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of the
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LOS ALAMOS • NEW MEXICO

Nuclear Safeguards
Research and Development

Pulsed Neutron Research Group, N-6

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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 - Computational

D. B. Smith and R. A. Forster

A series of neutronics calculations has been undertaken to supplement the previously-reported experimental investigation⁽¹⁾ of the effect of non-fissionable matrix materials in one-gallon recovery cans on delayed-neutron assay results. The one-dimensional, discrete-ordinates computer code, DTF-IV, was used with 25-group cross sections for these calculations. The experimental setup described previously⁽¹⁾ was modelled as concentric spherical shells of iron, cadmium, and carbon surrounding a cavity containing the (spherical) one-gallon can. The source spectrum used was that of 14 MeV neutrons moderated by two inches of lead, and was located on the inner surface of the carbon shell. Each configuration (i. e., density) of matrix material required two DTF-IV runs, the first to calculate the neutron flux distribution in the can due to the source, and the second (an adjoint calculation) to determine the neutron leakage probability from the can. The results of these two calculations were then combined by a computer code, GALCAN, to give the delayed neutron response of fissionable material as a function of radius in the

can. The LAMPS system⁽²⁾ was used to permit all three of these calculations to be combined in a single computer run. The agreement between calculated and experimental results for plutonium metal in a low-density ($\rho = 0.06 \text{ g/cm}^3$) CH_2 matrix indicated this calculational model to be sufficiently accurate to predict the effects of various matrix-material configurations.

Both calculation and experiment showed a large flux gradient in the void between the can and the wall of the cavity because thermal neutrons were absorbed in the cadmium liner. Such a gradient makes the response of an external normalizing device (e. g., a fission chamber placed in the void) extremely dependent upon position. Moving the cadmium to the outside of the iron shell permits more sub-cadmium neutrons to be reflected back into the can, thus reducing the flux gradient in the void and also raising the delayed-neutron response near the edge of the can. Calculations were done for four different cavity-liner combinations: Cd + Fe, Fe + Cd, Pb + Cd, and Pb + W + Cd, where the notation lists liner materials in order of increasing radius. The results of these calculations are shown in Fig. 1. Both expected effects of moving the cadmium to the outside of the cavity liner (i. e., reduced flux gradient in the void and flat-

¹ R. H. Augustson and H. O. Menlove, LA-4368-MS, pp. 1-3 (1969).

² See description of LAMPS system, this report.

Fig. 1. Calculated relative delayed neutron response per gram versus radius in one-gallon cans for Pu distributed uniformly in three densities of polyethylene matrix for various combinations of lining materials in the active interrogation cavity.

tening of the response across the can) were confirmed. The iron plus surrounding cadmium liner (Fe + Cd) was chosen as the most effective of the four configurations, and was used in all subsequent calculations.

Figure 2 shows the delayed-neutron response per gram of plutonium as a function of radius within the can for the full range of densities of polyethylene matrix. All results are normalized to the delayed-neutron response of one gram of plutonium placed in the void 0.5 cm from the edge of a can containing no matrix material. The calculations assumed a "flat-response" detector. The drop in the response curve for full-density ($\rho = 0.92 \text{ g/cm}^3$) polyethylene near the center of the can is largely

Fig. 2. Calculated relative delayed neutron response per gram versus radius in one-gallon cans for Pu distributed uniformly in polyethylene matrices of various densities.

compensated in the experimental measurements (See Ref. 1, Fig. 1) because the neutron counter used is more efficient for the lower-energy neutrons coming from the center of the can. The response curves for CH_2 densities greater than $\rho = 0.4 \text{ g/cm}^3$ are clearly compressed; this will not seriously affect the assay of dilute systems, but could pose difficulties in the assay of high-Pu-concentration samples. It is important to note here that the higher CH_2 densities (i.e., between $\rho = 0.2 \text{ g/cm}^3$ (tightly packed polyethylene chips) and $\rho = 0.92 \text{ g/cm}^3$) are not generally found in practical assay applications; e.g., as pointed out previously⁽¹⁾ most of the matrix materials contained in one-gallon cans at Rocky Flats correspond to the region below the $\rho = 0.2 \text{ g/cm}^3$ polyethylene curve.

The results shown in Fig. 2 assumed the fissile content of the can to be sufficiently low that it did not significantly alter the neutron flux set up

Fig. 3. Calculated relative delayed neutron response per gram versus radius in one-gallon cans for three masses of Pu distributed uniformly with no matrix material and in a polyethylene matrix ($\rho = 0.2 \text{ g/cc}$).

in the matrix material with no plutonium present (i. e., infinitely dilute plutonium). Figure 3 shows the results of calculations for cans containing 20 and 200 grams of plutonium in a polyethylene matrix of density $\rho = 0.2 \text{ g/cm}^3$. Clearly such masses of plutonium can no longer be considered dilute. The absorption cross section of the plutonium causes

almost no change in the shape of the response curve, but does produce a significant change in the magnitude of the response relative to the infinitely dilute case. If, however, the delayed-neutron response is normalized to the response of a plutonium fission chamber located in the void outside the can, the effects of the amount of Pu on the magnitude of delayed neutron response are significantly reduced. This renormalized response per gram of plutonium for the center, middle, edge, and volume-average of cans containing no matrix material and polyethylene with density $\rho = 0.2 \text{ g/cm}^3$ is given in Table I. The response normalized to the response of the "empty" can is also shown in Table I for comparison. Thus, using fission chamber normalization, the delayed-neutron response per gram in the active interrogation assembly is reasonably independent of Pu loading up to at least 200 grams of Pu.

It should be noted that the modelling and calculational methods described herein take detailed account of all major neutron interaction processes (fission, moderation, capture, leakage) and thus are able to provide very effective guidance in the development of optimized assay systems for a wide range of sample sizes (cc's to gallons), matrix material, and physical and chemical forms in which assay samples occur in the nuclear industry.

TABLE I

DELAYED NEUTRON RESPONSE PER GRAM OF PLUTONIUM
(Relative to Response of Dilute Distribution)

Matrix	Pu Loading	Relative Delayed-Neutron Response			Fission Counter Normalization			
		Center	Middle	Edge	Center	Middle	Edge	Vol Avg.
CH_2 , $\rho = 0.2$	20 g	0.88	0.87	0.86	1.07	1.06	1.05	1.06
	200 g	0.39	0.39	0.40	1.11	1.13	1.16	1.15
None ("empty" can)	20 g	0.98	0.98	0.98	0.99	0.99	1.00	1.00
	200 g	0.88	0.89	0.91	0.96	0.97	1.00	0.98

Delayed Neutron Assay of Fissionable Material In
One-Gallon Recovery Cans - Experimental

R. H. Augustson and H. O. Menlove

The development of a neutron interrogation system for fissionable material assay of one-gallon (and smaller) containers has progressed from the measurements made under laboratory conditions to an engineered configuration suitable for field assay applications in the MONAL. The effects of nonfissionable matrix material on Pu assay inside a Pb-C moderator were reported previously⁽¹⁾ and an assay procedure using Pu fission chamber monitoring was outlined. Based on experience gained from the initial measurements and the results of parallel neutronic calculations,⁽³⁾ a new Pb-C moderating assembly was designed and fabricated. In the course of field operations of the MONAL at

LASL's DP-Site, this assembly was used to perform measurements inside the experimental cavity of the MONAL. Based on this experience, a further design change, namely the addition of a removable B₄C sleeve, was incorporated into the moderator assembly. Because of MONAL scheduling pressures, it was necessary to utilize the Accelerator I facility for subsequent experiments with the Pb-C moderator assembly, taking care to provide an environment similar to the MONAL experimental cavity.

Figure 4 shows a cross-sectional view of the active interrogation system. The ³H target of the neutron generator is surrounded with a Pb-C tailoring assembly which can be mounted on the wall of the MONAL cavity. The graphite assembly has a sample chamber designed to accept samples

Fig. 4. Active interrogation system for recovery can assay.

³ cf. Delayed-Neutron Assay of Fissionable Material in One-Gallon Recovery Cans - Computational, this report.

of various sizes up to one-gallon recovery cans. This chamber is surrounded with a 0.7-in.-thick steel reflector, .16 in. of Cd and a 1-in.-thick outer annulus of B_4C . A Pu fission counter is mounted in the steel cylinder at 90° to the accelerator beam axis. Samples placed in the chamber can be rotated on a turntable with an angular speed of 1 rpm. A thin slab detector optimized for delayed neutron counting⁽⁴⁾ is placed 180° from the 3H target.

Assay Results

The new system was used to assay six cans containing Pu scrap in the form of sand, slag and crucible which were obtained from the Pu processing facility at DP-Site, LASL. These cans were interrogated with two different neutron sources: first, with a ~ 3 -MeV neutron source (obtained by

surrounding the 14-MeV neutron source with Pb) and secondly, with ~ 0.5 MeV neutrons obtained using the Pb-C MONAL moderator as shown in Fig. 4. To obtain standards, separate loadings of 20 and 200 g of PuO_2 were mixed with MgO sand, and packaged similarly to one-gallon scrap cans generated at Rocky Flats. The delayed-neutron response per fission chamber monitor count was measured for each of these cans and the results are listed in Table II. For the lower energy ($\bar{E}_n \sim 0.5$ MeV) neutron irradiation, there was only a 5% deviation from linearity over a range of a factor of 10 in the amount of Pu in the standard cans. For the higher energy ($E_n \sim 3$ MeV) irradiations, there was no significant absorption of neutrons in the Pu.

The standard can containing 20 g of PuO_2

TABLE II

ACTIVE Pu ASSAY OF RECOVERY CANS CONTAINING SAND, SLAG, AND CRUCIBLE MATRIX MATERIALS

Can No.	"By Difference"	Active Assay ^(b)	
		$\bar{E}_n \sim 3$ Mev	Pb-C Moderator
	weight (g Pu)	(g Pu)	(g Pu)
G-1 ^(a)	17.64	Standard	Standard
G-2	176.40	178.0 ± 5.0	168.0 ± 5.0
XBL-40-1	13	$22.5 \pm .7$	$20.6 \pm .6$
XBL-40-2	13	$16.6 \pm .5$	$15.0 \pm .8$
XBL-42	11	$14.4 \pm .5$	$14.7 \pm .6$
XBL-43	23	$24.0 \pm .7$	$21.0 \pm .8$
XBL-44	18	$15.8 \pm .5$	$15.8 \pm .5$
XBL-45	24	$16.5 \pm .5$	$15.0 \pm .5$

^a G-1 and G-2 are standard cans with known Pu content as listed.

^b Uncertainties quoted are statistical errors.

⁴ H. O. Menlove and R. H. Augustson, LA-4368-MS, pp. 13, 14 (1969)

D. B. Smith and R. A. Forster

was used for calibration in the assay of the six sand, slag and crucible cans and the results are given in Table II. Since the assay samples contained large amounts of F and Mg, there was a large neutron background from (α, n) reactions in these materials and it was necessary to use a high interrogating-neutron-source intensity to override this background.

It is evident from Table II that the active data for the two different neutron interrogating spectra essentially agree, but for some cans differ significantly from the "by difference" Pu content assigned at the processing facility. To provide a direct comparison between chemical and delayed neutron assay results, each of these six cans is scheduled to be recovered chemically at LASL's DP Site. The present active assay results (Table II) are compared with passive assay results on the same samples in a subsequent section of this report.

