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Pulsed Neutron Research for Nuclear Safeguards

Program Status Report
October-December, 1967

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NUCLEAR SAFEGUARDS RESEARCH SERIES

G. Robert Keepin, Editor

This LA...MS report presents the status of the nuclear safeguards research program at Los Alamos. Previous reports in this series are:

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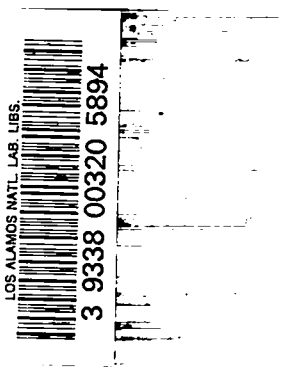

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PULSED NEUTRON RESEARCH FOR NUCLEAR SAFEGUARDS

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KINETIC RESPONSE TECHNIQUES FOR NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS

During the fourth quarter of 1967, the experimental program for application of delayed neutron kinetic response techniques to nuclear safeguards has entered a more quantitative phase. Data acquisition has been facilitated by the use of a high-efficiency, "flat"-energy-response neutron detector (cf. LA-3802-MS), a newly designed and constructed time base generator, and improved time analyzer readout ability (cf. later section on Instrumentation and Detector Development). A systematic study was made of irradiation and counting times to determine an optimum data taking cycle for isotopic abundance determination. Measurements of the kinetic response of various mixtures of fission isotopes were taken, and the data analyzed for isotopic abundance in each case. Measurements were repeated many times to determine the reproducibility of the results and to observe any systematic errors in the experiment. The question of how close kinetic response measurements using 14 MeV neutron irradiations can approach the theoretical discrimination ratios indicated for fission-spectrum-induced fission (cf. LA-3741) was also investigated.

The experimental arrangement used in the various measurements was similar to that previously reported (cf. N-6 Program Status Report, LA-3802-MS, July-Sept. '67). Samples weighing ~300 grams were placed 3/4" from the (D, T) neutron source and the N-6 high-efficiency long counter was positioned 10" from the samples. Care was taken during sample changing to reproduce the sample position precisely, and auxiliary measurements were carried out to insure negligible neutron backscattering from detector to sample. The lower energy of any such back-scattered neutrons would result in a somewhat enhanced ^{235}U delayed neutron response compared to that of ^{238}U. It was found that background under normal modulated-beam operating conditions but with no sample in place is not constant with time, but rather decays with a half-life of ~4 sec. This background is believed largely attributable to n, p processes in the oxygen of the detector (cf. Instrumentation and Detector Development section), the main contributing reaction being

$$^{17}O(n, p)\,^{17}N \xrightarrow[4.14\ \text{sec}]{\beta^-} {}^{17}O \searrow {}^{16}O + n$$

In the present measurements this background is the order of one percent of total counts, but neglecting even such a small time-dependent background would lead to a 1 to 2% error in calculated isotopic abundances.

Table I shows measured isotope discrimination ratios as a function of irradiation time, keeping the total period (irradiation plus counting time) fixed at 20 seconds. The counting time fiducials used in these measurements were the first 0.10 second and the last two seconds of the delayed neutron decay curve. The resulting discrimination ratios reflect, of course, the differences in relative delayed neutron group abundances among the different fission isotopes. Clearly higher discrimination ratios permit more accurate isotopic abundance determination in a given mixture of isotopes, providing the

necessary neutron counting statistics can be obtained in a reasonable time. From Table I it is seen that shorter irradiations lead to higher discrimination ratios as expected, but these require correspondingly longer counting times to accumulate data of comparable statistics.

TABLE I

ISOTOPE DISCRIMINATION RATIOS[a] AS A FUNCTION OF NEUTRON IRRADIATION TIME

Fission Isotopes	Irradiation Time (seconds)		
	.1	.5	1.0
^{238}U, ^{235}U	2.41	2.11	2.04
^{238}U, ^{239}Pu	2.64	2.28	2.09
^{235}U, ^{239}Pu	1.09	1.08	1.03

[a] See text for definition.

Additional data on the kinetic response of mixtures of isotopes were taken using a 1-sec irradiation and both a 14-sec and a 20-sec counting time. Figure 1 presents the measured decay data for ^{235}U-^{238}U mixtures and for the pure isotope runs using a 14-sec counting time. (The ^{235}U sample is 93% enriched Oralloy.) The decay curves in Figure 1 were normalized to equal counts at t = 13 seconds. These data were analyzed by comparing the measured pure isotope decay curves to the unknown-mixture decay curve, i.e., by dividing the latter curve into time bins and determining the best fit for all bins by the method of maximum likelihood (cf. N-6 Report N-6-1009). Various bin divisions were tried and the results are shown in Table II for a 2-bin and a 6-bin analysis. It can be seen that by proper choice of a 2-bin division, results are nearly as good as for a 6-bin division. A more complete analysis of assay precision is presently being pursued, but is not yet completed; therefore, the total error associated with the fit is not yet known. The statistical uncertainty is 1% or less in each bin. Background was neglected in these runs, and this introduces an additional 1% error. The additional pure ^{238}U run listed in Table II was introduced into the analysis as an unknown mixture, and thus was measured separately from the ^{238}U run used as a standard. It can be seen from Table II that relative isotopic abundance can be determined, under these experimental conditions, to within 4%.

