To investigate this error, calculations of the multiplication factor k were performed using the DTF-IV code (with the Hansen-Roach 16-group cross sections) for the most reactive[24] Rocky Flats scrap cans, i.e., homogeneous 100-mesh graphite scarfings. The results are shown in Fig. 27. For loadings of up to 500 g ^{239}Pu and 5.5% ^{240}Pu, the error introduced in the assay is less than 5% and thus is not of major concern. Higher ^{240}Pu abundance means reduced multiplication effects since ^{240}Pu is a good poisoning material.

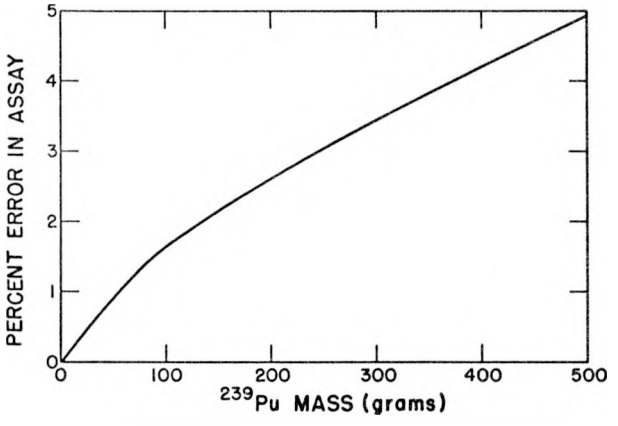

Fig. 27. Calculated percent error of assay in the 4π-gallon can counter caused by multiplication effects vs ^{239}Pu mass for a matrix of 100-mesh graphite scarfings.

Electronics Development and Fabrication

L. V. East, M. M. Stephens, and J. E. Swansen

The following electronic instruments were designed or fabricated during this reporting period.

Eight Channel Pulse Amplifier

A single width NIM module containing eight independent pulse amplifiers was designed and fabricated. Fast integrated circuit operational amplifiers and output drivers were used. Each amplifier has a variable gain of 0 to 2, and maximum pulse output of 8V. This unit was designed to be used to equalize input pulses to the Eight-Input Single Channel Analyzer previously described.[25]

Clock Generator

A crystal-controlled pulse generator having output periods of 10 ms, 100 ms, 1 sec, 10 sec, and 0.1 min, housed in a single-width NIM module was designed and built. The output can be controlled by an external start-stop signal, and an input is provided for a "dead-time" signal. The unit is designed to control the counting time of scalers in pulse-counting systems.

Proportional Counter Preamplifier

The design of a compact general purpose charge-sensitive proportional counter preamplifier was started during this reporting period. The design is similar to that of preamplifiers presently used, with changes being made to reduce the circuit complexity and physical size of the unit.

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[23] D. B. Smith and R. A. Forster, LA-4457-MS, p. 11 (1970).

[24] i.e., largest k_{eff} for a given amount of fuel.

[25] L. V. East and M. M. Stephens, LA-4457-MS, p. 34 (1970).

Neutron Coincidence Unit

Four more of the neutron-coincidence units previously described[26] were aligned, tested, and readied for use during this reporting period.

Ge(Li) Detector Recovery

L. V. East and M. M. Stephens

A Ge(Li) detector that had suffered loss of resolution due to some unknown cause was recovered. The germanium crystal surface was lapped and etched, and a "clean-up" drift was performed at a temperature of about -20°C. After the drift process was completed, the detector performance was better than when originally purchased.

COMPUTATIONAL AND DATA-ACQUISITION FACILITIES

Coupled Neutron-Gamma Transport Code

C. R. Weisbin and R. A. Forster

The Morse code[27] has been obtained from the Radiation Shielding Information Center at Oak Ridge. It solves time-dependent problems in generalized three-dimensional geometry[28] as does the Monte Carlo code already being extensively used in nuclear safeguards analytical work at LASL. In addition, however, it has two significant features not incorporated in the present code. The first of these is the capability to handle either neutron, gamma, or coupled neutron-gamma problems. This feature is particularly important for those assay techniques based upon the (γ, n) or (n, γ) reactions. Secondly, the code inputs group-averaged cross sections in the DTF format. Thus, the cross section libraries which have been used for discrete-ordinates calculations can be incorporated with little or no change. Finally, the group cross-section feature makes possible a direct comparison of solutions obtained from these two different transport calculational techniques.

The version of the code received was operational on the IBM 360/91. Incompatibilities between the Fortran languages for the CDC-6600 and the IBM-360/91 have since been resolved. Presently, those Morse routines written in assembly language for the 360 are being reprogrammed for the CDC-6600.