The one-gallon can assay system just described will be used extensively with the MONAL at Rocky Flats. The inventory of gallon can standards at Rocky Flats includes a wide range of production scrap materials with known Pu loadings varying from a few grams per can to hundreds of grams per can. The planned program of measurements on these standards should enable a thorough evaluation of the accuracy and precision attainable using this active interrogation system.

The concept of an active assay system involving neutron interrogation within a graphite moderator-reflector assembly is applicable not only to gallon cans, but also to a wide range of container sizes, matrix materials and fissile material loadings (^{233}U and ^{235}U as well as Pu). This type of active assay system will undoubtedly find widespread application in industrial and field use.

Both the active neutron interrogation and passive neutron counting of scrap contained in one-gallon (paint) cans will involve inserting a can containing an unknown (and perhaps large) amount of plutonium dispersed in an unknown (and perhaps hydrogenous) matrix material into a large, reflecting counting geometry. A series of computer calculations has been undertaken to investigate the possibility that sufficient plutonium could be contained in one of these cans to produce a critical system when the can is placed in the counter, as well as to provide a basis for the establishment of Standard Operating Procedures (SOP) for these counting geometries.

The calculations were performed using the discrete-ordinates code, DTK-IV, and the Hansen-Roach⁽⁵⁾ 16-group cross sections. These cross sections have been used extensively by Stratton⁽⁶⁾ in criticality calculations which agree well with available experimental data. As a check on present calculational procedures, the critical radius of three bare spherical systems of ^{239}Pu (α -phase) and water having H/ ^{239}Pu ratios of 0, 3, and 2000 were calculated and compared with Stratton's results. The excellent agreement obtained gives added confidence in the present techniques used for the criticality analysis.

The parameter of interest in these calculations is the multiplication factor, k . In all cases, it was desired that a calculation be as accurate as possible, but that any necessary approximations be chosen to produce conservative results (k larger

⁵ Gordon E. Hansen and William H. Roach, Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies, Los Alamos Report, LAMS-2543 (1961).

⁶ W. R. Stratton, Criticality Data and Factors Affecting Criticality of Single Homogeneous Units, Los Alamos Report LA-3612 (1967).

than the actual value). For this reason, a phase plutonium metal ($\rho = 19.6 \text{ g/cm}^3$) was homogeneously dispersed throughout the matrix material in the one-gallon container. Both the choice of the higher-density-phase plutonium and the choice of plutonium metal rather than the oxide (PuO_2) permits the largest number of moderating atoms for any given mass of plutonium in a fixed volume. A hydrogen-moderated system is more reactive (i. e., a higher value of k) for a given mass of plutonium than a system containing any other type of moderator. Polyethylene (CH_2) was chosen as the matrix material for the criticality study because it has a higher nuclear density of hydrogen than water and because the carbon in polyethylene is a more effective scattering material than is the oxygen in water.

For calculational purposes, the one-gallon can was modelled as a sphere having a volume of one gallon (3.785 liters). The spherical geometry is more reactive than the actual cylindrical can for a given volume,⁽⁷⁾ but only slightly so since the one-gallon can has a height/diameter ratio of about one. The moderating assembly for the active-interrogation of the one-gallon can consists of a large block of carbon having a steel-lined cylindrical cavity into which the sample can is placed for irradiation and delayed-neutron counting. The steel liner and the carbon moderator are separated by 16-mil cadmium. The cavity is closed at both ends by steel-and-cadmium-faced carbon plugs. This moderating assembly was modelled in the calculations as spherical shells of the same thickness as the corresponding material in the actual assembly. The 4π passive neutron counter consists of a ring of (18) six-atmosphere, one-inch-diameter, twenty-four-inch-long ^3He proportional counters imbedded in a cylindrical polyethylene moderator.

⁷ H. C. Paxton, J. T. Thomas, Dixon Callihan and E. B. Johnson, Critical Dimensions of Systems Containing ^{235}U , ^{239}Pu , and ^{233}U , TID-7028 (1964), p. 3.

The sample cavity is lined with cadmium and plugged at both ends with aluminum reflectors. For calculational purposes, the sample was replaced by a mixture of plutonium plus matrix material having densities appropriate to a one-gallon sphere. Because an infinite-cylinder calculation was expected to be over-conservative, a buckling length was used. This height was determined by replacing the aluminum plugs with water (a more effective reflector than aluminum) and using a value for the reflector-savings for water inferred from data given by Stratton.⁽⁸⁾

Calculations were made for a range of ^{239}Pu mass from 100 grams to 6 kilograms. Figure 5 illustrates the change in the multiplication factor caused by the effective plutonium cross-sections and differing ^{240}Pu content for both the reflected (i. e., can in the active interrogation assembly) and the unreflected one-gallon spherical can for a polyethylene ($\rho = 0.92 \text{ g/cm}^3$) matrix. The effective cross sections⁽⁹⁾ take into account the features of resonance behavior in fissionable materials by considering the ratio of the macroscopic scattering cross section of the moderating atoms to the number of fissionable atoms present. The results of using infinitely-dilute plutonium cross sections in the calculations are shown by the two dashed curves in Fig. 5. The use of effective cross sections increases the value of k for any given mass of plutonium because the fission resonances tend to have a greater width than the capture resonances. It is clear from Fig. 5 that ^{240}Pu is a very effective poison in the system because of its high absorption cross section and high capture-to-fission ratio. Calculations in which 0.5% ^{241}Pu was included (isolated points in Fig. 5) indicate that ^{241}Pu has a negligible effect on k .

The results of the calculations using effective cross sections and assuming 6% ^{240}Pu content

⁸ Stratton, *ibid.*, p. 34.

⁹ George I. Bell, Theory of Effective Cross Sections, Los Alamos Report LA-2322 (1959).

Fig. 5. Effects on the multiplication factor in a one-gallon can containing various amounts of plutonium in a polyethylene matrix ($\rho = 0.92$) caused by the ^{240}Pu content and the cross sections used. The solid lines were calculated using effective cross sections for the fissionable isotopes. Dashed lines use infinitely dilute cross sections and no ^{240}Pu .

are shown in Fig. 6 for two different densities of the polyethylene matrix-- $\rho(\text{CH}_2) = 0.92$ and $\rho(\text{CH}_2) = 0.20$ --for both the unreflected gallon can and for the can in the active interrogation assembly. The corresponding weights of the cans are given in Table III. On the basis of these results, any can having $\sim 6\%$ ^{240}Pu content and weighing less than ~ 15 lbs. can be safely placed in the active interrogation counting geometry with no possibility of k exceeding 0.9. Cans weighing more than ~ 15 lbs. will very likely have to undergo additional screening before they can be put into the counting geometry.

The count rate in the 4π passive-neutron

Fig. 6. Criticality evaluation of a one-gallon can inside (reflected) and outside (unreflected) of the active interrogation counter for two different polyethylene matrix densities. Six percent ^{240}Pu and effective cross sections were assumed.

TABLE III

WEIGHT OF ONE-GALLON CAN CONTAINING ^{239}Pu AND FULL DENSITY POLYETHYLENE

^{239}Pu Mass (Kg)	Can Weight (lb.)	
	$\rho(\text{CH}_2) = 0.92$	$\rho(\text{CH}_2) = 0.20$
6	20.3	14.8
5	18.2	12.6
4	16.1	10.4
3	14.0	8.2
2	11.9	6.0
1	9.8	3.9
0.5	8.7	2.8
0.2	8.1	2.1
0.1	7.9	1.9

Fig. 7. Relative count rate of an unreflected one-gallon can containing plutonium in various matrix materials as a function of the multiplication factor of the can when placed inside the 4π passive neutron counter.

counter is limited to the rate produced by ~ 500 grams of PuO_2 (6% ^{240}Pu). This rate is divided equally between spontaneous fission neutrons from ^{240}Pu and neutrons from the (α, n) reaction with oxygen. Since this counter is count-rate limited, the counting rate of 500 grams of PuO_2 outside the counter can be used as a screening criterion. Figure 7 shows the relative count-rate of unreflected one-gallon cans as a function of the multiplication factor of the same can when placed in the 4π counter. The count rate was computed from the relation^(1c)

$$\text{CR} \sim \frac{\text{SL}}{(1-k)},$$

where S is number of ^{240}Pu atoms present (i.e., the source strength), and L and k are the episcadmium leakage and multiplication factor respectively of the unreflected can. The count-rate produced by 500 grams of PuO_2 (6% ^{240}Pu) was

also calculated (assuming spontaneous fission rate = (α, n) rate) and is shown by the horizontal line in Fig. 7.

Three densities of polyethylene matrix were assumed— $\rho = 0.92$, $\rho = 0.80$, and $\rho = 0.20$ —and various masses of ^{239}Pu (6% ^{240}Pu) were added. The results (solid curves in Fig. 7) indicate that any can having a count rate less than that of 500 grams of PuO_2 (6% ^{240}Pu) will have a multiplication factor of less than 0.9 when placed in the 4π counter. Even when the ^{240}Pu content is reduced to 1.5% (dashed curve in Fig. 7), the multiplication factor will still be less than 0.9. This limiting count-rate corresponds to ~ 1 kg Pu metal in polyethylene of density $\rho = 0.2$ and ~ 500 g of Pu metal in full density ($\rho = 0.92$) polyethylene.

Several calculations were performed for configurations that might conceivably produce a low count rate when unreflected but yet have a high k-value when placed in the counter. These consisted of a boron ($1/v$ absorber) or a tantalum (resonance absorber) liner in the can completely surrounding the Pu- CH_2 mixture. The results of these calculations are shown as isolated points in Fig. 7, and are seen to lie on the "safe" (to the left) side of the full-density CH_2 -moderated curve.

One case, consisting of 6 kg ^{239}Pu (6% ^{240}Pu) in 6 kg of $\text{MgO} + \text{SiO}_2$ was run because such matrices are expected in practical assay problems. As indicated in Fig. 7, the calculation shows this to be an entirely safe configuration, as anticipated.

Fifty-Five-Gallon Barrel Assay

M. M. Thorpe and T. D. Reilly

With the completion of the MONAL and its associated passive and active assay systems, research on the assay of 55-gallon scrap barrels has begun in the experimental cavity of MONAL. The

¹⁰ Harold Etherington (Ed.), Nuclear Engineering Handbook, p. 8-22.

add-a-gram apparatus⁽¹¹⁾ was installed and additional lead was added to the D, T source tailoring assembly to reduce the delayed neutron contribution from the $^{17}\text{O}(n, p)^{17}\text{N}$ reaction.⁽¹²⁾ The add-a-gram method was developed to take account of the variation in delayed neutron yield versus incident neutron energy in the region from ~ 3 MeV to 14 MeV. In the MONAL cavity the average energy of the neutrons inducing fission was found to be well below 3 MeV, i. e., below the region of varying delayed neutron yield.⁽¹³⁾ Consequently, it has proved possible to replace the mechanically complex add-a-gram apparatus with a simpler assay procedure based on fission counter normalization.

To study the response of ^{235}U in different matrices, several 55-gallon barrels of representative mock scrap were prepared. These were fitted with three vertical tubes located at different radii in the barrel so as to permit placement of fissile samples at different representative positions throughout the matrix. The matrix materials selected for these barrels were: rags and paper wipes wrapped in plastic bags and weighing 25 lbs. (see Fig. 8); polyethylene chips weighing 50 lbs. (see Fig. 9); polyethylene pipe segments weighing 100 lbs. (see Fig. 10); graphite, broken rods and chunks weighing 500 lbs. (see Fig. 11); and steel pipe segments (partially full barrel) weighing 750 lbs. (see Fig. 12).

