LA-3974-MS

LOS ALAMOS SCIENTIFIC LABORATORY
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Pulsed Neutron Research for Nuclear Safeguards

Program Status Report

April-June, 1968

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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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PULSED NEUTRON RESEARCH FOR NUCLEAR SAFEGUARDS

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NEUTRON INTERROGATION TECHNIQUES FOR NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS

The delayed neutron kinetic response measurements reported in previous progress reports have been extended to include the R_{f-} and R_{f+} isotope discrimination ratios[1] for ^{239}Pu-^{238}U mixtures. The energy of the interrogating neutrons was 14 MeV, and the irradiation procedure was similar to that previously described in LA-3921-MS. Some measured ratios and corresponding theoretical predictions (for fission spectrum neutrons)[1] are given in Table I. As in previous measurements, R_{f+} ratios were observed to decrease as the time fiducial, f, was decreased from the maximum f value used (f = 150 sec); it is difficult to extend f much beyond 150 sec due to the neutron background from spontaneous fission of ^{240}Pu.

TABLE I

R_{f-} AND R_{f+} ISOTOPE DISCRIMINATION RATIOS FOR ^{238}U AND ^{239}Pu

$E_n \simeq 14$ MeV (D, T reaction; Irradiation Time: 0.2 sec; Total Counting Time: 200 sec

	$R_{f-}\left(^{238}U/^{239}Pu\right)$ f = 0.10 sec	$R_{f+}\left(^{239}Pu/^{238}U\right)$ f = 150 sec
Measured	1.83 ± .09	2.73 ± .33
Theoretical	2.0	2.80

It is clear from Table I that the combination of R_{f-} and R_{f+} measurements (both obtained during the same pulsed neutron irradiation) provide an overall measurable isotope discrimination factor of 5 or greater between ^{238}U and ^{239}Pu. Such clear-cut, incisive discrimination between these two major isotopic constituents of future fast breeder reactor fuels seems destined to assume considerable practical importance in the kinetic response method of nondestructive assay of these breeder fuels. Further kinetic response measurements on prototype FBR fuels are planned as part of the N-6 safeguards program during FY 69 and subsequently.

Nondestructive Assay of Reactor Fuel Elements Using a "Fast-Tailored" Neutron Beam

In the practical assay of many types of reactor fuel elements it is desirable to interrogate with neutrons that have energies high enough to be very penetrating, but which lie below the thresholds (several MeV) for (n, p), (n, 2n), and (n, n'f) reactions. This desired result can be readily achieved by "spectrum tailoring" of 14 MeV (D, T) neutrons.[a] The (D, T) source neutrons are moderated by an 8"-thick Pb slab to reduce the average neutron energy from 14 MeV to a few MeV.

Delayed neutron response measurements have been performed on an MTR-type fuel element mock-up to determine the absolute amounts of fissionable material in elements with various loadings. The mock-up element has the same maximum loading density and uranium-to-aluminum ratio as found in a typical MTR fuel element. Details of the mock-

[1] G. R. KEEPIN, "Nondestructive Detection, Identification, and Analysis of Fissionable Materials," Los Alamos Scientific Laboratory Report, LA-3741 (1967).

[a] E. g., from Accelerator I; see later section on Accelerators and Neutron Sources.

up element are given in the last quarterly progress report (cf. LA-3921-MS). For the new measurements, using a "fast-tailored-spectrum" beam, the geometry and irradiation procedure was similar to that previously described (cf. LA-3921-MS) using Accelerator I and the N-6 slab detector. The production rate of the accelerator neutron source was monitored using a ^{238}U fission chamber positioned near the (D, T) source, and the relative delayed neutron counting efficiency was calibrated using the fully loaded fuel element (18 plates) in the normal interrogation position. The delayed neutron counting rates were typically greater than 15,000 cpm, thus counting statistics of better than 1% could be obtained in a few minutes.

The results of the measurements are given in Table II, where it is apparent that the mass of fissionable material determined from the delayed neutron response agrees closely (average deviation = 0.51%) with the actual mass. A particularly significant result of this work is the fact that absolute total amounts of fissile material can be measured to within 1% accuracy, for large deviations, 20% or greater, from the standard calibration mass. Thus the delayed neutron method of nondestructive assay seems very well suited to the determination of absolute amounts of fissionable materials in various types of reactor fuel elements. The application of this technique to other types of reactor fuel elements (thermal, intermediate, and fast reactors) is foreseen in future safeguards R&D work at LASL.