Analytical Studies Related to Fuel Burnup

C. R. Weisbin and R. A. Forster

The Cinder code,[29] a fuel depletion and decay heating program has been obtained from Prof. Charles Maynard of the University of Wisconsin, along with an extensive data collection of fission product yields, branching ratios, cross sections, etc. The program is to be applied to the three problems cited below:

1) Predict the concentrations and activities of selected fission products which might prove useful in discriminating between ^{239}Pu and ^{235}U. For example, two such fission products are ^{106}Ru and ^{95}Zr which have characteristic gamma-ray energies and yields (in thermal fission) as shown in Table VIII.
2) Predict fission product backgrounds for a capture gamma-ray discrimi-

[26] L. V. East, M. M. Stephens, and J. E. Swansen, LA-4368-MS, p. 16 (1969).

[27] E. A. Straker et al., ORNL 70-2-31, Feb. 1970.

[28] It should also be noted that the Morse code contains specialized one-dimensional geometry packages as well.

[29] See T. England, University of Wisconsin doctoral thesis, August, 1969.

TABLE VIII

GAMMA-RAY ENERGY AND YIELD CHARACTERISTICS OF ^{106}Ru AND ^{95}Zr

Fission Product	Gamma Energy (keV)	% Yield in ^{235}U Fission	% Yield in ^{239}Pu Fission
^{106}Ru	510, 620, 2066	0.389	4.55
^{95}Zr	726, 758	6.45	4.86

nation technique and optimum time for performing the measurement.

3) Determine the sensitivity of group cross sections and burnup as a function of the reactor history.

Problems of fission product migration and fuel contamination will be considered with regard to Problems 1) and 2) if the techniques appear feasible.

A sample problem is presently being prepared. Ultimately, it is hoped that a detailed gamma spectra option as a function of time can be incorporated into the calculation.

Cross Sections: Acquisition and Processing

C. R. Weisbin

In addition to the LASL compilation[30] of neutron cross sections, Group A-1 has added to its library the ENDF/B[31] collection of evaluated neutron data. This addition is significant in two important respects. First, the ENDF/B program is a national effort to develop a standardized, reliable set of neutron information. Thus, the data are continuously being tested by a wide variety of users and therefore should be expected to improve with time. In addition, an organized procedure has been outlined for periodically updating (and presumably, upgrading) the cross-section library. Secondly, the ENDF/B library is available to any user, upon request, in an intelligible format along with the necessary processing codes (written in Fortran IV). Access to the LASL library is presently[32] achieved through the use of special processing codes written for the MANIAC, which are not directly available to the user. It is hoped that the addition of the ENDF/B collection will complement the LASL data, which has been used successfully for the past three years, and that intercomparison of results using different evaluated sets would lead to a better understanding of the information in both libraries.

In preparing for calculation of a hot, spent MTR fuel element, group-averaged cross sections for ^{27}Al in the Goad-Sandmeier[33] 25-group structure were required. These were obtained both by request from R. Lazarus[34] and by operating upon the ENDF/B data with ETOG-1,[35] an associated group-averaging code. The group-averaged ENDF/B data were then put in a form suitable for comparison with the Lazarus data by a short program written to convert the ETOG-1 output to DTF format. Differences in the group-averaged smooth cross-sections of up to 25% and in the transfer matrices of up to 60% were observed, particularly for the higher energy group (i.e., 14 MeV primary source). At first, these seemingly large differences were attributed merely to the different evaluations.[36]

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30 R. Lazarus and L. Stewart, collection of cross-section evaluations from a variety of sources including LRL, KAPL, UK, and LASL, updated 1968.

31 H. C. Honeck, BNL-50066, revised July 1967.

32 Plans are in progress to convert the LASL library processing codes into Fortran IV for the CDC-6600.

33 H. Sandmeier et al., LA-3415 (1965).

34 R. Lazarus, LASL, private communication (1970).

35 D. E. Tusner and R. A. Dannels, WCAP-384501, (1969).

36 ENDF/B evaluation for ^{27}Al, "Neutron Cross Sections for Aluminum", Joanou and Stevens, GA-5884, (1964).

However, upon group averaging oxygen (used in the calculation of the BWR fuel assembly)[37] wherein the microscopic data in both libraries were the same,[38] similar discrepancies were observed. Apart from the possibility of a coding error, it is quite probable that the group-processing codes represent the scattered spectra differently (since in both of the above cases the discrepancies in the differential data were generally a factor of two higher than those for the smooth cross sections). A detailed study of these group-averaging procedures is being carried forth with regard to Pb, a major component of many nondestructive assay devices.

Computer-Based Data Acquisition System

L. V. East, R. H. Augustson, and H. A. Walter

During this reporting period, a considerable amount of effort went into software development for the PDP-9/L computer system. General routines were written for use with the display and plotter, and for handling pulse-height data stored on "DECtape". The multiscaling software was modified to provide spectrum normalization and control functions for fuel-element scanning. A general polynomial least-squares fitting program was also written.

The installation of a controller for a computer-compatible magnetic tape transport was begun. The use of the tape transport will allow data from the Geoscience pulse height analyzer to be read directly into the PDP-9/L computer and will allow more efficient communication with LASL's CDC-6600 computers.