The experimental arrangement in the cavity is shown schematically in Fig. 13. The delayed neutron counter used is the "thin" slab detector.⁽¹⁴⁾ A ^{238}U fission counter is used to monitor the source intensity of the neutron generator. The

Fig. 8. Fifty-five gallon barrel containing matrix of rags and paper wipes wrapped in plastic bags (wt., 25 lbs.). The three vertical tubes permit fissile material placement at different representative positions throughout the matrix.

^{235}U fission counter is used to normalize ^{235}U delayed neutron response from the barrel.

For each sample position in the barrel, there were three irradiations made at different barrel elevations (barrel center at target level, and barrel 9" above and 9" below target). This provides some spatial averaging in the vertical direction while the three tubes located at different radii in the barrel (cf. Figs. 8 through 13) provide radial-

Fig. 9. Fifty-five gallon barrel containing matrix of polyethylene chips wrapped in plastic bags (wt., 50 lbs.). The three vertical tubes permit fissile material placement at different representative positions throughout the matrix.

¹¹ M. M. Thorpe, T. D. Reilly and D. B. Smith, LA-4368-MS, pp. 14-15 (1969).

¹² *ibid.*, p. 15.

¹³ M. S. Krick and A. E. Evans, LA-4368-MS, pp. 11-12 (1969).

¹⁴ H. O. Menlove and R. H. Augustson, LA-4368-MS, pp. 13-14 (1969).

Fig. 10. Fifty-five gallon barrel containing matrix of polyethylene pipe segments (wt., 100 lbs.). The three vertical tubes permit fissile material placement at different representative positions throughout the matrix.

Fig. 12. Fifty-five gallon barrel containing matrix of steel pipe segments (partially full barrel, wt., 750 lbs.). The three vertical tubes permit fissile material placement at different representative positions throughout the matrix.

dependence information. To provide averaging in the azimuthal direction the barrel was rotated at approximately 6 rpm. The neutron generator was pulsed every 200 msec with a duty cycle consisting of an 80 msec irradiation, a 20 msec "wait time" to allow the counters to recover from the interrogating neutron pulse, and a 100 msec counting period. One or more 100-sec counts were obtained at each position.

A study was made of the variation in delayed neutron response throughout the barrel. For this purpose a sample of ^{235}U was placed in nine representative positions throughout the barrel volume (i. e., at three radii and three heights). Two different samples were used; a "thick" sample consisting of 18.3 grams of ^{235}U foil tightly coiled in a 1" brass cylinder and a "thin" sample consisting of 0.8 grams of ^{235}U foil of approximate dimensions 1.5" x 1" x .001". The specific responses of

Fig. 11. Fifty-five gallon barrel containing matrix of graphite, broken rods and chunks (wt., 500 lbs.). The three vertical tubes permit fissile material placement at different representative positions throughout the matrix.

Fig. 13. Schematic of experimental arrangement for barrel assay in MONAL cavity.

TABLE IV

SPATIAL VARIATION OF NORMALIZED DELAYED NEUTRON
RESPONSE IN REPRESENTATIVE 55-GALLON SCRAP BARRELS

EMPTY BARREL, THICK SAMPLE

	Reflected			Unreflected		
	Center	1/2 Rad.	Edge	Center	1/2 Rad.	Edge
Top ^a	.74	.77	.86	.97	.79	1.00
Center ^a	1.00	1.04	1.11	1.00	1.05	1.25
Bottom ^a	.80	.76	.73	.84	.86	.78

STEEL BARREL, THICK SAMPLE

	Reflected			Unreflected		
	Center	1/2 Rad.	Edge	Center	1/2 Rad.	Edge
Top ^b	.78	.90	.86	1.16	1.37	1.68
Center ^b	1.00	1.04	1.51	1.00	1.16	1.32
Bottom ^b	.56	.58	.90	.79	.77	.91

POLY BARREL (100 LBS.), REFLECTED

	Thick Sample			Thin Sample		
	Center	1/2 Rad.	Edge	Center	1/2 Rad.	Edge
Top ^c	.37	.31	.19	.72	.82	.33
Center ^c	1.00	.99	.52	1.00	.93	.36
Bottom ^c	.21	.40	.36	.24	.29	.17

GRAPHITE BARREL (500 LBS.)

	Thick Sample, Unreflected			Thin Sample, Reflected		
	Center	1/2 Rad.	Edge	Center	1/2 Rad.	Edge
Top	.46	.41	.29	.57	.50	.31
Center	1.00	.81	.41	1.00	.87	.50
Bottom	.56	.47	.26	.59	.48	.31

RAG BARREL (25 LBS.)

POLY BARREL (50 LBS.)

	Thick Sample, Reflected			Thin Sample, Reflected		
	Center	1/2 Rad.	Edge	Center	1/2 Rad.	Edge
Top	.46	.48	.45	.56	.49	.18
Center	1.00	.92	.69	1.00	.84	.27
Bottom	.65	.58	.61	.60	.50	.20

^a The ²³⁵U sample was centered in the barrel and 6 inches from the top and bottom of the barrel.

^b This barrel was only partially full. Top, bottom, and center refer to the matrix rather than the barrel.

^c For thin sample, "top" is 6 inches below top of barrel and "bottom" is the sample actually resting on bottom of barrel.

these samples vary due to self-shielding effects. The results of this spatial response study are presented in Table IV, where the nine measured values in each barrel have been normalized to the response at the center of that barrel. The reflector referred to in Table IV consists of two sheets of polyethylene, 2 ft. x 4 ft. x 1/2", placed near the surface of the barrel, as shown schematically in Fig. 13.

In order to compare the effects of various matrix materials, a total (volume-averaged) response value for each barrel was calculated from the spatial response data for that barrel. This volume averaged response, presented in columns 3 and 4 of Table V shows that the fission counter

method of normalization is quite effective, as predicted by previous calculations.⁽¹⁵⁾ i.e., over a wide range of materials, and consequent wide (order of magnitude) variations in delayed neutron response (e.g., see Fig. 14, LA-4029-MS), the fission counter method of normalization is used very effectively to "flatten" most of the dependence of delayed neutron response on matrix material characteristics. The fifth column of Table V indicates how the fission rate observed at the edge of the barrel is influenced by the matrix material. The sensitivity for ²³⁵U detection (column 2) is taken as that number of grams of ²³⁵U located at the center of the barrel which will give a delayed neutron response equal to background.

TABLE V
MATRIX MATERIAL DEPENDENCE IN
REPRESENTATIVE 55-GALLON SCRAP BARRELS

WITHOUT REFLECTOR				
Matrix	Detection Sensitivity ^a (Grams ²³⁵ U)	Volume Averaged Response/gm Normalized to Empty Barrel (Thin Sample)		²³⁵ U/ ²³⁸ U Fission Ratio Normalized to Empty Barrel
		Thick Sample	Thin Sample ^b	
Empty	5.5	.89	1.00 ^b	1.00
Steel	6.3	.96	1.45 ^b	.99
Graphite	0.53	1.81	3.74 ^b	2.1
WITH REFLECTOR				
Empty	2.5	.55	1.00 ^b	1.00
Steel	3.7	.46	---	.97
Graphite	0.35	---	1.68	2.32
Rag	1.4	.55	---	1.85
Poly (50 lbs)	.05	---	2.41	6.89
Poly (100 lbs)	.05	.38	1.96	8.83

^a See text for definition of detection sensitivity.

^b Volume average actually measured with thick sample; a measured correction for the self-shielding has been applied in this case to obtain thin sample response.

¹⁵ LA-4029-MS, pp. 14-16 (1968)

The data presented herein marks the beginning of an experimental program of research on 55-gallon barrel assay techniques. Further work will involve optimization of reflector material and configuration; studies of the differences in response between plutonium and uranium; studies of other matrix materials, (including cases intermediate between the representative extreme cases chosen here); and further studies of self-shielding effects.

^{252}Cf Spectrum-Tailoring

D. B. Smith

The calculational investigation of a spectrum-tailoring assembly for the 1 mg ^{252}Cf neutron source⁽¹⁶⁾ continued during this reporting period. Of particular interest in the present calculations is the enhancement of thermal neutron flux within the assembly for delayed-neutron assay of small samples (or alternatively for activation analysis). One-dimensional DTF-IV calculations were carried out for various combinations of moderating materials. The results of these calculations are given in Fig. 14, which shows the thermal neutron flux as a function of radial position within the tailoring assembly. Two of the moderator combinations are especially interesting. Fifteen centimeters of Be followed by polyethylene produces a relatively high thermal flux near the center of the moderator which changes by only a few percent per centimeter over the entire Be region. This permits much flexibility in the choice of location of sample reentrant hole. The moderator should be quite compact and the relatively flat flux should result in quite uniform irradiation of small samples. Pure polyethylene moderator produces a thermal neutron flux near the source which is a factor of three or four times that produced by Be + CH_2 . However, in order to take advantage of this increased flux, the sample reentrant hole would have to be located

very close to the ^{252}Cf source in a position which could be mechanically inconvenient. In addition, the flux drops by $\sim 15\%$ per centimeter in this region, and even small samples might require rotation to obtain a near-uniform irradiation.

Fig. 14. Thermal neutron flux produced by ^{252}Cf spontaneous fission source in various spectrum-tailoring configurations.

¹⁶ D. B. Smith and H. O. Menlove, LA-4368-MS, p. 4 (1969)

Precision Gamma-Ray Energy Measurements

L. V. East

A Ge(Li) gamma-ray spectrometer is being utilized to make precision energy measurements of gamma-rays from high-Z radioactive isotopes and certain fission products. Gamma-ray energies from some useful calibration sources will also be determined. Energy determinations are made relative to a series of "standard" gamma rays emitted from sources of ^{241}Am , ^{109}Cd , ^{203}Hg , ^{139}Ce , ^{198}Au , ^{57}Co , ^{75}Se , ^{22}Na , ^{137}Cs , ^{54}Mn , ^{88}Y , and ^{60}Co .

Precise energy values of the gamma rays emitted by these sources were taken from recent publications.^(17, 18) Energy uncertainties of the standards range from < 10 eV for low energy gamma rays up to about 40 eV for ~ 1 MeV gamma rays.

The locations of gamma-ray photopeaks in a pulse height spectrum are obtained by a least-squares computer code⁽¹⁹⁾ that fits a modified Gaussian function to each photopeak of interest. Using this technique, photopeak locations can be determined with a precision of the order of 0.02 - 0.05 channel in most cases for well-resolved peaks. This code is presently running on the LASL CDC-6600; it is hoped, however, that a modified version of the code will be running on the N-6 PDP-9/L computer in the near future.