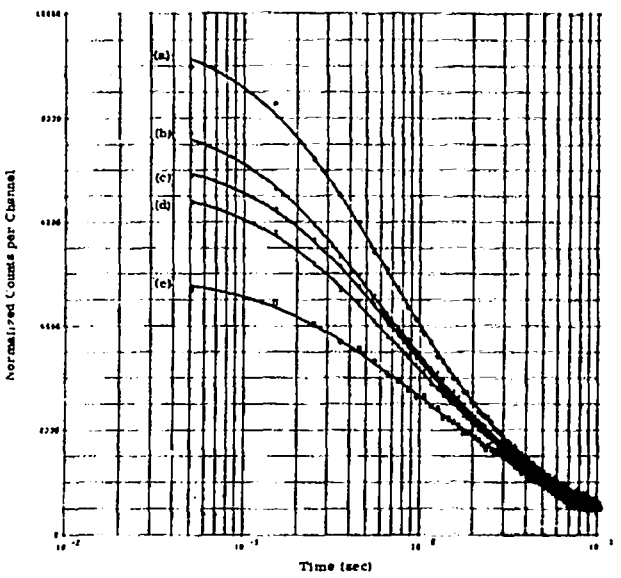

Fig. 1. Delayed neutron decay curves for: (a) 100% ^{238}U; (b) 69% ^{238}U + 31% ^{235}U; (c) 52% ^{238}U + 48% ^{235}U; (d) 35% ^{238}U + 65% ^{235}U; (e) 100% ^{235}U. (Computer microfilm; 4020 Display System).

TABLE II

MEASURED RELATIVE ISOTOPIC ABUNDANCES FOR ^{238}U-^{235}U COMPOSITE SYSTEMS

Actual Relative Abundance		Measured Relative Abundance			
		2-Bin Analysis		6-Bin Analysis	
^{238}U	^{235}U	^{238}U	^{235}U	^{238}U	^{235}U
1.000	.000	.999	.001	.991	.009
.686	.314	.673	.327	.675	.325
.520	.480	.548	.452	.541	.459
.351	.649	.376	.624	.380	.620

To improve on these results and to test reproducibility, another series of runs was made with the goal of repeating a given mixture a number of times. Care was taken to reduce the background and to apply detailed corrections for the remaining decaying background. The counting time was increased to 20 seconds, which resulted in a slightly increased discrimination. Data were taken on two different days for a 50-50 mixture of ^{235}U and ^{238}U for a total of 9 determinations of the isotopic abundance. Two sets of pure-isotope runs were taken each day and one set was included as an unknown in the analysis. These results are presented in Table III. For the 50-50 mixtures, the average deviation from the mean is 1.6%. There seems to be a systematic trend for the ^{235}U results to be low. A 1% error in the standard runs would account for this. The combined statistical uncertainty for each determination is at least 1%, and much longer running time would be required to improve significantly on this. The evidence is that the error introduced by small shifts in the electronics of the timing circuit and the amplifier gain setting during a day's running is small, and the accuracy of relative isotopic abundance measurements for ^{235}U and ^{238}U is approximately 3%.

TABLE III

REPRODUCIBILITY OF RELATIVE ISOTOPIC ABUNDANCE ANALYSES

Actual Isotopic Abundance		Measured Isotopic Abundance[a]	
^{238}U	^{235}U	^{238}U	^{235}U
0.50	0.50	0.509	0.491
"	"	0.519	0.481
"	"	0.518	0.482
"	"	0.518	0.482
"	"	0.514	0.486
"	"	0.487	0.513
"	"	0.501	0.499
"	"	0.511	0.489
"	"	0.506	0.494
1.0	0.0	0.988	0.012
"	"	1.019	-0.019
0.0	1.0	-0.011	1.011
"	"	-0.023	1.023

[a] Mean for all .50-.50 results: 0.509 for ^{238}U and 0.491 for ^{235}U
Average deviation from mean: 0.008

The availability of neutron generators with 5-10 times higher neutron flux than the present N-6 Cockcroft-Walton accelerator promises further significant improvement in isotope discrimination ratios by counting for longer times after irradiation. To investigate such possible improvements in isotope discrimination ratio, kinetic response data were taken for a 100-millisecond irradiation and a 100-second counting time. These data were analyzed to give the quantities R_{f-} and R_{f+} (cf. LA-3741) for ^{238}U and ^{235}U. Theoretical values of R_{f-} and R_{f+} have been calculated assuming an instantaneous irradiation, infinite counting time, and using the relative delayed neutron group abundances measured for fission-spectrum-induced fission rather than 14 MeV-neutron-induced fission. Table IV lists the calculated and measured R_{f-} (0.05 sec time fiducial) and R_{f+} (20 and 40 sec time fiducials) and corresponding overall isotope discrimination ratios (R_{f-} times R_{f+}). In order to compare R_{f-} values directly, it is necessary to correct experimental numbers for delayed neutron decay during the finite irradiation time of 0.1 sec.

TABLE IV

COMPARISON OF THEORETICAL AND MEASURED VALUES OF R_{f-} AND R_{f+} RATIOS FOR ^{238}U AND ^{235}U

R_f Ratios	R_{f-} (f = 0.05 sec)	R_{f+} (f = 20 sec)	R_{f+} (f = 40 sec)	Isotope Discrimination Ratio [R_{f-} and R_{f+} (40 sec)]
Theoretical[a]	1.77	1.75	1.83	3.17
Measured[b]	1.75 ± .08	1.59 ± .03	1.63 ± .05	2.85 ± .16

[a] See Los Alamos Report, LA-3741 (1967).