The system was used for data acquisition and analysis in passive gamma-ray assay work and biased neutron detector development work.

SS Materials Inventory Code

D. B. Smith and M. M. Thorpe

In the course of Nuclear Safeguards research at LASL, Group A-1 has acquired a large number of sources, standards, and samples containing special nuclear material (primarily isotopes of uranium and plutonium). Each of these items may be stored in any one of several repositories. It is important to maintain a record of the current location of each sample, the total mass of material stored in each repository, and the total mass of each isotope in the inventory. A computer code, SSINVEN, has been written to maintain the necessary records.

Each sample is assigned a unique identification number when it is transferred into the group. A record of this number along with a description of the sample, its location, isotope, mass, percent enrichment (if available) and transfer number are maintained on magnetic tape. SSINVEN records transfers into and out of the group as well as internal transfers of a sample from one repository to another and produces a listing of all current transfers. It also produces listings of all items currently in the inventory by identification number, by isotope, and by repository, and gives the total number of items in the inventory, the total mass of each isotope, and the total mass of fissile material stored in each repository.

The code appears to be working well and is expected to greatly reduce the effort necessary to maintain the SS material inventory records in the Nuclear Safeguards Research Laboratory at LASL.

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37 See R. H. Augustson and A. E. Evans, "Nondestructive Assay of Power Reactor Fuel Elements", this report.

38 E. L. Slaggie and J. T. Reynolds, KAPL-M-6452 (1965).

Field Experience

J. H. Menzel, B. R. Dennis (ENG-6), D. E. Helfer, and J. H. Kottmann

On May 8, 1970, the MONAL arrived at the Dow Chemical Co. plutonium facility at Rocky Flats, Colorado, after a 350-mile trip from Los Alamos. Except for a minor logistical problem, all systems including the neutron generator and the rack-mounted electronics survived the trip in excellent condition.

In addition to the assay results presented in the following sections, valuable field experience was gained at Rocky Flats:

First, the magazine-type large-sample handling system has proven to be an operational necessity. For example, plant procedure requires the use of approved fork-lift operators to move 55-gallon barrels containing fissile materials from one location to another. The MONAL conveyor system for 55-gallon barrels and other large samples eliminates the necessity of fork-lift operators who are unfamiliar with the assay procedure to stand by continuously; periodic use of these operators such as once every three to four hours can usually be fitted conveniently into their regular duties. In addition, batch handling of samples greatly simplifies the record keeping and health physics monitoring required in fissile-material transfers from the plant to the MONAL.

Second, active interrogation of Rocky Flats plutonium scrap barrels has shown that the consumption of tritium targets for the 14-MeV neutron generator is much lower than originally anticipated. Present experience indicates the use of one target ($60) per 100 barrels based on a target half-life of 5 ma·hr.

Third, complete sets of tools, maintenance manuals, replacement parts, safety and health physics supplies, and radiation monitoring equipment are essential for the operation of a mobile nondestructive assay laboratory.

And fourth, interdisciplinary knowledge in nuclear physics and the chemistry and materials technology involved in nuclear-materials processing is an immense asset in determining the most efficient solution to a nondestructive assay problem; the MONAL provides the appropriate working environment for such technical exchange between materials people and the developers of nondestructive assay instrumentation.

Neutron Assay of 55-Gallon Barrels

T. D. Reilly and M. M. Thorpe

Since 1964, Rocky Flats Division has used a barrel counter[39] for nondestructive assay of plutonium in process residues and waste. The shielded barrel counter consists of an annular array of eight Geiger-Mueller detectors for passive gamma assay and sixteen BF_3 detectors for a neutron count to correct the observed gamma count (e.g., gamma rays from (α, n) reactions). Because of gamma attenuation variations, individual correction curves for each matrix (except insulation, fire brick, and scrap metal) are used to obtain the proper assay; an accuracy of ± 20% is quoted by Rocky Flats for the barrel counter under these conditions:[39] For this essential calibration as well as to obtain periodic measurement-bias and variability information, the Rocky Flats Chemistry Standards Laboratory has prepared an extensive set of 55-gallon barrel standards.[40] These standards have been designed to simulate as closely as possible the plutonium-bearing solid process materials and residues routinely assayed in the barrel counter. A summary of the salient characteristics of these standards is presented in Table IX. Since these

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39 O. H. Willoughby and D. R. Cartwright, "Measurement of Plutonium in Process Materials and Contaminated Waste", Proc. INMM, pp. 130-142 (1969).

40 L. Doher, "A Measurement Control Scheme for Plutonium Counting Systems", Proc. INMM, Annual Meeting, Gatlinburg, Tenn., May, 1970.