Energy calibration of the pulse height analyzer is accomplished by making a least-squares fit of the photopeak channel positions of the standards, as determined above, versus the photopeak

energy. A fourth-order polynomial fitting function has been found adequate to compensate for analyzer nonlinearities. The values of the fitted parameters are then used to calculate the energies corresponding to "unknown" photopeak. Whenever possible, a spectrum of the unknown gamma rays is obtained simultaneously with a series of standards in order to minimize errors due to analyzer gain shifts. For very complicated spectra, this procedure is not always possible. For these cases, separate runs are made for the unknown and standards, with at least two photopeaks common to the separate runs, one near the low end and one near the high end of the pulse height scale, so that corrections may be made for any gain shifts occurring between runs. The precision obtainable by this method is indicated in Table VI, which compares the calculated energies versus the "known" energies of a series of standard sources. The energy scale

TABLE VI

COMPARISON OF CALCULATED AND
"STANDARD" GAMMA-RAY ENERGIES OF
A SERIES OF CALIBRATION SOURCES

Source	Calibration Energy, keV	Calculated Energy, keV	Diff., eV
^{241}Am	59.537 ± 0.001	59.5364	-0.6
^{109}Cd	88.036 ± 0.008	88.0362	0.2
^{75}Se	96.732 ± 0.007	96.7338	1.8
	121.113 ± 0.010	121.1127	-0.3
	135.998 ± 0.010	135.9961	-1.9
	198.600 ± 0.020	198.6021	2.1
	264.651 ± 0.015	264.6488	-2.2
^{57}Co	279.525 ± 0.012	279.5291	4.1
^{203}Hg	122.061 ± 0.010	122.0614	0.4
	136.471 ± 0.010	136.4701	-0.9
	279.188 ± 0.006	279.1853	-2.7

Calculated values based on fourth-degree least-squares fit to calibration energies.

¹⁷ J. B. Marion, Nuclear Data A4, 301 (1968).

¹⁸ R. C. Greenwood, R. G. Helmer, and R. J. Gehrke, Nucl. Inst. and Meth. 77, 141 (1970).

¹⁹ H. M. Murphy, Jr., "PPA, A Computer Program for Photopeak Analysis," AFWL-TR-65-111, March 1966. The original code was modified to include weighting of the least-squares fit and a calculation of the expected variances of the fitted parameters.

TABLE VII

GAMMA-RAY TRANSITION ENERGIES

Decaying Nuclide	Gamma-Ray Energy, keV	
	Present Measurement	Previous Measurements ^a
²⁴¹ Pu	148.569 ± 0.020	145
²³⁹ Pu	129.297 ± 0.015	129.27 ± 0.03 ^b
	144.173 ± 0.028	144.18 ± 0.08
	146.12 ± 0.05	146.07 ± 0.08
	171.378 ± 0.030	171.37 ± 0.08
	203.552 ± 0.020	203.55 ± 0.04
	255.379 ± 0.035	255.38 ± 0.07
	297.44 ± 0.04	297.48 ± 0.07
²³⁷ U	164.596 ± 0.025	164.61
	208.012 ± 0.018	208.005 ± 0.023 ^c
	267.545 ± 0.030	267.54
²³⁵ U	109.0 ± 0.1	110 ^d
	143.766 ± 0.022	143 ± 2
	163.353 ± 0.020	163
	182.58 ± 0.06	180
	185.719 ± 0.018	185 ± 2
	202.113 ± 0.022	201
	205.316 ± 0.018	204
²³⁴ U	53.24 ± 0.05	53.3
	120.888 ± 0.021	120.9
²³¹ Th	84.229 ± 0.017	84.17
⁹⁴ Nb	702.630 ± 0.030	702.5 ± 0.1 ^e
	871.104 ± 0.035	870.8 ± 0.1

^a Except as noted, gamma-ray energies were taken from C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes, Sixth Edition, John Wiley and Sons, Inc. (1967).

^b ²³⁹Pu gamma-ray energies from J. E. Cline, Nuclear Physics A106 (1968) 481.

^c 208 keV transition in ²³⁷Np from ²⁴¹Am decay reported by T. Yamazaki and J. M. Hollander, Nuclear Physics 84 (1966) 505.

^d ²³⁵U gamma-ray energies from unpublished data of R. C. Pilger, F. S. Stephens, F. Asaro and I. Perlman reported by E. K. Hyde, I. Perlman, and G. T. Seaborg, The Nuclear Properties of the Heavy Elements, Vol. II, Prentice Hall, Inc. (1964) 730.

^e R. A. Kuebbing and K. J. Casper, Nucl. Phys. A98, 75 (1967).

was established as described above using these same standards. In all cases, the calculated energies are well within the uncertainties of the calibration energies.

A Ge(Li) detector with an active volume of 30 cm^3 is being used for these measurements. This detector has very good resolution for its size; typical FWHM resolutions using a room temperature preamplifier are 1.20 keV at 122 keV and 2.0 keV at 1.33 MeV. The pulse height spectra were obtained using N-6's new computer-based system de-

scribed later in this report. A typical gamma-ray spectrum obtained from a ^{235}U sample, along with the measured gamma-ray energies, is shown in Figure 15.

Table VII lists the measured gamma-ray energies and measurement uncertainties that have been obtained to date. Gamma rays from the Pu and U isotopes are of interest for passive assay of fissionable material. ^{94}Nb , with its long half life of 2×10^4 years, is a convenient calibration source for many purposes.

Fig. 15. Gamma-ray spectrum from a 93% enriched ^{235}U sample. The measured gamma-ray energies are given in keV.

Gamma-Ray Scanning System for Assay of Fifty-Five Gallon Drums

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

The passive gamma-ray assay systems under development for MONAL include a unit for scanning and assaying of 55-gallon, or smaller, drums. This passive assay unit should supplement and complement the active interrogation techniques available in MONAL, particularly for barrels containing plutonium-bearing homogeneous waste.

Design criteria for this barrel scanning unit have included:

1. Provision for rapid and accurate quantitative assay. In the case of homogeneous wastes, a quantitative assay should involve; 1) integral counts taken at only a few representative vertical positions in the barrel and; 2) subsequent corrections for backgrounds and gamma-ray attenuation.

2. Provision for the detection of lumps of high-Z materials which could cause a serious underestimate of the plutonium content. It is essential to quickly recognize drums not amenable to assay by gamma scanning methods.
3. Optional capability for determination of the spatial distribution of plutonium in the drum.
4. Mechanical compatibility with the MONAL interrogation chamber and barrel handling system. Space limitations in the chamber constrain the possible configurations of the passive scanning unit. The gamma-ray detectors should be mechanically mounted so as to permit rapid setup and removal, since these detectors must not be present in the interrogation chamber when it is being used for active assay measurements.

Rotating the barrel while counting is a major factor in reducing the dependence of response on radial distribution of plutonium. With a detector located at one barrel radius from the periphery of the barrel, the radial variation in response is about $\pm 15\%$ for barrels containing homogeneous material with densities less than about 0.1 g/cm^3 . If the detector is located closer to the barrel, (e.g., as constrained by the MONAL irradiation chamber) or if the density of the matrix material in the barrel is greater than 0.1 g/cm^3 , the radial variation increases, the observed response becoming larger for material near the periphery of the barrel.

For the design of the present system, experimental studies of spatial response were performed for barrel rotation combined with horizontal and vertical collimation. Figure 16 shows the two-dimensional case, i.e., the Pu and the detector are coplanar and horizontal collimation is utilized. The effect of the horizontal collimation is to selectively reduce the response of material located near

Fig. 16. Two-dimensional schematic for gamma-ray assay of Pu in cylindrical containers. L_1 , L_2 , and D are varied to achieve flat response within the plane.

the periphery of the barrel. The collimation angle and displacement D can be adjusted to yield an almost flat radial response for matrices which attenuate the "385-keV" ^{239}Pu gamma rays by as much as a factor of 25 across the container diameter. If the matrix material is reasonably uniform, an assay (in two dimensions) would then consist of an attenuation measurement, an integral count with the collimation angle and displacement adjusted to yield flat response for this attenuation, and finally correction of the integral count for the measured attenuation to give the desired plutonium assay value for the sample.

By positioning the detector close to the barrel and using horizontal collimation, one can also obtain the radial distribution of plutonium from a measurement of the counting rate vs angle of rotation. The multiscaling unit of a multichannel analyzer offers a convenient means for collecting and displaying this type of data. Figure 17 shows the counting rate of a Pu sample as a function of the angle of rotation for three different radial positions of the Pu sample: A-at the center of the barrel, B-at one-half the barrel radius, C-at the barrel periphery. The widths of the peaks in the data are a function of the radial position of the sample and hence provide information on the radial distribution

Fig. 17. Multiscale records of response from a "point" Pu sample in rotating barrel: (A) sample at center of barrel, (B) sample at mid-point of barrel radius, (C) sample at outer edge of barrel.

of plutonium within the barrel.

The three-dimensional barrel scanning system now under construction consists of eight 2-in. diam. by 2-in. thick NaI crystals placed in a vertical plane and spaced so as to view the entire barrel height. The output of each crystal is passed through a single-channel analyzer which brackets the "385-keV" complex. For spatial distribution and transmission measurements, the eight detector outputs can be multiscaled separately and simultaneously with the multichannel analyzer in the MONAL. Eight 100 μ Ci ^{22}Na sources, placed diametrically opposite the gamma detectors will be utilized for the external transmission scan. The detector outputs may also be added to obtain directly and rapidly a single count for the assay of a barrel.

A full scale prototype test unit for gamma-ray assay of barrels has been constructed and

tested. The eight crystals are spaced vertically six inches apart on centers and their faces are approximately five inches from the barrel periphery. The crystals view the barrel through a 2-in. vertical collimator which consists of parallel lead plates extending four inches forward of each crystal and four inches on each side of the crystal axis. Variable horizontal collimation is also provided.

With this prototype geometry, it was necessary to count the barrel at two positions—one displaced vertically from the other by 3" (half the distance between crystals)—in order to achieve a flat response in three dimensions. For convenient identity, this technique will be referred to hereafter as the "double position scan" method. For the 0.1 g/cm³ polyethylene test barrel, the variation in response over the volume is less than $\pm 10\%$. For this case no horizontal collimation was required, but for higher densities such collimation will be needed to achieve flat response. With a 150-keV single channel analyzer bracketing the "385-keV" complex, the count rate of the system is $\sim 25 \text{ sec}^{-1}\text{-g}^{-1}$ and the background rate in the well-shielded experimental location used is approximately 16 sec^{-1} .

Gamma-Ray Scan Computations

T. D. Reilly

A computer program has been written which predicts the response of either a single gamma-ray detector or vertical array of detectors to passive gamma rays originating uniformly or from any given point within a cylinder. It is assumed that the crystals view the cylinder through rectangular collimators whose vertical and horizontal dimensions can be varied independently. The cylinder contains a uniform matrix of variable attenuation, and, to help achieve a flat spatial response, the cylinder is rotated in front of the detectors. The following is a summary of the results obtained with this computer program.

The first case considered was a single detec-

tor viewing a one-gallon can, which is essentially a two-dimensional geometry if the detector is located approximately 6.5" (one can diameter) from the periphery of the can. Through the use of horizontal collimation, it is possible to obtain a response which is independent of position in a horizontal plane to better than $\pm 5\%$. This occurs because of the averaging effect of barrel rotation and because the collimator selectively reduces the response of sources located near the edge of the barrel (see Fig. 16). For a given attenuation there is a collimation angle that optimized ("flattens") the spatial response in the barrel; the higher attenuation requires narrower collimation for greater suppression of peripheral response, thereby compensating for the more-highly-attenuated central response. Table VIII gives some examples of the optimum collimation angles as a function of matrix attenuation for a one-gallon can viewed by a 2" crystal.