[b] 0.1 sec irradiation, 100 sec count

Saturation irradiations were performed in order to investigate the isotope discrimination available in the longer-lived delayed neutron groups. $S_{f+/\Delta}$ ratios (cf. LA-3741) were measured for ^{238}U, ^{235}U, and ^{239}Pu and the results compared with theoretical calculations. Irradiation times of 40 and 330 sec were used in order to measure the sensitivity of discrimination ratios to the degree of saturation in the delayed neutron precursors ($T_{1/2} \leq 55$ sec). The results shown in Table V correspond to counting time fiducials of 50 sec and 100 sec.

TABLE V

COMPARISON OF THEORETICAL AND MEASURED VALUES OF $S_{f+/\Delta}$ RATIOS FOR VARIOUS ISOTOPES

	$S_{f+/\Delta}$ (f = 50 sec)			$S_{f+/\Delta}$ (f = 100 sec)	
Isotopes	Measured[a]	Measured[b]	Theory[c]	Measured[b]	Theory[c]
^{238}U, ^{235}U	1.79 ± .09	1.81 ± .05	2.18	2.05 ± .05	2.59
^{238}U, ^{239}Pu	1.98 ± .10	1.95 ± .05	2.60	2.22 ± .06	2.79
^{235}U, ^{239}Pu	1.11 ± .07	1.09 ± .03	1.13	1.08 ± .03	1.08

[a]The irradiation time was 40 sec and the total counting time was 200 sec.

[b]The irradiation time was 330 sec and the total counting time was 310 sec.

[c]See Los Alamos Report, LA-3741 (1967).

The product of the experimental ratios, R_{f-} (= 1.75 at f = 0.05 sec) and $S_{f+/\Delta}$ (= 2.05 at f = 100 sec) gives an experimental discrimination ratio of 3.60 for ^{238}U and ^{235}U, which may be compared with the limiting theoretical value of 4.59. By comparing the 40 sec irradiation with the 330 sec irradiation, it can be seen that the degree of saturation does not appreciably affect discrimination ratios, and so it is possible to optimize data collection time in practical applications. Thus in saturation irradiations a complete data cycle time (irradiation plus counting) of a few minutes can be used, and adequate counting statistics for accurate $S_{f+/\Delta}$ ratios can typically be obtained in less than 30 minutes. As seen in Tables IV and V, experimental values of R_{f+} and $S_{f+/\Delta}$ are significantly smaller than the theoretical expectation; this difference is believed attributable to differences in relative delayed neutron group abundances for 14 MeV fission as compared to fission-spectrum-induced (1-3 MeV) fission.

Further indications of changes in delayed neutron group abundances with increasing energy of the neutron inducing fission are seen when kinetic response curves are analyzed by fitting them to a sum of exponentials. In particular, the least-squares-fitted relative abundances and periods of the delayed neutron groups indicate that a_i and λ_i values are indeed somewhat different at 14 MeV than for fission-spectrum-induced fission. (As noted in LA-3802-MS, Group N-6 is preparing a series of detailed measurements and analyses of the periods and relative abundances of delayed neutrons from 14 MeV neutron-induced fission of the major fissioning species.) The sum-of-exponentials fitting procedure just mentioned has been applied to all data reported herein, and offers an alternative method of analysis of kinetic response data to determine isotopic abundances. The resulting abundance values so obtained compare well with the area analysis described earlier, although there is more scatter in the results.

Another type of kinetic response measurement is being directed toward isotope discrimination based on the characteristic differences in delayed neutron fractions, β, between the major fissile species (e.g., a factor of 3 between β values for ^{235}U and ^{239}Pu). Attempts are being made to distinguish prompt (fission spectrum) neutrons from the 14 MeV neutrons of the primary interrogation beam. In preliminary experiments the neutron shielding and collimation have proved inadequate, but work is continuing on improved collimation and shielding, as well as possible energy discrimination to help distinguish the prompt fission neutrons produced in the unknown sample from the 14 MeV primary-beam neutrons.

PASSIVE NEUTRON COUNTING TECHNIQUES

It is well known that samples containing plutonium emit neutrons from spontaneous fission of ^{240}Pu. If light elements are present in the sample, additional neutrons can result from (α, n) reactions initiated by alpha decay of the plutonium isotopes. Consequently, detection of these neutrons may provide a basis for non-destructive quantitative analysis of ^{240}Pu and ^{239}Pu in unknown samples (one practical case in point being irradiated reactor fuel elements). The method has already proved valuable for routine monitoring of plutonium in fabrication and recovery processes at LASL (especially in cases where the relative abundances of ^{239}Pu and ^{240}Pu are already known).

To illustrate a specific application of the "passive neutron counting" technique to nuclear safeguards, some preliminary estimates have been made for an irradiated reactor fuel element with characteristics approximating those of a Yankee fuel element. This element is a matrix of fuel pins and water cooling channels having outer dimensions of approximately 8" x 8" x 92". At the end of its power cycle, a "spent" reactor fuel element is expected to contain heavy isotopes (in oxide form) in the representative amounts given in Table VI.

TABLE VI

REPRESENTATIVE HEAVY ISOTOPE CONCENTRATIONS IN "YANKEE-TYPE" SPENT FUEL ELEMENT

Isotope	Quantity (in kilograms)
^{238}U	270.0
^{235}U	6.6
^{239}Pu	1.5
^{240}Pu	0.23

A synthesis of the neutron source strengths estimated for these heavy isotope concentrations is presented in Table VII.