TABLE IX

SUMMARY OF ROCKY FLATS 55-GALLON BARREL STANDARDS

Description	Matrix	Matrix Composition (Wt. %)	Matrix Avg. Net Weight (kg)	Matrix Avg. Density (g/cm^3)	Plutonium (as PuO_2) Loadings Assayed (g Pu)
Graphite molds	60-mesh graphite	100	110	0.53	60, 145, 195
Dry combustibles	Carbon Plastics Cellulose	90 5 5	24	0.12	10, 165, 175
Wet combustibles	Cellulose Water Plastics	80 15 5	51	0.25	28.5, 166
Washables	Polyvinyl Lead gloves Polyethylene Cellulose Surgeon gloves	42 28 20 7 3	32	0.15	10, 90, 160
Raschig Rings	Pyrex glass w. 12% Boron as B_2O_3	100	82	0.39	40, 95, 185
Resin	Dowex 1 x 4	100	-	-	25, 110
Benelex-Plexiglas	Benelex-Plexiglas	-	-	-	75

standards involve a very substantial investment in money and time, they present a unique opportunity for testing the range of active and passive techniques employed in the MONAL.

The delayed-neutron response of the 17 standards was studied during MONAL deployment at Rocky Flats. The high efficiency, "thin" slab detector was used to measure delayed neutrons, and a ^{239}Pu fission chamber was used to correct for changes in the irradiating neutron spectrum caused by moderating material in the barrel. The fission chamber response varied by an order of magnitude over these barrels indicating the presence of much moderating material. The barrels were unreflected so there were large spatial variations in delayed-neutron response within the barrels.[41] This is illustrated in Fig. 28 (top curve), where the ratio of the observed plutonium loading

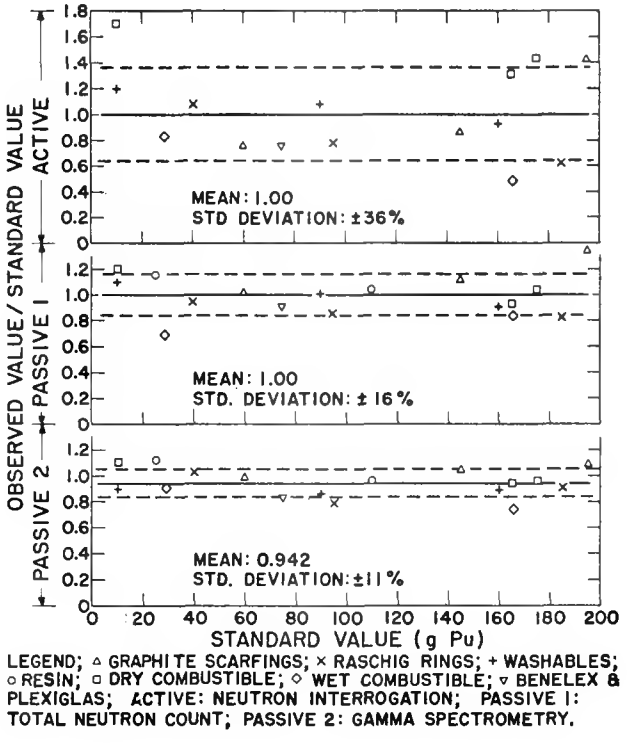

Fig. 28. Active and passive assay results of Rocky Flats 55-gallon standards.

41 M. M. Thorpe and T. D. Reilly, LA-4457-MS, pp. 11-16 (1970).

to the known standard value is plotted against the standard value. The standard deviation of these measurements is 36%.

The addition of an effective neutron reflector around the barrel will reduce these spatial effects but will increase self-absorption in the fissile material. The latter effect should be tolerable in many cases involving hydrogenous waste. This will also improve the sensitivity of the method which is already very good (0.1 g or better, in a 55-gal drum of hydrogenous waste).

The middle curve in Fig. 26 shows the results of passive neutron assay of these same standards using the gross neutron count from the drums. These neutrons come from the spontaneous fission of ^{240}Pu and the (α, n) reaction on oxygen in the PuO_2. Since the standards contain the same percentage of ^{240}Pu and no other strong (α, n) sources, this neutron count is a good measure of plutonium content. The standard deviation of these measurements is 16%. Such an assay method is subject to large errors whenever (α, n) emitters are present in the waste matrix but could be of use if the absence of such reactions could be independently verified (e.g., using a simultaneous gamma measurement).

Since metal wastes do not present the same moderation difficulties for active assay, a sampling of seven such barrels was assayed using delayed-neutron response. These were process line wastes and the only Pu value available for comparison was that assigned by the Rocky Flats barrel counter. No standard was available for this matrix as the high attenuation of metals makes gamma-ray assay difficult. Results of these active measurements are presented in Table X. The passive neutron results are included to illustrate the problems caused by strong (α, n) reactions. The agreement between the two assay results on the first six barrels is surprisingly good (~ 20%) in view of the basic difference between the two methods. Examination of the seventh barrel with the NaI gamma-scanning system and a Ge(Li) detector showed a strong, localized source of ^{208}Tl (2.61 MeV and related lines). This

TABLE X

ACTIVE NEUTRON INTERROGATION OF ROCKY FLATS 55-GALLON METAL SCRAP BARRELS

Sample Identification Number	Total Neutron Technique (g Pu)[a]	Rocky Flats Barrel Counter[b] (g Pu)	Neutron Interrogation Technique[c] (g Pu)
CO-60083	673	85	108 ± 5[d]
CO-50108	2050	71	91 ± 4
TO-60084	415	48	67 ± 3
RO-59930	34	21	20 ± 2.4
CO-59991	66	5	3.5 ± 1.5
TO-59875	636	200	227 ± 8
LO-60009	77	362	14 ± 1.5

[a] Presented for illustration purposes only.