For a 55-gal drum and the eight-crystal array (definitely a three-dimensional geometry) the collimation requirements change. It was assumed that the eight crystals are positioned 6" on centers in a vertical array. Two possible scanning procedures are suggested by the code. For a "single

position scan" procedure in which the barrel is not moved vertically, no vertical collimation is utilized but there is horizontal collimation which may be adjusted for the appropriate attenuation of the material in the barrel. The optimum spatial response for a 0.1 density CH_2 -loaded barrel is obtained with no horizontal collimation. At higher attenuations it becomes necessary to add collimation in this direction to compensate for the high peripheral response. The response for the 0.1 density barrel varies only $\pm 23\%$ over the entire volume, and this is primarily due to end (i. e., top and bottom) effects; over most of the volume it is much flatter than this. The response of a source uniformly distributed throughout the barrel is identical to that of a point source at the barrel center; this can simplify calibration of the system.

In the "double position scan" method (as studied with the prototype gamma scan unit for 55-gallon drums, cf. previous section) the maximum vertical collimation (about 4" of Pb beyond the crystal) is used along with variable horizontal collimation. In this method the barrel is moved into position beside the detector array and is counted; then the barrel is displaced 3" upwards and counted again; this second scan enables the

TABLE VIII

GAMMA SCAN CHARACTERISTICS (ONE-GALLON CANS)^a

$\mu(\text{cm}^{-1})$	$e^{-\mu \cdot \text{radius}}$	$e^{-\mu \cdot \text{diam}^b}$	Collimation		% Variation of Response	Volume Average Response Relative To Center Response
			Angle	L2(in.)		
.03	.77	.61	73°	2.72	$\pm 2\%$	1.00
.06	.60	.37	64°	3.20	$\pm 2\%$	1.00
.10	.44	.19	48°	4.45	$\pm 4\%$	1.00
.13	.34	.12	41°	5.40	$\pm 5\%$	1.01
.16	.26	.07	35°	6.43	$\pm 6\%$	1.02

^a For 2" crystal centered vertically and located 6.5" out from periphery of gallon can.

^b Diameter of gallon can = 16.5 cm.

gamma detectors to "look" at those regions which were shielded in the first scan by the vertical collimators. The results of these two scans are averaged to provide the total barrel response. Here again no horizontal collimation is required for a 0.1 density barrel. After averaging the two scans, the response throughout the barrel is flat to $\pm 3\%$. With the same geometry a 0.2 density barrel yields a response which is flat to $\pm 12\%$. This, and higher attenuating cases, can be improved with the appropriate horizontal collimation.

Gamma-Scan of Medium-Density ^{239}Pu Scrap Cans

L. V. East and J. L. Parker

Six one-gallon cans containing plutonium-contaminated slag and broken crucibles were obtained from the LASL plutonium fabrication facility (DP Site) for assay of ^{239}Pu content. These cans were estimated to contain between 10 g and 20 g of ^{239}Pu in a matrix of MgO and CaF_2 , with an average density of about 2.3 g/cm^3 . Two "standard" cans were also obtained, one containing 20.0 g

and the other 200.0 g of PuO_2 uniformly mixed in the same type of matrix material.

These cans were gamma-scanned using a $30 \text{ cm}^3 \text{ Ge(Li)}$ detector. This detector was chosen over NaI(Tl) since more information, such as ^{241}Pu content and the presence of (α, n) reactions, could be obtained due to the inherently better resolution of Ge(Li) detectors (cf. Fig. 18). Also the precise energy resolution of the Ge(Li) detector very effectively separates the uncollided (full energy) ^{239}Pu gamma rays from small-angle Compton-scattered photons which limit the interpretation of NaI data for some matrix materials. The Ge(Li) detector had an energy resolution of 1.45 keV FWHM for 400 keV gamma rays.

The scan arrangement used is shown in Fig. 19. The 414 keV and 375 keV gamma rays from ^{239}Pu were used for assay. Because of the moderately high density of the cans, gamma rays in this energy range are attenuated by a factor of about 25 in traversing the width of a can. For this reason the technique of sample rotation combined

Fig. 18. Comparison of ^{239}Pu gamma-ray spectra obtained with a high resolution Ge(Li) detector and a NaI(Tl) scintillator.

Fig. 19. Gamma-scan set-up used with one-gallon ^{239}Pu scrap containers.

with detector collimation⁽²⁰⁾ was used to "flatten" the detector response to gamma rays emitted from various volume elements of the can. In addition, the 511 keV gamma rays from the ^{22}Na source shown in the figure were used to measure the total gamma-ray attenuation of each can. The 400 keV gamma rays from ^{75}Se would be somewhat better suited for these attenuation measurements; however, the strength of available sources was found to be inadequate. The 0.03 in. Cd absorber in front of the detector was used to reduce the count-rate from the 59.6 keV ^{241}Am gamma rays emitted by the samples.

Detector response flatness for a given density sample is a function of the amount of collimation used and the offset of the detector-collimator center line with respect to the sample center. In order to optimize these parameters for the scrap cans, a one-gallon can containing approximately the same density of $\text{MgO} + \text{CaF}_2$ (no Pu) as the Pu cans was obtained. The 400 keV gamma rays from a small 100 μCi source of ^{75}Se that could be located at various positions within the can were then used to measure detector response. The detector response obtained with the collimator configuration shown in Fig. 19 is presented in Fig. 20 as a func-

Fig. 20. Detector response as a function of ^{75}Se source radial location for one-gallon cans containing different matrix materials. The can containing the CaF_2 - MgO mixture was a mock-up of the slag and crucible ^{239}Pu scrap containers.

tion of the radial position of the ^{75}Se source. Also shown in Fig. 20 are responses for a gallon can containing SiO_2 ("sand") and one containing a mixture of lead shot and carbon pellets. The C + Pb can had a total gamma-ray attenuation of about eight times that of the $\text{MgO} + \text{CaF}_2$ can; the can of sand had a total attenuation about half that of the $\text{MgO} + \text{CaF}_2$. The point spread for the C + Pb can response results from inhomogeneities in the mixture. It is estimated that the response over the entire volumes of the scrap cans was constant to within 15%.

In order to correct for the different gamma-ray attenuations of the various scrap cans, a series of measurements were performed to establish the relationship between the attenuation measured by the external ^{22}Na source and the attenuation of 400 keV gamma-rays originating from within a can. The 100 μCi ^{75}Se source was placed half-way between the center and edge of a series of cans containing matrix materials of varying densities, and the attenuations of the 400 keV line as well as the 511 keV ^{22}Na line were determined. The results of

²⁰ See earlier section on Gamma-Ray Scanning System for description of the collimation-rotation technique.

the measurements are shown in Fig. 21. Using this curve, the assay results were corrected for attenuations differing from the standard can.

It was found that for all of the scrap cans, different ^{22}Na gamma-ray attenuations were obtained through different parts of the can. For this reason, two assay measurements were made, one with the can center located about even with the detector ^{22}Na source center line, and the other with the can center about 1.5 in. above this center line. The time required for one assay was about 18 min.

In each measurement, the 375 and 414 keV ^{239}Pu gamma-ray intensities were determined by subtracting a straight line background and summing the counts in the peaks. The measured ^{239}Pu content of each can (obtained by averaging these two measurements) is given in Table IX, along with the average 511 keV gamma-ray attenuations. The uncertainties shown were obtained from counting statistics and an estimate of the measurement error resulting from inhomogeneities of the can contents. Cans XBL-40-1 and XBL-43 were found to be very inhomogeneous from external source atten-

Fig. 21. Attenuation of external 511 keV and internal 400 keV gamma rays as a function of can density for the assay set-up shown in Fig. 19.

uation measurements. On the other hand, cans G-1, G-2, XBL-40-2 and XBL-44 were found to be fairly uniform. A comparison of these gamma-ray passive assay results with assays by active neutron interrogation and passive neutron counting is given in a later section.⁽²¹⁾ Considering the large

TABLE IX
RESULTS OF GAMMA-SCAN ASSAY OF
ONE-GALLON Pu SCRAP CONTAINERS

Pu Scrap Can No.	Total Wt. (kg)	Average 511 keV γ-Attenuation	Assay of Pu Content (g)
G-1	7.5	25.1	Standard
G-2	7.7	27.2	174.0 ± 10.0 [*]
XBL-40-1	7.2	25.0	18.5 ± 2.5
XBL-40-2	6.7	27.7	15.0 ± 1.5
XBL-42	4.8	15.9	12.6 ± 2.0
XBL-43	7.2	25.5	19.6 ± 2.5
XBL-44	6.0	21.9	14.1 ± 1.5
XBL-45	5.7	25.2	14.7 ± 2.0

^{*}Known content of Can G-2 is 177.4 g Pu (difference of 2% from reported assay).

²¹ See section entitled "4π Coincidence Unit for One-Gallon Cans and Smaller Samples".

gamma-ray attenuations involved and the fact that such attenuations vary both within a given can and from one can to another, the gamma assay results are in quite good agreement with the other (neutron) assay methods used on these ^{239}Pu scrap cans.

In addition to the ^{239}Pu content, the $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratio was determined for each can (unknowns and standards) by measuring the ratio of intensities of the 203.6 keV ^{239}Pu gamma ray and the 208 keV ^{237}U (in equilibrium with ^{241}Pu)

gamma ray. This isotopic ratio was found to be constant to within 8% for all cans. This indicates that the assumption required for passive neutron counting, namely that all cans had the same $^{239}\text{Pu}/^{240}\text{Pu}$ ratio, was probably valid. For Pu scrap containers such as these one-gallon cans, gamma-ray scanning using a Ge(Li) detector in conjunction with passive neutron counting represents a very powerful combination of assay methods.

PASSIVE NEUTRON COUNTING TECHNIQUES AND APPLICATIONS

Assay for Pu by Neutron Coincidence Counting

R. B. Walton

The utility of coincidence counting prompt neutrons from the spontaneous fission of ^{240}Pu for quantitative assay of plutonium has been demonstrated for a wide range of small samples and for fabricated fuels such as FFTF and SEFOR. Recent research has been directed to neutron coincidence systems suitable for assaying plutonium-bearing scrap. Application of the neutron coincidence method to scrap is more complicated because of the influence of unknown matrix materials and because of the wide variations in Pu loading encountered in scrap materials, i.e., from very low Pu concentrations in discards up to several hundred grams Pu/gallon for recoverable scrap. A general purpose 4π coincidence detector which will accommodate various sample sizes ranging up to one-gallon cans has been developed and is described herein. In addition, the current stage of development of a 4π coincidence detector for 55-gallon barrels is reported later in this section.

(α , n) Neutrons

Coincidence counting of ^{240}Pu spontaneous fission neutrons is accomplished with 4π polyethylene moderated assemblies filled with ^3He (or

BF_3) counters.⁽²²⁾ In compounds or mixtures containing plutonium, neutrons can also originate from (α , n) reactions on low-Z materials (e.g., O, Be, F, Li, C). If the chemical composition and Pu isotopic abundances are known and controlled, a gross ("singles") neutron count may suffice for quality control; however in many practical situations these assumptions may not be valid. i.e., the (α , n) neutrons can complicate the ^{240}Pu coincidence method by contributing to the accidental (random) coincidence rate and to the real coincidence rate by multiplication via ^{239}Pu (n, f).