TABLE VII

NEUTRON SOURCE STRENGTHS FROM THE HEAVY ISOTOPES IN A "YANKEE-TYPE" SPENT FUEL ELEMENT

	O(α, n) *	Spontaneous Fission
^{238}U	76 neuts/sec	3.8×10^3 neuts/sec
^{235}U	12 "	--- "
^{239}Pu	8×10^4 "	38 "
^{240}Pu	4.3×10^4 "	3.5×10^5 "

*(α, n) yields were calculated assuming all α's have an energy of 5.3 MeV and the yield of the oxide compound is one third of the infinite-dilution yield, 7×10^{-8} neutrons/α, obtained from the data for ^{210}Po α's on oxygen.

The total neutron source strength of this composition is approximately 5×10^5 n/sec, of which about 80% are attributed to ^{240}Pu spontaneous fission and ^{240}Pu (α, n) reactions on Oxygen. The single dominant source is clearly the ^{240}Pu spontaneous fission neutrons, which account for approximately 75% of the total neutron source strength from the fuel element.

The chain leading to the formation of ^{239}Pu and ^{240}Pu is:

$$^{238}U + n \rightarrow {}^{239}U \xrightarrow[23.5\ min]{\beta^-} {}^{239}Np \xrightarrow[2.35\ days]{\beta^-} {}^{239}Pu + n \rightarrow {}^{240}Pu \ldots$$
$$\searrow \text{Fission}$$

The expressions for the yields of ^{239}Pu and ^{240}Pu at a given spatial point in the fuel element are (assuming negligible ^{238}U burn-up):

$$N^{49} = \frac{\sigma_c^{28}}{\sigma_R^{49}} N^{28} \left(1 - e^{-\sigma_R^{49} \tau}\right)$$

$$N^{40} = \frac{\sigma_c^{49}\sigma_c^{28}}{\sigma_R^{49}\sigma_R^{40}} N^{28}$$

$$\times \left\{ 1 - \frac{\sigma_R^{49}}{(\sigma_R^{49} - \sigma_R^{40})} e^{-\sigma_R^{40}\tau} + \frac{\sigma_R^{40}}{(\sigma_R^{49} - \sigma_R^{40})} e^{-\sigma_R^{49}\tau} \right\}$$

where

N^X = atom density of nuclide X designated by the last numerals of its Z and A,

σ_R^X = flux-averaged removal cross section of nuclide X,

σ_c^X = flux-averaged capture cross section of nuclide X,

τ = time-integrated neutron flux $= \int_0^T \varphi(t)dt$.

Since the equations for the ^{239}Pu and ^{240}Pu populations are parametric in τ, the ratio of the concentrations of these nuclides can, in principle, be calculated knowing only the flux-averaged cross sections. To illustrate this last point, the yields of ^{239}Pu and ^{240}Pu relative to that of ^{238}U (which for practical purposes may be taken as a constant) have been calculated using tabulated thermal neutron cross sections instead of specific flux-averaged quantities. These results are presented in Figure 2.

A quantitative analysis of the ^{240}Pu and ^{239}Pu in a spent fuel element based on its neutron emission may be feasible if the ^{240}Pu/^{239}Pu ratio can be ascertained (e. g., from burnup calculations), and if the (α, n) conversion factors for the specific fuel material are known. Also, the neutron counting method described above can be calibrated directly against an independent isotopic analysis (destructive) of a spent fuel element. In principle, the ^{240}Pu concentration can be obtained independently by co-

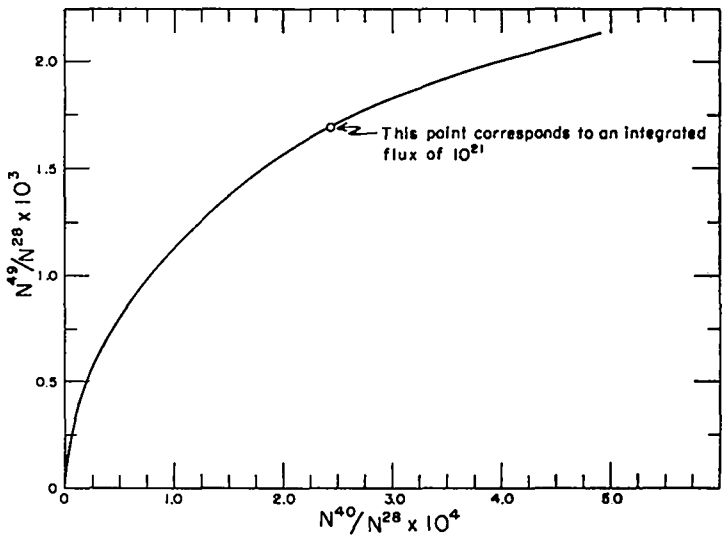

Fig. 2. Relative ^{239}Pu and ^{240}Pu concentrations in a typical spent reactor fuel element (calculations based on thermal neutron cross sections).

incidence counting the spontaneous fission neutrons; however, the feasibility of the coincidence method for fuel element assay is uncertain because of problems such as low source intensity and neutron multiplication effects.

Factors which may affect practical application of the passive neutron counting method to irradiated fuel elements are:

1) the requirement for low-level neutron counting in the presence of a high gamma radiation field;
2) extraneous neutron sources such as photofission and photoneutron reactions, and nuclides other than Pu isotopes which either spontaneously fission or emit alphas.