[b] See Ref. 39

[c] See Ref. 41

[d] Standard deviation based on statistics only.

gamma spectrum indicated the presence of ^{232}U (probably in conjunction with ^{233}U) and a small amount of ^{239}Pu, consistent with the active assay. This intense gamma source was the cause of the erroneous assay given by the Rocky Flats barrel counter. This incident illustrates the value of a gamma waste assay system having inherently high gamma-ray energy resolution.

Gamma-Ray Assay of Fifty-Five-Gallon Barrels

J. L. Parker, T. D. Reilly, and R. B. Walton

The gamma-ray scan system for 55-gallon barrels described in the previous progress report[42] has been completed. The system has eight detector modules, each containing a shielded 2 x 2 in. NaI crystal and incorporating a fixed 4 in. of vertical collimation and adjustable horizontal collimation. These modules are stacked vertically and can be spaced at 4, 5, or 6 in. on centers. Opposite the detector stack is an array of eight collimated, 100 μci ^{22}Na sources which allow a transmission measurement of the barrel on a detector-by-detector basis. While counting the gamma-activity of the sample drum these sources are shielded from the detectors. Figure 29 shows a general view of the complete 8-channel passive system for gamma-ray assay of 55-gal barrels. The complete system fits nicely into the MONAL interrogation cavity.

The system was installed in MONAL at Rocky Flats and tested on the 17 PuO_2 barrel standards described in Table IX. For this assay the "double-position scan" was used wherein the barrel is centered in the array, counted, moved half the crystal spacing vertically, and counted again. For this series of measurements a 6-in. detector spacing was used. With the 6-in. spacing only six of the eight detectors and sources are effective, each viewing a small vertical slice of the barrel (~ 4.5 in. for the two end tubes, and ~ 6 in. for the four center tubes). A barrel is moved into the cavity and a 200-sec transmission measurement made which gives the transmission through each barrel section. As described previously[42] this transmission measurement determines the appropriate horizontal collimation for each detector. Then the plutonium activity is measured by a 100-sec count at each of the two vertical positions for the "double-position scan". By treating the response of the individual tubes separately, errors due to matrix inhomogeneities are reduced considerably. The whole procedure takes about 10 minutes; this time can be reduced if favorable conditions (such as uniform matrix density) allow a simpler scan procedure.

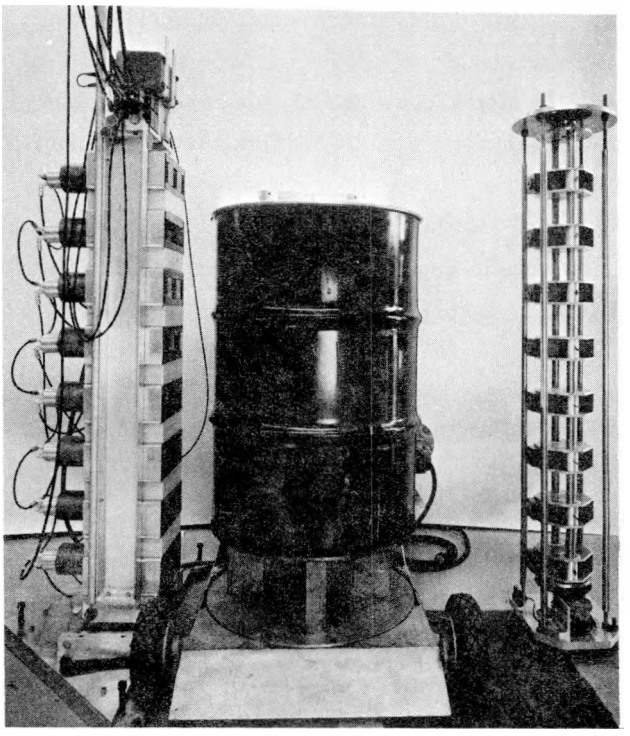

Fig. 29. Eight-channel gamma scanning system for 55-gallon barrels employing the rotation-collimation principle. The eight NaI scintillation detectors are seen at left in the articulated Pb-collimator geometry, and the eight transmission sources are at right in individual Pb collimator-shields.

The calibration for this test was obtained by combining computer calculations of the system spa-

[42] J. L. Parker, R. B. Walton, T. D. Reilly, LA-4457-MS, pp. 19-23 (1970).

tial response with experimental numbers from a 50-g sample of Pu (as PuO_2) dispersed in a 2-qt can of diatomaceous earth. Results of the assay are presented in Fig. 28 (bottom curve); the number given here is the ratio of the observed value to the known standard value. The measured values differ from the known values by a standard deviation of 11%.