It is well known that (α , n) yields vary widely over the gamut of light elements. Thick-target yields measured with Po- α sources are presented in Table X for the most important light elements (Handbuch der Physik, Vol. 38/2 Amaldi, p. 108).

In Pu-plus-light-element mixtures the (α , n) neutron yield varies with the Pu/light-element atom ratio and with the degree of mixing, the most effective mixing occurring in actual chemical compounds, e.g., PuO_2 or PuF_4 . In the commercially-common case of PuO_2 , the (α , n) yields are typically the order of 100 n/sec-g. For pure PuF_4 in powdered form, the (α , n) neutron yield is approximately 300 times greater than for PuO_2 .

²² See Proceedings of the AEC Symposium on Safeguards R&D, WASH-1147, pp. 113-115, (1969).

TABLE X

 (α, n) YIELDS FOR VARIOUS LIGHT ELEMENTS

Element X	Neutron Yield (Po- α Source) per $10^6 \alpha$'s	Yield Ratio X/Be
Be	80	1.00
B	24	3.0×10^{-1}
F	12	1.5×10^{-1}
Na	1.5	1.9×10^{-2}
Mg	1.4	1.8×10^{-2}
Al	0.74	9.3×10^{-3}
Si	0.16	2.0×10^{-3}
Cl	0.11	1.4×10^{-3}
C	0.11	1.4×10^{-3}
O	0.07	8.8×10^{-4}
N	0.01	1.25×10^{-4}

To utilize the ^{240}Pu coincidence counting method for assay of large amounts of PuO_2 (as in SEFOR reds) or scrap containing light elements having high (α, n) yields, coincidence detectors with short die-away times are required to maximize the contribution of real coincidence events relative to accidental events. This requirement in turn led to the concept of varying the physical die-away time of a ^{240}Pu neutron coincidence detector by varying the amount and geometrical configuration of polyethylene moderator and Cd (or B) neutron absorber. ⁽²³⁾ Thus "short-die-away-time" neutron coincidence detectors have been developed and used very effectively in the Pu assay of SEFOR fuel rods, ^(24, 25) to name one example.

4π Coincidence Unit for One-Gallon Cans and Smaller Samples

H. O. Menlove and R. B. Walton

A versatile 4π -neutron detector has been designed and fabricated for Pu assay applications; it has been used for assay of PuO_2 samples varying in mass from a few milligrams up to several hundred grams. The cylindrical counting well of the detector can accommodate sample sizes up to 8 in. diam. (e.g., one-gallon scrap cans) and can be operated in an open-ended mode to facilitate the measurement of long fuel rods. A photograph of the detector assembly (exploded view for clarity) is shown in Fig. 22, and a schematic diagram of the tube locations and polyethylene dimensions is given in Fig. 23. Removable sheets of 30-mil Cd are located on the inside and outside of the polyethylene cylinders to shorten the neutron die-away time ⁽²⁶⁾ and to give variable efficiency for neutron detection. The moderating material (CH_2 or C) on the outside of the larger polyethylene cylinder acts as a shield for exterior background neutrons when the outer Cd sheath is in place; if however the Cd

Fig. 22. Neutron coincidence detector (exploded view for clarity) for counting a large range of Pu samples. The inner annulus of ^3He neutron counters can be removed for measuring samples up to ~8-in. diam.

²³ *ibid.*, p. 115.

²⁴ LA-4315-MS, pp. 20-22 (1969).

²⁵ NRL Memorandum Report 2107 (1970);

²⁶ LA-4315-MS, pp. 21 (1969).

Fig. 23. Schematic diagram of neutron coincidence detector showing location of ^3He neutron counters and thickness of polyethylene moderator. The exterior fast neutron reflector increases the detection efficiency and also acts as a shield against background neutrons.

sheath is removed, the exterior moderator increases the efficiency of the detector. The 1-in.-diam. counter tubes have an active length of 20 in. and are filled with ^3He gas to 3-atm.-pressure. When both rings of counter tubes are used and the Cd is removed, the absolute efficiency of the detector is $\sim 40\%$. The efficiency is reduced to $\sim 9\%$ when the inner ring of tubes is removed and Cd sheaths are placed on the inside and outside of the larger ring of tubes. For assay applications, the configuration of tubes and Cd is chosen to best fit the assay problem. Figure 24 shows the neutron detector, together with its associated electronics, assembled as a complete assay system. Such a system is being used to assay one-gallon cans of Pu scrap typical of the material generated at the Rocky Flats Pu facility. All electronic components are commercially available with the exception of the variable-gate-width coincidence circuit,⁽²⁷⁾ designed and built at LASL. Aluminum or graphite reflectors serve to plug both ends of the detector

²⁷ L. V. East, M. M. Stephens and J. E. Swansen, LA-4368-MS, p. 16 (1969).

Fig. 24. Experimental set up for measuring Pu in one-gallon scrap cans. The NIM BIN contains all the electronics needed for detector operation and data collection.

and thus reduce neutron leakage from the ends. In this configuration of a single ring of tubes, the neutron die-away-time was measured to be $\sim 11 \mu\text{sec}$; Fig. 25 shows a plot of the fraction of coincidence counts observed as a function of coincidence gate width for this configuration. It can be seen that $\sim 80\%$ of the available coincidence events are counted in a gate width of $25 \mu\text{s}$.

For the assay of scrap materials and larger samples, it is necessary to determine the influence of the matrix materials and sample inhomogene-

Fig. 25. Fraction of coincidence counts detected as a function of coincidence gate width for the neutron detector configuration shown in Fig. 24 (single annulus of ^3He neutron detectors).

ities on the measurements. To measure these matrix effects, several one-gallon cans were filled with scrap materials typical of Pu scrap output. The different materials that were investigated are listed in the first column of Table XI. A small ^{252}Cf source of spontaneous fission neutrons was systematically moved throughout the volume of each can and the measured volume-average counting efficiencies are listed in Table XI for counting both single neutron events as well as coincident neutron events. Both of these efficiencies have been normalized to unity for the case of an empty can; thus the deviation from unity represents the net effect of the added matrix material. It can be seen that all of the materials increase the absolute efficiency by $\sim 5\%$ (on the average), with the exception of hydrogenous materials which can change the efficiency by as much as 20% or more, depending on the hydrogen density. These matrix effects on the counting efficiency are roughly squared for the case of coincidence counting.

When a standard which closely matches the sample is available for an assay application, the influence of the matrix material can be effectively "normalized out", thereby achieving the goal of "matrix material independence". However, when little is known about the assay sample, the matrix and multiplication perturbations can be measured by the new "source addition technique", as follows. In this technique, a small spontaneous fission neutron source such as ^{252}Cf (~ 1 nanogram) or ^{240}Pu (a few grams) is positioned in the detector well, and its count rate is observed. Then the unknown sample is positioned in the detector well directly on top of the fission source. The observed change in the known source counting rate then provides a measure of the perturbation due to the matrix material in the unknown sample. This technique was applied to the materials listed in Table XI and the corrected efficiencies are given in the last column of the table. It was necessary to increase the measured source perturbation by

TABLE XI
MATRIX MATERIAL EFFECTS ON THE PASSIVE NEUTRON
ASSAY OF Pu IN ONE-GALLON SCRAP RECOVERY CANS

Matrix Material	Material Mass (kg)	Relative Singles Counting Efficiency ^(a)	Relative Coincidence Counting Efficiency ^(a)	Corrected Coinc. Efficiency ^(b)
Empty 1-Gal. Can	---	1.00	1.00	1.00
Carbon Pellets	1.89	1.03	1.05	0.97
Metal Parts	3.60	1.04	1.09	1.02
Slag-Crucible	1.80	1.03	1.08	1.01
Concrete	3.24	1.05	1.10	1.02
String Filters	0.60	1.07	1.17	1.05
CH_2 ($\rho=0.065$ g/cc)	0.27	1.06	1.11	1.00
CH_2 ($\rho=0.12$ g/cc)	0.43	1.09	1.19	0.98
CH_2 ($\rho=0.27$ g/cc)	0.97	1.19	1.36	1.04
H_2O ($\rho=1.00$ g/cc)	3.62	0.98	0.98	0.96

(a) Efficiencies normalized to case of an empty can.

(b) The counting rate was corrected using the "source addition technique" described in text.

~ 5% to account for the difference in geometric coupling between the actual source location (bottom of detector well) as compared to a (more exact) central source location in the interior of the can. It can be seen that the corrected coincidence counting efficiencies are all within a few percent of the empty can case.

To help evaluate new nondestructive assay techniques and instrumentation, cans of ash residue spiked with known amounts of PuO_2 were obtained from the Atlantic Richfield Hanford Company, Richland, Washington. These cans were packed into heavy (~ 40 lbs) 6-M containers (~ 6-in.-diam. x 12-in. high) for shipment to Los Alamos. For the passive neutron assay, the cans were never removed from their shipping containers. A suitable standard was produced by placing a metallic disk of Pu inside an empty shipping container. The ^{240}Pu content of the samples ranged from 8% to 22% and the assay results are listed in Table XII together with a comparison of the PuO_2 loading given by Hanford. The Pu isotopic composition given by Hanford was used to convert from g ^{240}Pu to g PuO_2 . The average deviation of the passive neutron assay from the Pu loadings given by Hanford was only 2.0%; it should be emphasized

that this accuracy was obtained without removing Pu samples from their shipping containers.

As another example of passive neutron assay of Pu scrap material, one-gallon cans of contaminated slag and crucible were obtained from the process line of the LASL Pu facility at DP-Site. In addition to these production samples, two standards of MgO containing 20 and 200 g of PuO_2 were produced. The one-gallon can 4π neutron detector was then used to assay the slag and crucible samples using the 20 g PuO_2 standard for calibration. The large (α, n) neutron background from the Mg and F in the slag and crucible samples made neutron coincidence assay very difficult because of the low coincidence-to-accidental count ratio (~ 0.05). The results of the assay are listed in Table XIII together with assay data obtained using other assay techniques previously described in this report. The various nondestructive assay results clearly agree within the measurement uncertainties; it is quite significant that the new results differ in some cases by nearly a factor of two from the "by-difference" estimates given by the plant. It is now planned to chemically recover the Pu in the slag and crucible material to provide a direct comparison between the nondestructive assay results, and actual (rather than "by difference") Pu content.