The neutron source intensity from the ^{240}Pu and ^{239}Pu in a typical spent fuel element should be sufficient to permit a longitudinal scan of the element with suitable gamma-insensitive neutron detectors. If we consider scanning increments of 1 foot, the source strength would be $\sim 5 \times 10^4$ n/sec-foot for a practical measurement. For source intensities of this magnitude,

neutron detection efficiencies of 10^{-4} or greater should yield adequate counting statistics in reasonable counting times. Although conventional neutron detectors having much greater efficiencies are available, they would probably not be usable in the very high radiation fields in which our low level neutron counting must be performed. Some promising systems for detecting neutrons in the "hot" fuel element environment are: (a) fission chambers, with characteristically high gamma-ray discrimination properties; (b) BF_3 proportional counters which are shadow-shielded from the gamma rays; (c) foil or solution activation.

Background neutrons may arise from photofission and (γ, n) reactions which are initiated by the energetic gamma rays from long-lived fission products. Estimates are available (cf. KAPL-M-JRS (1960)) on the intensity of high energy gamma rays emitted from a reactor core as a function of reactor operating and cooling times. For cooling times of the order of 6 months the most energetic gamma ray is the 2.9-MeV ^{140}La 12.8-day γ activity. Calculations based on present low-energy photofission cross-section data (Nuclear Physics 64, 420-432 (1965) show that after extended cooling periods the neutron yield from photofissions induced by fission-product delayed gamma rays are completely negligible. When calculated intensities of neutrons from $D(\gamma, n)$ reactions in ordinary water (cf. KAPL-M-JRS (1960)) are applied to our "Yankee-type" fuel element, the resulting neutron source is small compared with the intensity of neutrons from the plutonium. Detailed calculations are now being performed to obtain an estimate of the neutrons generated by α-emission and spontaneous fission of other nuclides which may be present in the irradiated fuel element.

In summary, these preliminary studies show that passive neutron counting may offer a direct, practical method for fuel element analysis. To implement this method, it is planned to investigate the response of various types of neutron detectors in high radiation fields, to continue isotope production calculations, and to verify present estimates of (α, n) yields from the metallic oxides of the plutonium isotopes.

SELF-INDICATION TECHNIQUES FOR DIA APPLICATIONS

The distinctive resonance structure in the neutron fission cross sections of the different fissile isotopes offers another promising method for nondestructive DIA (Detection, Identification and Analysis) applications. This resonance structure is most pronounced in the neutron energy range from 0.3 eV to roughly 10 keV, and the resonance peaks are typically orders of magnitude larger than the valleys.

One method of utilizing the resonance structure is to pass a beam of epithermal neutrons through a sample of fissile material and then to monitor this neutron beam with thin-foil fission detectors containing the same fissile isotope(s) as the sample. The sensitivity of this method depends on using the same fissile materials in the fission detectors as are under investigation in the sample, since the selective resonance absorption in the sample is amplified by the (n, f) resonance reaction in the fission foil with the same resonance structure (thus the term "self-indication").

Some preliminary measurements have been performed to evaluate this method for DIA applications. A schematic diagram of the experimental arrangement is shown in Fig. 3.

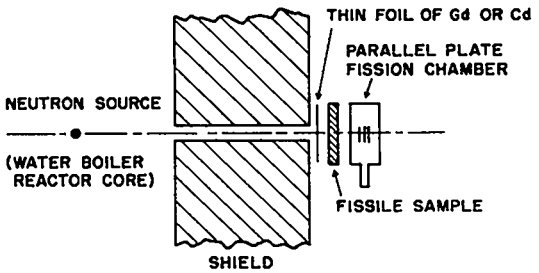

Fig. 3. Experimental setup for self-indication method.

The collimated neutron beam from the LASL Water Boiler Reactor was first passed through a foil of Gd or Cd to remove the thermal neutrons. The epithermal neutron beam then passed through a fissile sample (^{235}U or ^{239}Pu) and the parallel plate fission chamber which contained back-to-back foils of ^{235}U and ^{239}Pu. The fission rates in the two halves of the detector were recorded both with and without the fissile sample in the beam.

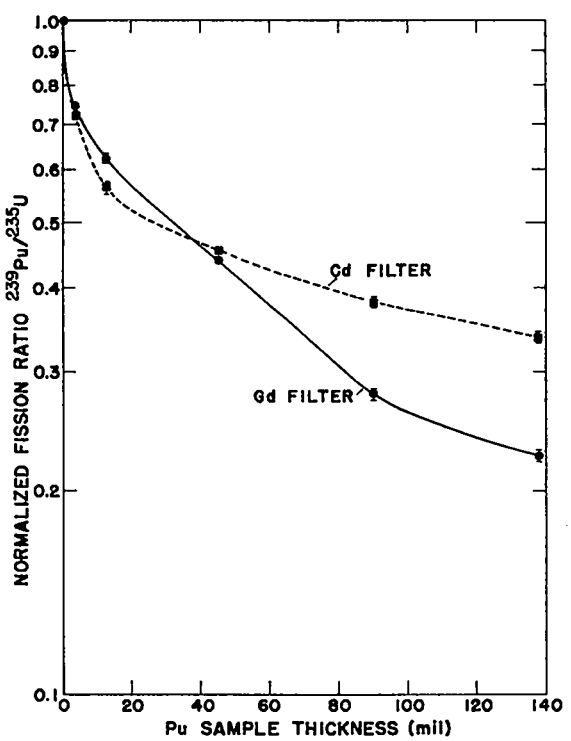

Fig. 4. $^{239}Pu/^{235}U$ counting rate ratio versus Pu sample thickness, basic data for self indication method.