The eight-channel NaI system was also used to assay a representative sampling of normal production barrels before leaving Rocky Flats. Table XI lists typical results along with the value indicated by the Rocky Flats barrel counter. The most glaring difference in assay results occurred in the barrel of filters (Sample No. FO60270). Here the NaI system showed a large concentration of Pu in the bottom 6 in. of the barrel and near the periphery. This situation led to a count rate too high for the Geiger tubes of the Rocky Flats counter; the resulting large dead time causes the low assay value. The amount and distribution of fissile material is not mocked up well in the filter standard upon which the Rocky Flats counter system depends.

These results show the advantages of an energy-sensitive gamma-ray assay system which is also capable of reasonable spatial resolution in large containers. The eight-channel gamma-scan method which has proved so useful for passive assay of Pu is presently being extended to assay of ^{235}U scrap and waste.

One-Gallon Plutonium Scrap Cans

H. O. Menlove, B. R. Dennis (ENG-6), and J. H. Menzel

Rocky Flats Division performs plutonium assays on gallon-size waste and residue packages, produced in the process area, with a can counter [39] of similar design as the barrel counter previously described. In addition, a "helix counter" [39] is used to scan gallon-size containers of homogenized process residues with a NaI (Tl) detector; the transmission of gamma radiation from an external plutonium source through the sample yields information on sample homogeneity and density. Again, the Rocky Flats Chemistry Standards Laboratory has assembled a set of plutonium standards with a range of plutonium contents for each representative matrix, as summarized in Table XII.

For the can counter, the plutonium (PuO_2) mixed with the matrix material is enclosed in small containers which can be distributed heterogeneously in one-gallon polyethylene bottles; the small containers can be changed periodically to alter the plutonium loading and the spatial distribution within each one-gallon standard. For the helix counter,

TABLE XI

GAMMA-RAY ASSAY OF ROCKY FLATS PRODUCTION SCRAP

Sample Identification Number	55-Gallon Barrel Contents	Rocky Flats Barrel Counter (g Pu)	Eight-Channel NaI Assay (g Pu)
CO59952	Burnables	35	18
CO60396	Resin	34	21
71-7500	Graphite	56	53
LO60647	Combustibles	93	95
FO60270	Filters	203	468
CO60108	Hot Metal	71	124
RO60637	Hot Plastic	21	20

TABLE XII

SUMMARY OF ROCKY FLATS ONE-GALLON CAN STANDARDS

Description	Matrix	Matrix Composition (Wt. %)	Matrix Avg. Net Weight (kg)	Matrix Avg. Density[a] (g/cm^3)	Plutonium (as PuO_2) Loadings Assayed (g Pu)
Graphite scarfings	60-mesh graphite	100	2.26	.60	10, 100, 145
Insulation	SiO_2	68	2.70	.70	10, 80.4, 145
	Al_2O_3	10			
	MgO	10			
	CaF_2	5			
	$Ca(OH)_2$	5			
	Asbestos	2			
Slag	CaF_2	96	2.27	.60	10, 150, 155
	MgO	2			
	$CaCO_3$	2			
Ash	Carbon	51	1.50 and	.40	10, 25,
	SiO_2	30	1.25	.33	65
	Fe_2O_3	15			
	MgO	3			
	Al_2O_3	1			
Glass	6 mm Pyrex Glass Beads w. 2% Boron in B_2O_3	100	2.72	.72	3, 21.1, 53.2
Ash[b]	SiO_2	40	3.87 to	1.02 to	10, 25, 50, 100
	Carbon	20	4.26	1.13	
	Fe_2O_3	20			
	MgO	20			
	CaF_2	0.1			
Graphite scarfings[b]	100-mesh graphite	100	3.63 to 4.23	0.96 to 1.12	10, 25, 50, 100, 200, 350, 500
	CaF_2	0.1			

[a] Based on the one-gallon volume. Local density for heterogeneous matrix ≈ 2 x avg. density.

[b] Homogeneous matrix, all others are heterogeneous.

the plutonium (PuO_2) has been blended with the matrix material to achieve a homogeneous plutonium distribution as well as a uniform matrix distribution.

Matrix and spatial effect studies and neutron response calibration of the portable coincidence counter combined with the "source addition" technique[43] had been performed at the Los Alamos Nuclear Safeguards Laboratory prior to field deployment of the MONAL at Rocky Flats. Excellent results[43] had been obtained on a set of Hanford ash standards still in their 6-M shipping containers. Thus, the coincidence counter was ready for assay work at Rocky Flats.