TABLE XII
PASSIVE NEUTRON COINCIDENCE
COUNTING OF PuO_2 IN ASH RESIDUE

Hanford Can No.	Passive Neutron Coincidence Counting			Hanford Loading PuO_2 (g)
	^{240}Pu (g)	Pu (g)	PuO_2 (g)	
29-9-2	3.03	35.30	40.32	40.07
29-9-3	1.10	8.86	10.02	10.09
29-9-4	4.48	36.20	40.46	40.08
29-9-5	0.11	0.87	0.97	1.02
29-9-6	1.99	8.77	9.80	10.11
29-9-7	7.91	34.96	39.34	40.07

Another practical application of neutron coincidence counting is in the assay of $^{238}\text{PuO}_2$. In addition to determine the amount of ^{238}Pu present, it was necessary to determine the fraction of neutrons originating from (α, n) reactions in the oxygen and other impurities. The Pu samples (~ 80% ^{238}Pu) were either in the form of metal (Samples Nos. 179, 180, 181) or metal oxides having various enrichments of ^{16}O . The samples were counted without removing them from their 2-R shipping containers and the results are listed in Table XIV. The ratio of single events to coincidence events gives the (α, n) neutron contribution

TABLE XIII

NONDESTRUCTIVE Pu ASSAY IN
ONE-GALLON CAN SCRAP CONTAINERS

Slag & Crucible Can No.	Total Wt. (kg)	"By-Difference" Mass Assignment (g Pu)	Passive		Active Neutron	
			Neutron (g Pu)	Gamma (g Pu)	(~ 3 MeV) (g Pu)	(~ 0.5 MeV) (g Pu)
G-1	7.5	17.64	Standard	Standard	Standard	Standard
G-2	7.7	176.4	177.0 ± 10.0	174.0 ± 10.0	178.0 ± 5.0	168.0 ± 5.0
XBL-40-1	7.2	13	23.4 ± 3.8	18.5 ± 2.5	$22.5 \pm .7$	$20.6 \pm .6$
XBL-40-2	6.7	13	18.5 ± 2.6	15.0 ± 1.5	$16.6 \pm .5$	$15.0 \pm .8$
XBL-42	4.8	11	13.5 ± 2.0	12.6 ± 2.0	$14.4 \pm .5$	$14.7 \pm .6$
XBL-43	7.2	23	25.5 ± 3.9	19.6 ± 2.5	$24.0 \pm .7$	$21.0 \pm .8$
XBL-44	6.0	18	17.1 ± 2.6	14.1 ± 1.5	$15.8 \pm .5$	$15.8 \pm .5$
XBL-45	5.7	24	14.8 ± 2.3	14.7 ± 2.0	$16.5 \pm .5$	$15.0 \pm .5$

and it was estimated in Table XIV that the purest metallic sample had a 1% (α, n) neutron contribution. Sample No. 60, which has a 490% (α, n) contribution, is representative of normal $^{238}\text{PuO}_2$ before ^{16}O enrichment.

TABLE XIV

PASSIVE NEUTRON COUNTING OF ^{238}Pu
SAMPLES TO DETERMINE
(α, n) BACKGROUND CONTRIBUTION

Sample No.	(α, n) Neutron Contribution	Neutron Assay g ^{238}Pu	Chemical Assay g ^{238}Pu
181	1%	Standard	1.046
180	1%	.785	.786
179	3%	1.00	1.01
139	15%	1.26	1.27
143	16%	1.39	1.43
128	36%	1.44	1.46
123	59%	1.52	1.55
119	175%	1.76	1.63
60	490%	1.51	1.69

4 π Neutron Counter for Fifty-Five Gallon Drums

J. E. Foley, D. B. Smith, and R. B. Walton

Calculations and experiments were performed to ascertain the feasibility of coincidence counting 55-gallon barrels. The leakage of neutrons from barrels containing plutonium dispersed in various matrix materials is of particular interest because the coincidence counting rate is approximately proportional to the square of the neutron leakage probability. One-dimensional calculations using DTF-IV ($S_n = 8$, Hansen-Roach 16-group cross sections) were performed for a spherical container (24-inch diameter) to investigate both the fractional leakage and the leakage neutron spectrum. The spontaneous fission source--a Maxwellian with $T = 1.275$ (28)--was distributed uniformly throughout a polyethylene matrix.

The number of neutrons per unit source leaking from the spherical "drum" is shown as a function of polyethylene density in Fig. 26. Curves are given for total leakage and for neutrons having

²⁸ James N. Terrel, LASL, private communication, (1970).

Fig. 26. Leakage of spontaneous fission neutrons from ^{240}Pu in a simulated 55-gallon drum containing polyethylene matrix material. Isolated points show the corresponding results when 200 lbs. of Fe are added to the system.

energies above and below the cadmium cutoff. The calculated results indicate that greater than 90% of the neutron from spontaneous fission of ^{240}Pu will leak from a system containing polyethylene having a density of $\rho = 0.2 \text{ g/cm}^3$. This density corresponds to compressed polyethylene chips and is perhaps the highest CH_2 density that would ordinarily be found in practice. About 80% of the source neutrons leaking from the system have energies greater than $\sim 0.4 \text{ eV}$. Furthermore, the addition of 200 lbs of iron to the drum (isolated points in Fig. 26) does not greatly change the neutron leakage characteristics. The number-spectrum of the neutrons leaking from this system (i.e., $\rho(\text{CH}_2) = 0.2 \text{ g/cm}^3$) is shown in Fig. 27. The ^{240}Pu source spectrum is also shown in Fig. 27 for comparison. On the basis of these results, it appeared that assay errors due to attenuation of the ^{240}Pu spontaneous fission neutrons should not be excessive, and it

Fig. 27. Number-spectrum of spontaneous fission neutrons from ^{240}Pu in a simulated 55-gallon drum containing polyethylene of density $\rho = 0.2 \text{ g/cm}^3$. Dashed line indicates ^{240}Pu source spectrum.

just might be feasible to develop a practical 4π neutron coincidence detector for 55-gallon barrels.

Based on the encouraging feasibility calculations just described, a 4π neutron coincidence detector for assay of Pu waste contained in 55-gallon drums has been designed and is under construction at LASL. This unit can be used independently or in conjunction with the MONAL assay laboratory. The unique features of this detector are its high efficiency, its capability for ^{240}Pu neutron coincidence assay, and its shielding and geometrical design which permit mobile field operation. It is expected that this detector will prove quite useful in screen-

ing waste storage barrels for discard limits, (29) as well as for quantitative assay of scrap having higher concentrations of fissile material.

Measurements performed in a mock-up assembly of neutron counters embedded in polyethylene showed that an intrinsic efficiency of about 15% can be obtained with a single annular array of 2-in. diam. BF_3 counters. Based on these studies, a system was designed which consists of an annular ring of polyethylene completely surrounding the barrel and containing 36 BF_3 -filled detectors, each detector being two inches in diameter, 36 inches long, and filled to a pressure of 68 cm Hg. The top and bottom of the barrel are enclosed similarly with a total of 18 2-in. -diam. BF_3 counters embedded in slabs of polyethylene. The polyethylene is four inches thick and the counters are spaced approximately 0.75 inches apart. These dimensions give sufficient mechanical strength along with high neutron detection efficiency. There is provision for adding another layer of counters to achieve higher efficiency, but this may be done only after the first system has been thoroughly tested.

The polyethylene detection assembly is enclosed by a one-foot-thick water shield which consists of an annular array of small interlocking aluminum tanks.

One-half of the cylindrical body of the tank and detector assembly can be rolled back on horizontal tracks to permit convenient horizontal loading of barrels. The entire system can be assembled by two people; it can be set up in the vicinity of MONAL and operated using the data acquisition facility in MONAL. Alternatively, with separate electronics and data acquisition equipment, it can be operated entirely independently of MONAL.

Calculations have been made to predict the coincidence counting rate from this 4 π barrel

detector for various amounts of ^{240}Pu in the barrel. Estimates have also been made of the counting time required to obtain a given precision in the ^{240}Pu assay. The counting time required for a specified precision is dependent on the detector efficiency, neutron lifetime, coincidence gate width, and accidental counting rate. Figure 28 illustrates the estimated time to obtain a precision of ten percent in the coincidence counting rate for various values of the accidental counting rate, as would be recorded by the accidental gate scaler. A neutron die-away time of 75 μsec and a coincidence gate width of 100 μsec were assumed for this example. Furthermore, it was assumed that the samples contained five percent ^{240}Pu . (30) It is seen that counting times less than 100 sec are sufficient for a five-gram sample with low accidental gate rates (< one count/sec), while counting times on the order of

Fig. 28. Time required to obtain 10% precision in coincidence counting rate vs accidental counting rate for 4 π neutron barrel counter.

²⁹ cf. AEC General Manager's recent Immediate Action Directive No. 0511-21 re Pu waste burial, and similar directives to come. (^{235}U as well as Pu.)

³⁰ e.g., the five-g sample of Pu contained 0.25 g of ^{240}Pu , etc.

15 minutes are required for accidental rates of 100 counts/sec. For smaller samples, counting times will be correspondingly longer. At low accidental counting rates, the counting time required for a given precision is inversely proportional to the amount of plutonium in the sample, while at high

accidental rates (≈ 10 counts/sec) the counting time is inversely proportional to the square of the amount of plutonium. Of course, counting times can be reduced somewhat by increasing the detection efficiency and reducing the die-away time of the system.

NUCLEAR INSTRUMENTATION AND APPARATUS DEVELOPMENT

Electronics and Apparatus Development

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

The design of several electronic instruments was undertaken during this reporting period. In addition to this design work, (see description of instruments below) complete specifications for a Ge(Li) spectrometer system were prepared for group CMB-11, LASL.

Quad NaI(Tl) Scintillation Detector Preamplifier

A compact package containing four charge-sensitive preamplifiers was designed for use with a NaI(Tl) barrel scanning system being developed for use in the MONAL.⁽³¹⁾ Each circuit consists of a charge sensitive input stage followed by a gain-of-ten line driver. The preamplifier pulses are RC-CR shaped, with a rise time of about 0.5 μ s and a decay time of 0.75 μ s. Output pulses are linear to about 6 V. Sufficient gain is incorporated in the preamplifiers to obviate the need for further pulse amplification in ²³⁹Pu gamma-ray counting applications. The preamplifiers are operated from a single + 12 V power source.

Eight-Input Single Channel Analyzer

A single dual-width NIM module containing eight independent single-channel analyzers with common threshold controls was developed for use with the gamma scanning system for 55-gallon barrels, as described earlier.⁽³¹⁾ The analyzer module, along with a quad preamplifier described above, is shown in Fig. 29. The analyzer input

connectors are located on the rear of the module; output connectors, test points, and threshold controls are located on the front panel. Input pulse range is 0 to + 5 V, so that output pulses from the quad preamplifier may be used directly without further amplification. The circuitry has been tested in a "bread-board" model, and a prototype unit is presently nearing completion.

Fig. 29. View of Quad Preamplifier (left) and Eight Input Single Channel Analyzer designed for the MONAL NaI(Tl) barrel scanner.

³¹ See previous section on Gamma-Ray Scanning for Assay of Fifty-Five Gallon Drums.

Gated Preamplifier for NaI(Tl) Scintillators

A simplified version of the gated preamplifier described previously⁽³²⁾ was designed specifically for use with NaI(Tl) scintillation detectors. This preamplifier has less gain and improved pulse overload characteristics than the previous one developed for neutron counting.

Random Pulse Generator

Circuit design and testing is underway on a random pulse generator for use in making dead-time-loss and time-resolution measurements in

various counting systems. It is planned to include many features in this unit that are not available in commercial units. Such features include:

1. True randomness-in-time down to 0.5 μ s or less.
2. Variable pulse shaping.
3. Variable randomness in pulse height as well as time.

It is expected that a complete prototype unit will be constructed in the near future.

COMPUTATIONAL AND DATA ACQUISITION FACILITIES

LAMPS, Los Alamos Modular Programming System

D. B. Smith

A modified version of the KAPL DATATRAN modular programming system⁽³³⁾ (renamed LAMPS for Los Alamos Modular Programming System) has been made operational on the LASL CDC-6600 computers.⁽³⁴⁾ This system allows users to easily connect a series of functional program units (modules) into an integrated computational package. Free-standing codes (with only slight modification) may be readily executed and linked to other free-standing codes or to modules. Data lists generated by either free-standing codes or modules can be saved easily on tape and recalled for later use. Blocks of data can be stored as modules and are then available to either free-standing codes or to other modules.