Shown in Fig. 4 is a plot of the counting rate ratio of ^{239}Pu and ^{235}U in the fission chamber as a function of the Pu sample thickness. The solid curve corresponds to using a 1-mil-thick Gd filter, and the dashed curve corresponds to using a 20-mil-thick Cd filter. The thickness of the metallic Pu samples ranged from approximately 4 mil to 280 mil, giving a maximum discrimination ratio (^{235}U fission rate/^{239}Pu fission rate) of over 5.

When ^{235}U samples of corresponding thicknesses were inserted in the neutron beam, there was little change in the observed counting ratio ($^{235}U/^{239}Pu$) in the fission chamber.

Calculations of the resonance integrals of ^{235}U and ^{239}Pu have indicated that the observed discrimination ratio should be increased by roughly a factor of two if a thicker Gd foil were used. Accordingly, a 4-mil Gd foil has now been ordered.

The source of neutrons for practical isotope assay applications of this resonance self-indication method could be a moderated radioactive source such as ^{252}Cf, an accelerator-produced neutron source, or a simple low-power reactor (quite conveniently accessible in nearly all regions of the world), as was used in the preliminary experiments described above. For a hot fuel element it may be possible to use available (γ, n) reactions as source neutrons for the self-indication method; however, initial calculations have indicated that the neutron intensity would be low, though not necessarily prohibitive, for DIA applications.

In future work at LASL on self-indication methods, it is planned to obtain epithermal neutrons using 14 MeV (D, T) neutrons which have been slowed down in a moderator. Computer calculations using a DTF (neutron transport) code are being performed in order to optimize the thickness and choice of material(s) for the moderating assembly.

TRANSPORT THEORY CALCULATIONS AND ANALYSIS TECHNIQUES

The Los Alamos DTF-IV neutron-transport code (cf. LA-3373) is being used to calculate the leakage spectrum of neutrons produced by various moderator configurations surrounding the 14 MeV (D, T) neutron source at the Cockcroft-Walton Accelerator. Such calculations are expected to delineate a near-optimum moderator configuration in which the maximum number of neutrons are slowed down into the energy region from about 0.3 eV to a few hundred keV. Neutrons in this resonance energy range are of particular interest for practical DIA applications, such as the self-indication techniques described earlier in this report. A library of group-averaged cross sections is being built up for use with DTF-IV in projected computer studies supporting the N-6 safeguards research program.

Studies on Time Analysis of Kinetic Response Data

With reference to the area analysis or time-bin method of determining isotopic composition from kinetic response, it is clear that more detailed time-decay data should permit more accurate isotopic assay. Viz, detailed multichannel decay-vs-time data is more sensitive to intrinsic differences in the delayed neutron decay curves for the various isotopes, and therefore should lead in turn to a more accurate determination of relative isotopic abundance. However, the question arises whether in practice a given improvement in accuracy is worth the accompanying complications in data acquisition and analysis when using the detailed multichannel approach.

This problem has been formulated mathematically, coded in Fortran IV, and calculations of relative isotopic abundances and their propagated errors in representative systems have been performed on a CDC 6600 computer (cf. Report N-6-1009).

For the cases studied, the calculations show typically an 8% reduction in the error on relative isotopic abundance when the number of time bins is increased from the minimum of 2 to 3. In the limit of a large number of time bins (typically 50 or more, as in detailed multichannel analysis) there is an indicated decrease in error of about 13% as compared to the simple two-time-bin case. Thus, on the basis of this study it would appear that the detailed multichannel approach may not always offer a sufficient improvement in assay accuracy to warrant the accompanying complexities of data acquisition and analysis, and the simpler two-time-bin approach may indeed be preferable for many practical applications where maximum assay accuracy is not essential.

CW ACCELERATOR; EXPERIMENTAL USE AND FACILITY DEVELOPMENT

During the fourth quarter of 1967, operating time on the N-6 Cockcroft-Walton accelerator was devoted primarily to nuclear safeguards research, including instrumentation and detector development.

In addition to routine maintenance, increasing problems with spurious pulses necessitated accelerator shutdown early in the quarter for replacement of the einzel lens and dismantling the ion beam deflection chamber in order to replace insulators for the pre-acceleration deflection plates. The spurious pulses have been attributed to electrical breakdown across the deflection-plate insulators. The zero-degree beam

port of the CW accelerator is being activated to permit greater experimental flexibility; this should also provide better beam stability under long pulse operation (pulse width > 0.1 sec), since the accelerator high voltage power supply regulation is not adequate to smooth out completely the voltage transients associated with switching from no beam to full beam and vice versa. These voltage transients in turn cause a momentary decrease in intensity of the deflected beam thru the 25° ports, but not in the undeflected beam thru the 0° port.

Standard operating procedures for the N-6 Cockcroft-Walton have been formulated. More rigid and formalized procedures have become necessary as the number of qualified accelerator operators has increased.

Group N-6 is presently conducting a survey of commercially-available small portable pulsed neutron sources (D, T and D, D reactions). Thus far, inspection visits have been made to Accelerators, Inc., and Texas Nuclear Corporation, both of Austin, Texas. A similar inspection visit will be made in early February, 1968, to the third U. S. supplier of intense neutron sources, the Kaman Nuclear Corporation of Colorado Springs, Colorado.