Fifteen heterogeneous and eleven homogeneous standards whose plutonium content was unknown to MONAL personnel were assayed with the neutron-coincidence system. A small (~ 10^{-9} g) ^{252}Cf source of spontaneous-fission neutrons was used for the "source addition" technique to "nor-

43 H. O. Menlove and R. B. Walton, LA-4457-MS, pp. 27-31 (1970).

malize out" matrix and multiplication effects without prior knowledge of the content of any standard. The data were corrected for dead-time to obtain the final results. The assay results were then presented to Rocky Flats personnel for comparison with their standard values. The results are presented in Figs. 30 and 31 for the heterogeneous and homogeneous cases, respectively.

The plutonium loading of the heterogeneous standards ranged from 3 g Pu located on the bottom of a one-gallon bottle to 155 g Pu fairly evenly distributed, and the chemical composition varied from pure graphite to slag containing 96% CaF_2 (see Table XII. Yet, a least-squares fit to the assay results plotted in Fig. 30 show a bias of only 1% and a standard deviation (σ) of only 8%. The counting statistics of coincidence events were 2% or better, and the counting time per sample varied up to a maximum of 1000 sec.

The homogeneous standards contained only two matrix materials, namely either graphite or ash, but the plutonium content ranged up to 500 g. A least-squares fit to the assay results plotted in Fig. 31 again shows a bais of 1% but the standard deviation is reduced to 3% over the complete 10-500 g Pu range. The counting statistics of coincidence events were 2% (up to 25 g Pu) or better, and the counting time per sample was 1000 sec for ≤ 25 g Pu content and 400 sec for the higher plutonium loadings.

These assay results clearly demonstrate the matrix independence, sensitivity, and accuracy of the portable neutron-coincidence passive-assay system combined with the "source addition" technique. For nondestructive assay of plutonium whose isotopic composition is known or independently measurable this passive-assay system should provide a powerful tool for use by production, accountability, and safeguards inspection personnel.

Neutron Fluxes and Adjacent Barrel Background in the MONAL Cavity - Computational

R. A. Forster and D. B. Smith

The neutron flux (n/cm^2/unit source) in various energy bins at several points in the MONAL cavity has been obtained with a detailed Monte Carlo calculation. The lead tailoring assembly is in place around the 14-MeV source, and all the cavity walls except the floor are covered with borated PVC. There is no PVC on the ground, and the ground scatter shield is in place. The results of

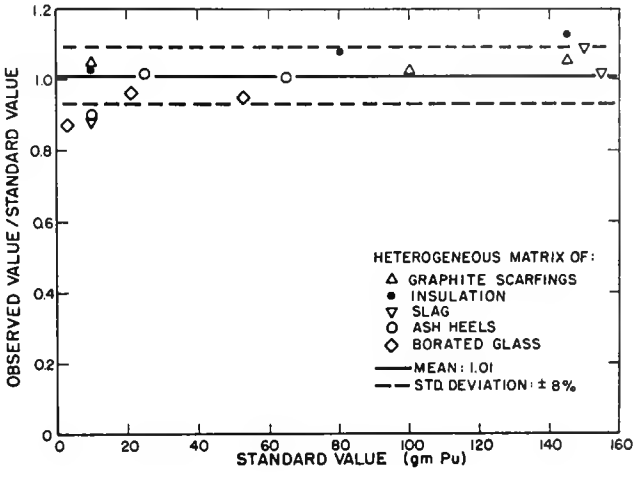

Fig. 30. Nuetron coincidence counter; assay results of Rocky Flats heterogeneous scrap cans.

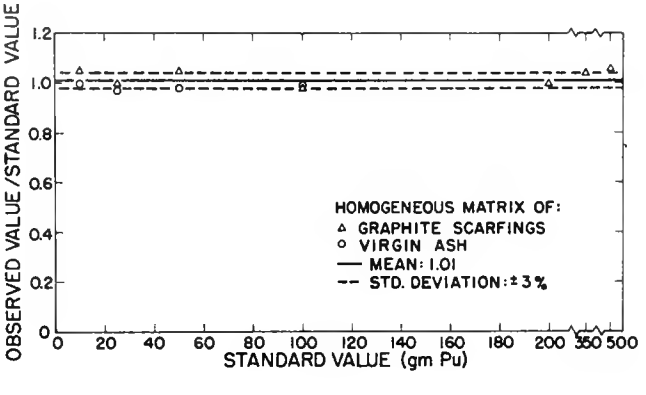

Fig. 31. Neutron coincidence counter; assay results of Rocky Flats homogeneous scrap cans.

the calculations are shown in Fig. 32 where the neutron flux

$$\Phi(E_1, E_2) = \int_{E_1}^{E_2} \Phi(E)dE$$

is plotted versus energy. An energy cutoff of 0.025 eV was chosen to expedite the calculation. All points are directly in line with the source. The error bars represent the largest standard deviation of each case. Note that the fast groups decrease roughly as $1/r^2$ while the thermal flux appears to be relatively constant throughout the cavity.