These features of LAMPS make the system attractive for such parametric studies as matrix-material effects on delayed-neutron response.

Each calculation in such a study formerly required execution of two separate DTF-IV problems and a third computer calculation which used the scaler fluxes from the DTF-IV runs to compute parameters of interest such as fission densities, delayed neutron response, etc. With the modular programming system, both DTF-IV problems and the computation or connecting problem are all executed in a single computer run.

To use LAMPS, the DTF-IV code was modified, compiled, and stored on a properly initialized LAMPS system tape along with the connecting code and cross sections needed for the DTF-IV problems. The parameters and data required for the DTF problems is input to the LAMPS job as data lists. These data lists and the appropriate cross sections are processed by an input processor (also stored on the LAMPS system tape) which prepares a disc file in the correct DTF-IV format. DTF-IV reads this file instead of the normal input stream. The scaler fluxes generated in both DTF-IV problems are available to the connecting code and also are saved in readily accessible form for future use.

³² L. V. East, Nucl. Inst. and Meth. 71, 328 (1969).

³³ H. J. Kopp, J. D. Morris, and W. E. Schilling, DATATRAN Modular Programming System for Digital Computers, KAPL-M-6997, Knolls Atomic Power Laboratory (1968).

³⁴ Frank McGirt, private communication, LASL (1970).

Computer Based Data Acquisition System

L. V. East and K. H. Augustson

The data acquisition and analysis system described previously⁽³⁵⁾ was received in February, 1970. Basically, the system hardware consists of the following:

1. A small general purpose computer with 8192 18-bit word core memory (Digital Equipment Corporation model PDP-9/L).
2. Two high resolution analog-to-digital converters (Northern Scientific model NS-624's).
3. Two "DECTape" tape transports and controller.
4. A high speed paper tape punch and reader.
5. A digital plotter (Calcomp model 565).
6. A 14-inch cathode ray tube display with light pen.
7. A software interrogated functional control panel (Tennecomp model TP-1347).
8. A model KSR-35 teleprinter.

A photograph of the system is shown in Fig. 30. The display is at the left with the control panel and ADC's directly above. The blank cabinet on the right is for future expansion.

In addition to the usual system programs (assembler, Fortran IV compiler, editor, etc.), a set of programs for single and dual parameter pulse height analysis and generalized multiscaler operation were also obtained. In these programs, maximum use is made of the control panel and light pen for such operator-computer interactions as the set up of initial run conditions, selection of regions of interest for displaying, printing, or plotting, and for analysis of data for peak locations and area under a peak.

During single-parameter pulse height analysis, the two ADC's can be used independently to store data in two separate sections of a 4096-channel data region of memory. The size and relative location of the data region associated with each ADC is selected by a series of hardware switches and can be any size from 0 to the full 4096 channels

Fig. 30. Overall view of the computer-based data acquisition system. The left cabinet contains the display scope, control panel, light pen, and ADC's. The center section contains the PDP-9/L central processor, ADC interface control, and DECTape units.

³⁵ LA-4227-MS, p. 21 (1969).

available. While data are being collected in one section of memory, the contents of another (or the same) section can be plotted, printed, or operated on to determine peak locations, intensities, etc.

In the near future we plan to add an inexpensive card reader, and interface electronics for a computer-compatible magnetic tape transport obtained from another group at LASL. The card reader will greatly simplify off-line program preparation and editing. Use of the magnetic tape transport will ease the problem of communication with the larger computers located at the LASL central

computer facility. Such communication must presently be done via punched paper tape.

The system has already been extensively used to acquire data for precision gamma ray energy measurements,⁽³⁶⁾ and for the acquisition and analysis of plutonium-scrap gamma-ray scans.⁽³⁷⁾ Codes are presently being written for the analysis of delayed neutron yield data and least-squares photopeak fitting. FOCAL, a real-time programming language, is available on the system and has been used for several small data reduction and calculational problems.

MOBILE NONDESTRUCTIVE ASSAY LABORATORY (MONAL)

J. H. Menzel, B. R. Dennis, D. E. Helfer and J. H. Kottman

Progress on the MONAL itself during the period of January through April 1970 can be summarized as follows:

1. Neutron generator testing and acceptance, and installation of a new post-acceleration beam deflection system.
2. Completion of equipment room facilities, mounts, auxiliary shield tanks, and large sample handling system improvements.
3. Installation of an automatic fire protection system.
4. Complete MONAL checkout, debugging and upgrading, including installation and checkout of all electronic instrumentation, data acquisition and health monitoring equipment.
5. Field-deployment tests at LASL's plutonium facility (DP-Site) including logistics, personnel training, and nondestructive assay of fissionable material in one-gallon cans and 55-gallon drums.

The detection portion of the automatic fire protection system consists of six pairs of product-of-combustion and fixed-temperature/rate-of-rise

devices mounted at strategic locations throughout the MONAL. The fire extinguishing system uses pressurized bromotrifluoromethane (Freon 1301), an extremely efficient extinguishing agent for all types of fires. Unlike CO_2 , Freon 1301 gas is non-toxic when inhaled. Audio and visual alarms, internal and external to the MONAL, indicate both detector malfunction as well as automatic actuation of the fire extinguishing system.

Proper radiation-monitoring equipment is essential for any laboratory involving the use of radioactive sources and materials. Consequently, the MONAL is fully equipped with portable units such as REM, alpha, geiger, and tritium counters as well as permanently-mounted monitors located at the MONAL console. The neutron dose level at the console is automatically monitored whenever the neutron generator is operating, and the data is permanently recorded along with a time code and neutron generator yield monitor. Potential plutonium contamination is monitored throughout the

³⁶ See earlier section, "Precision Gamma-Ray Energy Measurements," this report.

³⁷ See earlier section, "Gamma Scan of Medium-Density ^{239}Pu Scrap Cans," this report.

MONAL by a manifold system connected to an alpha air monitor which employs a Rustrak recorder to maintain permanent records.

The neutron interrogation--delayed neutron detection method of nondestructive assay of fissionable materials requires the use of a neutron generator with an on/off ratio of $\sim 10^8$. Measurements showed that the Kaman A-1254 neutron generator as originally procured did not satisfy this requirement. The subsequent installation of a 7-degree post-acceleration beam deflection system eliminated the problem of neutrons produced by residual beam "dark current", and resulted in a beam on/off ratio well in excess of the required value of 10^8 .

On March 17, 1970, the first field test of the MONAL began at the Los Alamos plutonium facility (DP-Site). The purpose of this test was to evaluate and develop the logistics of field deployment, to train MONAL personnel in the set-up, operation

and maintenance of all systems, and to study the accuracy, sensitivity, and applicability of active neutron interrogation and assay methods in the MONAL environment. (Attention was first focused on assay of one-gallon cans and 55-gallon barrels). Experimental results of this research are described in other sections of this report. Figs. 31 through 36 show some operational features of the instrumentation and facilities aboard the MONAL.

Testing of the MONAL at LASL's DP-Site has proved extremely useful in preparing the MONAL for its first field deployment (~ 1 May) at an industrial nuclear facility, i.e., the Dow Chemical Company's Rocky Flats Plant near Denver, Colorado.

Fabrication of complete passive assay systems for MONAL is continuing and it is expected that some of this equipment will be installed and tested during the MONAL's deployment at Rocky Flats.

Fig. 31. Assay of 55-gallon barrels being conducted from the control console and data acquisition area located forward in the MONAL. The Ge(Li) system (lower right) provides complete on-board capability for high resolution passive gamma nondestructive assay.

Fig. 32. Inspecting the 14 MeV neutron generator located in the rear equipment room of the MONAL.

Fig. 33. Fifty-five gallon scrap barrel elevated into position for delayed neutron assay in the shielded interrogation chamber of the MONAL. The high-efficiency delayed neutron "slab" detector (unshielded) is seen in the foreground.

Fig. 35. Screening samples for radioactivity in sample preparation room of the MONAL prior to assay.

Fig. 34. Weighing samples on a precision balance in the sample preparation room of the MONAL.

Fig. 36. Inserting ^{235}U in access tube of 55-gallon barrel standard containing known polyethylene loading.

Van de Graaff Accelerator Facility

A. E. Evans and M. S. Krick

During this reporting period, work on the N-6 Vande Graaff accelerator facility was concerned principally with improving the sensitivity and reproducibility of small-sample assay by delayed-neutron techniques. Preparation has been made for accelerating deuterons in order to extend the energy range of incident neutrons (via the $D(D, n)He^3$ reaction) in the program of measuring delayed-neutron yields vs. neutron energy. In addition, some time was spent on Rover scrap analysis and on calibration of film badges and TLD dosimeters for the Health Physics Group at LASL.

It has been found that further improvement of the accuracy of small-sample analysis using the Van de Graaff will require reduction of backgrounds due to ambient radiation, neutron leakage from the accelerator room into the target area, and neutrons from target dark current caused by leakage of neutral particles and scattered ions through the post-acceleration pulsing system. The latter backgrounds vary unpredictably and result, at present, in a minimum uncertainty of 50 mg in the assay of plutonium. The effect of accelerator-room neutron leakage has been reduced to less than 10% of ambient background by the use of PVC-B₄C curtains between the experiment and the accelerator-room wall of the experimental area. In order to reduce the target dark-current background, a rotating mechanical beam shutter is being designed. This shutter will be synchronized with the existing post-acceleration beam-pulsing system.

The ambient neutron background, as measured with an N-6 slab detector, is equivalent to the delayed-neutron response from 100 mg of plutonium using optimum geometry and an interrogating-source strength of $\sim 2 \times 10^{10}$ neutrons/sec. In order to reduce this undesirable ambient back-

ground level, improve radiation decoupling between the accelerator and experimental areas, and permit safe operation of the accelerator at full intensity ($\sim 10^{12}$ neutrons/sec), the existing target-area shielding is being augmented. The new target shield design will provide at least three-foot-thick concrete walls and a two-foot-thick concrete ceiling. The new shielding structure will consist of pre-cast blocks, beams, and slabs, which are now being fabricated.

Cockcroft-Walton Accelerator Facility

C. N. Henry and J. J. Malanify

During this reporting period, the N-6 Cockcroft-Walton accelerator was in service for all but five days; this down time was used largely for maintenance of the ion source and gas bottle/palladium leak system.

Preliminary tests have been made to check the feasibility of gating the RF ion source for the slow pulsing mode (> 50 ms pulse width). Initial results indicate sufficient background reduction to warrant pursuing this upgrading of the Cockcroft-Walton accelerator. It is hoped that by July 1970 all necessary parts will be installed and operating to provide gated ion source capability in conjunction with a post-deflection system. Discussions were held with Ortec people at Oak Ridge on fast klystron post-acceleration bunched pulsing in the time region of 1 - 2 nsec. It appears that this upgrading of Cockcroft-Walton accelerator pulsing capability will be feasible at reasonable cost.

Accelerator I Neutron Generator

R. H. Augustson

Recent improvements on Accelerator I include installation of a shorter drift tube which can be accommodated inside N-6's various spectrum

tailoring systems, and allows more beam defocusing, when desired, to prolong target life.

Accelerator usage has been divided between measurements on active assay of Pu in scrap re-

covery cans and on Uhtrex fuel elements, with the major fraction of the time devoted to Pu scrap measurements.

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