Power, cooling, and safety-interlock systems are nearly completed in preparation for the mid-January installation of Accelerator I, which is being supplied on an interim-loan basis by Picker Nuclear (marketing firm for Accelerators, Inc.). The Accelerator I will undergo extensive and sustained performance evaluations in connection with the N-6 safeguards research program, and the development of a compact, portable isotopic assay system for in-the-field applications.

DENSE PLASMA FOCUS SOURCE

Construction of the N-6 Dense Plasma Focus (DPF) source has been completed and the device is undergoing detailed performance evaluations as a source of pulsed neutrons for nuclear safeguards research.

General features of the DPF source are shown in Figure 5. The electrical energy storage and switching gear are seen at the lower right, the vacuum and gas handling apparatus at the left, and the DPF discharge tube ("neutron gun") is the prominent aluminum cylinder protruding from the top of the system. The complete assembly is mounted on a steel frame fitted with wheels for mobility and convenience.

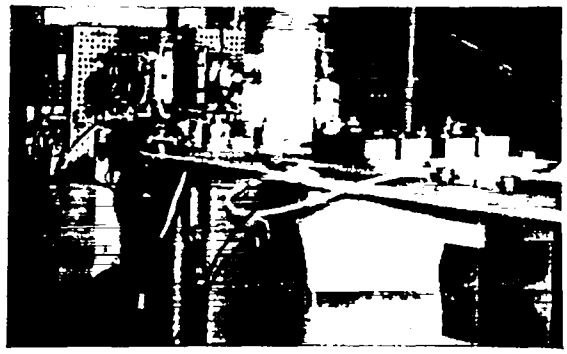

Fig. 5. The N-6 Dense Plasma Focus (DPF) pulsed neutron source; general view.

To date there have been four neutron-producing shots with yields varying between 2.5 and 4×10^8 neutrons per shot. A total of seventy test shots have been fired. The conditions necessary for production of neutrons in the DPF appear to be extremely exacting, and it has proved necessary to develop rather extensive diagnostic techniques to permit precise "tuning" of the device. Considerable effort has been spent in developing instrumentation for photographing and interpreting the simultaneous single voltage and current pulses associated with each shot. For example, special high-speed high-voltage, and high-current probes had to be

perfected and adapted to the system to provide the required diagnostics capability.

Performance of the strip-line dielectric switches has fallen somewhat short of our original "great expectations." There appears to be a considerable nonuniformity in the mylar dielectric layer which causes breakdown at one or two points rather than at many points as originally intended. As a result, there occurs excessive pitting of the copper plates (between which the dielectric switch is mounted), and this is believed to be responsible for the shot-to-shot uncertainty in the inductance of the switch. As a result of these and other frustrating difficulties with dielectric switches, a possible conversion to ignitrons or vacuum-spark-gap switching is being seriously considered for the DPF.

In more recent DPF shots there has also arisen a problem with breakage of the coaxial "glass hat" insulators, which is believed due to transverse motion of the center electrode during the discharge. Such motion could be caused by axial asymmetry of the discharge, which in turn may result from a noticeable imperfection in the axial symmetry of the outer electrode.

Steps are being taken to correct these problems and to resume, as soon as possible, the performance evaluation and yield optimization of the DPF source.

INSTRUMENTATION AND DETECTOR DEVELOPMENT

The high-efficiency "long counter" recently developed in Group N-6 (cf. LA-3802-MS) was found to have an unacceptably high background when used in precise measurements of absolute delayed neutron yield at 14 MeV incident neutron energy. After elimination of various possible sources of background, the major contribution to background is believed due to n, p reactions on oxygen in the epoxy of the boron-loaded epoxy counter shield (epoxy contains about 20% O_2 by weight).

The subject n, p reactions are:

a) $^{16}O(n, p)\ ^{16}N$, which yields a high energy gamma ray ($E_\gamma \sim 6$ MeV, 7.1 sec half-life) which in turn produces neutrons by γ, n reactions on materials such as ^{13}C, ^{2}D, ^{17}O in the counter and shield;

b) $^{17}O(n, p)\ ^{17}N$, which yields a delayed neutron following β-decay of the ^{17}N (4.1 sec half-life).

Both (a) and (b) are threshold reactions, with threshold neutron energies of ~ 11 and ~ 10 MeV, respectively. The delayed neutrons following ^{17}N β-decay are believed to be the principal contributors to the observed background. The original epoxy material used in the N-6 long-counter shield has been replaced by an oxygen-free boron-loaded paraffin shield; in addition, a new long counter which is free of both oxygen and carbon is being constructed for test purposes using ZrH as the moderating material.

High Efficiency "Slab" Detector

A high efficiency neutron detector consisting of thirteen 20-inch long, 1-inch diameter ^{3}He proportional counters imbedded in slabs of moderating material has been designed. Two such detectors are presently being fabricated for use as coincidence counters to study the feasibility of detecting fission neutrons in a background of (α, n) neutrons. These detectors will also be used in other applications requiring large-area or large-solid-angle neutron detection.

Twenty-six 6-atmosphere ^{3}He counters for use in slab detectors (and the 4π-detector--see below) have been received and tested. These detectors are matched to within 3% in pulse height, and have a resolution of 7% to 8% for

thermal neutrons.

The necessary electronic circuitry for coincidence counting with these detectors is presently being developed.