Reaction rates can be determined from Fig. 32 by folding in the appropriate cross sections for each energy bin. Estimates of ^{235}U cadmium ratios and cadmium-covered $^{235}U/^{238}U$ ratios are 1.03, 1.08, 1.26 and 4.7, 8.0, 19, respectively, at the three indicated distances from the source. Assay in the MONAL of a barrel containing plutonium is complicated by a continuous background of neutrons from the spontaneous fission of ^{240}Pu. In addition to this background from the barrel in the cavity, barrels on the monorail conveyor underneath the MONAL can produce a background at the detector. Several Monte Carlo calculations were performed to investigate the magnitude of this background and the possibility of reducing it.

The neutron flux at the detector position in the cavity from a fission spectrum source at the center of each of the two barrels adjacent to the elevator was calculated. Ground scatter--but no return from the cavity walls--was included. The same flux was than calculated for a 4-in. thick slab of borated (25% B_4C) CH_2 on the floor of the cavity. The calculations indicate the background from each of the adjacent barrels to be ~ 10% of the background from the barrel in the cavity, assuming equal amounts of plutonium in all three barrels. The slab of borated CH_2 between the conveyor and the detector should reduce the background from barrels on the conveyor by approximately an order of magnitude.

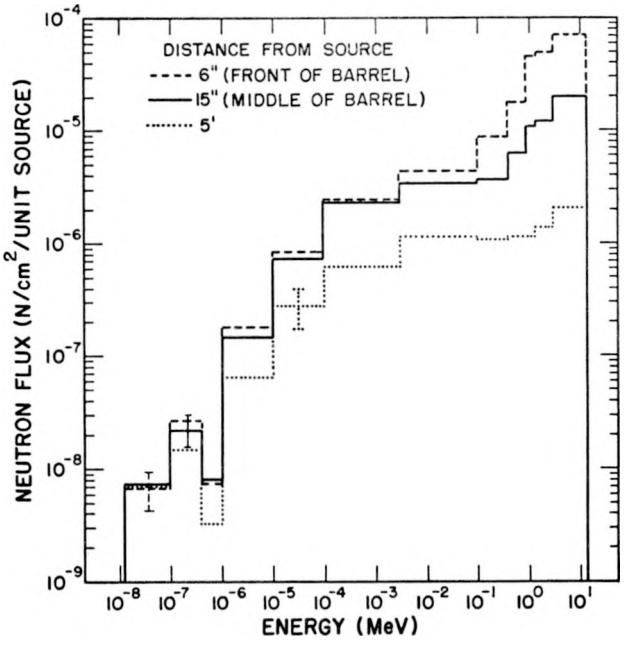

Fig. 32. Calculated neutron flux (n / cm^2 / unit source) vs energy for the MONAL cavity with borated PVC on all cavity walls except the floor, the 14-MeV source in the lead-tailoring assembly, and the ground shield in place.

PUBLICATIONS

1. J. H. Menzel, "From Safeguards to Field Exploration--A Mobile Laboratory," invited paper presented at the American Chemical Society National Meeting, February 26, 1970, Houston, Texas.

2. J. H. Menzel, B. R. Dennis, M. M. Thorpe, R. B. Walton, D. B. Smith, and G. R. Keepin, "The Los Alamos Mobile Nondestructive Assay Laboratory," paper presented at the Eleventh Annual Meeting of the Institute of Nuclear Materials Management, May 25-27, 1970, Gatlinburg, Tenn.

3. J. H. Menzel, B. R. Dennis, M. M. Thorpe, R. B. Walton, D. B. Smith, and G. R. Keepin, "A Mobile Laboratory for Nuclear Safeguards," paper presented at the IAEA Symposium on Progress in Safeguards Techniques, July 6-10, 1970, Karlsruhe, Germany.

4. R. H. Augustson, H. O. Menlove, R. B. Walton, L. V. East, A. E. Evans, and M. S. Krick, "Development of Techniques for Active and Passive Assay of Fissionable Materials," paper presented at the IAEA Symposium on Progress in Safeguards Techniques, July 6-10, 1970, Karlsruhe, Germany.

5. G. R. Keepin, H. O. Menlove, M. M. Thorpe, R. H. Augustson, C. N. Henry, D. B. Smith, and T. D. Reilly, "Application Areas and Results of Nondestructive Assay Measurements," paper presented at the IAEA Symposium on Progress in Safeguards Techniques, July 6-10, 1970, Karlsruhe, Germany.

6. H. O. Menlove, R. H. Augustson, L. V. East, A. E. Evans, and G. R. Keepin, "Neutron Interrogation Techniques for Fissionable Material Assay," paper presented at the Meeting of the Institute of Nuclear Materials Management, May 25-27, 1970, Gatlinburg, Tenn.

7. L. V. East, "Baseline Stabilization in Direct Coupled Counting Systems," Rev. Sci. Instr. 41, 1245 (1970).

8. E. E. Gross, E. V. Hungerford, J. J. Malanify, and R. Woods, "Nucleon-Nucleon Scattering Parameters from Final-State Interactions," Phys. Rev. 1C, 1365 (1970).