4π Neutron Detector

Design of the 4π Neutron Detector described in the previous N-6 progress report (LA-3802-MS) has been completed in detail, and parts are presently being fabricated. Evaluation of high pressure BF_3 counters for use in this detector has been completed, and forty 20-inch long, 1-inch diameter counters are on order. The 20-inch ^3He counters described above will also be used in this detector.

Neutron Spectrometers

A survey of various types of neutron spectrometers usable over the energy range of delayed fission neutron spectra is presently underway. Preliminary evaluation of a solid-state ^3He "sandwich" detector indicated severe background problems from gamma rays. For crude spectrometry, however, a similar sandwich-type detector using ^6Li rather than ^3He may effectively override relatively high gamma backgrounds, since the neutron detection reaction, $^6Li(n,\alpha)^3He$, has a positive Q value of 4.78 MeV compared to Q = + 0.75 MeV for the $^3He(n,p)^3H$ reaction. One strong motive for neutron spectrometer development is the prospect of increased isotope discrimination factors for safeguards inspection applications, based on both time and energy discrimination in kinetic response measurements.

^{238}Pu-Li Neutron Source

In conjunction with Group CMB-11 at LASL, a ^{238}Pu-Li neutron source was constructed which has a total α, n source strength of about 6×10^5 neutrons per second. The half-life of ^{238}Pu for α emission is 89 years, and the energy spectrum of the emitted neutrons approximates the delayed neutron spectrum to about as well as the latter spectrum is known. The availability of such a convenient, portable, steady-state (rather than transient) source with its unique spectral feature should be especially useful in DIA research, including development of a spectrometer to measure delayed neutron group spectra.

Variable Time-Base Generator

An electronic channel-advance unit has been designed and built which allows groups of channels in a multichannel analyzer, when operated as a multiscaler, to be advanced at different rates. The time from one channel to the next (i.e., channel width) can be varied from 10 μsec to 9.9 sec, and the number of channels with a pre-determined width setting can be varied from 1 to 999. Two different groups of channels, each of variable width, have been constructed and tested thus far; the complete channel-advance unit has been designed for future expansion to as many as eight different groups of channels, each with independently-variable channel widths. This new unit will enable the collection of pulsed neutron kinetic response data in nearly any desired distribution of channel-width versus time, e.g., to permit examination of both the short- and long-lived delayed neutron decay components to the same statistical accuracy.

Data Acquisition Equipment

Various types of small computers and pulse analysis equipment are presently being investigated for the development of extended on-line data acquisition and analysis capabilities. A small pulse height analyzer was ordered some time ago to augment present capabilities for multichannel time and pulse height analysis. However, the manufacturer has experienced unavoidable delays in delivery, and a similar instrument has been obtained on an interim loan

basis. The improved read-out capability of this instrument has greatly facilitated the acquisition and analysis of kinetic response data.

OTHER CONTRIBUTIONS TO NUCLEAR SAFEGUARDS RESEARCH AT LASL

Water-Boiler Reactor Irradiations (P-2)

Irradiations at the LASL Water Boiler Reactor, provided by Group P-2, were used to investigate fissile isotope discrimination factors available using new resonance self-indication techniques being developed by Group N-6.

Mathematical Simulation of Space-Dependent Delayed Neutron Kinetic Response (T-Division)

A mathematical analysis was carried out, using the techniques of singular perturbation theory, to show the mathematical rigor of the zero-prompt-lifetime approximation as applied to the transport theory calculations of the delayed neutron response in small samples (cf. LA-3732-MS). The zero-prompt-lifetime approximation was shown to be part of a consistent formalism which yields a solution for short as well as long times. This approximation will form the basis for using time independent two-dimensional S_n codes for the detailed calculation of delayed neutron kinetic response to pulsed neutron interrogation.

^{238}Pu-Li Source Fabrication (CMB and CMF)

Several LASL groups contributed to the fabrication of the ^{238}Pu-Li source to be used for detector calibration, spectrometer development, and possible future DIA applications:

CMF-4 - Prepared the ^{16}O isotope;

CMB-1 - Prepared the $^{7}Li_2{}^{16}O$ compound;

CMB-11 - Furnished the ^{238}Pu and assembled the source;

CMB-3 - Contributed to calculation of expected source yield.

Tritium Target Preparation for N-6 Cockcroft-Walton Accelerator (CMF-4)

Assistance in Preparing Group Averaged Cross Sections for Use with DTF Program (T-7)

PUBLICATIONS

1. Nondestructive Detection, Identification, and Analysis of Fissionable Materials, WASH-1076, 150 (1967); also WASH-1077 (Confidential, RD) (1967).

2. Neutron Radiative Capture Cross Sections for ^{23}Na, ^{55}Mn, ^{115}In, and ^{165}Ho in the Energy Range 1.0 to 19.4 MeV, Physical Review 163, 1299 (1967).

3. Fast-Neutron Bombardment of ^{64}Ni and the Decay of ^{61}Fe, Physical Review 161, 1118 (1967).

4. Activation Cross Sections for the ^{19}F(n, 2n)^{18}F, ^{23}Na(n, 2n)^{22}Na, ^{55}Mn(n, 2n)^{54}Mn, ^{115}In(n, 2n)^{114m}In, ^{165}Ho(n, 2n)^{164m}Ho, ^{115}In(n, n')^{115m}In, and ^{27}Al(n, α)^{24}Na Reactions, Physical Review 163, 1308 (1967).