TABLE II

DELAYED NEUTRON ASSAY OF ABSOLUTE AMOUNTS OF FISSIONABLE MATERIAL IN MTR-TYPE FUEL ELEMENTS

"Fast-Tailored" Neutron Beam ($\bar{E}_n \sim 3$ MeV)

Actual Weighed ^{235}U Content in Element (grams)	Delayed Neutron Assay Determination (grams)	Deviation
339.95 (fully loaded)	Calibration Point	---
321.18	321.2	- 0.03%
302.95	305.2	+ 0.75%
284.20	284.6	+ 0.13%
265.45	268.4	+ 1.12%
	Average Deviation:	0.51%

MSRE Salt Assay by Delayed Neutron Kinetic Response Methods

At the request of Oak Ridge, a series of experiments is in progress at LASL to develop neutron kinetic response methods for nondestructive, quantitative assay of ^{233}U-Th fuel-salt mixtures to be used in the Oak Ridge molten salt reactor experiment (MSRE reactor). A typical MSRE salt contains Be, F, and Li in addition to the fissionable nuclides, ^{233}U and ^{232}Th. Thus far, three 1-1/2" diameter samples, each weighing about 40 grams, loaded only with Th, have been furnished by ORNL; samples containing both ^{233}U and ^{232}Th are expected in the near future. Preliminary measurements are underway using LASL-fabricated metallic ^{233}U foils and discs in combination with the

ORNL Th salt.

Neutron kinetic response to 14 MeV neutron interrogation was measured for N-6 standard 2" diameter discs of ^{233}U, ^{232}Th, and ^{235}U using a one second irradiation and a 20 second counting period. (The ^{235}U standard response curve was used for comparison with earlier measurements.) Experimental results thus far indicate that ^{233}U-Th mixtures can be isotopically analyzed just as accurately as has already been demonstrated for ^{235}U-^{238}U composites (cf. LA-3859-MS). A comparison of the kinetic response curves for the metallic Th sample and the ORNL Th salt (42.7% Th by weight) showed no difference in their time decay.

Because MSRE fuel contains only a few percent ^{233}U, the presence of ^{233}U will tend to be masked by the much larger amount of Th. To enhance the delayed neutron response of ^{233}U over that of Th, various W-Pb-CH_2 spectrum-tailoring assemblies were placed around the accelerator target to moderate the 14 MeV (D, T) source neutrons below the Th fission threshold (~ 2 MeV).[b] A double-sided fission chamber containing ^{233}U and Th was used to measure the enhancement of ^{233}U/Th fission ratio resulting from the tailored source spectra. Table III lists the ratio of fissions for various tailoring conditions (i. e., moderator configurations).

TABLE III

$^{233}U/^{232}Th$ FISSION RATIOS FOR VARIOUS SPECTRUM-TAILORING CONDITIONS

Spectrum Tailoring Condition	$^{233}U/^{232}Th$ Fission Ratio
14 MeV	6
2" W - 4" Pb	26
2" W - 4" Pb - 2" CH_2 - .030" Cd	184
2" W - 4" Pb - 2" CH_2	1302

[b] Cf. calculations of tailored neutron leakage spectra from various moderator configurations, LA-3921-MS, pp. 11-13.

The ratio of delayed neutron yields per fission must be factored into these fission ratios to obtain the expected enhancement in ^{233}U/Th delayed neutron yield ratio. It should be noted here that delayed neutron fission yield ratios are essentially constant, independent of incident neutron energy--a fact of considerable practical importance in kinetic response techniques for isotopic assay. Accordingly, we assume a delayed neutron yield ratio,

$$\frac{n/F(^{233}U)}{n/F(Th)} = 0.13,$$

as indicated by recent LASL measurements[2] at 3 MeV, and we further assume the ^{233}U/Th fission ratios given in Table III for the W-Pb-CH_2 moderating assemblies with and without Cd (i. e., 184 and 1302, respectively). The product of the above ratios gives a ^{233}U/Th delayed neutron response ratio between 24 and 170 (corresponding to tailored spectrum assemblies with and without Cd). Considerable further improvement in ^{233}U/Th fission ratios is expected using more nearly optimum moderator configurations.

Neutron Interrogation and Delayed Neutron Measurements Applied to Scrap Assay

As has been pointed out in previous progress reports, the presence of hydrogenous material in fissile scrap configurations yields greatly enhanced delayed neutron response. One method of reducing this response is to normalize observed delayed neutron counts to fission counts in a fission chamber monitor containing the fissile species of interest. To implement this approach for practical assay applications--and to provide data on the spatial (one-dimensional) response as well--the delayed

[2] C. F. MASTERS, M. M. THORPE, and D. B. SMITH, "Measurement of Absolute Delayed Neutron Yields from 14 MeV Fission," Trans. Am. Nucl. Soc., 11, 179 (1968).

neutron counting rate from a 20-gram disc sample of ^{235}U per average fission monitor count was measured as a function of position within a 4"-thick by 12" x 12" polyethylene block. The resulting data are labeled "Primary Response" in Fig. 1. In practical assay applications the sample may be inverted with respect to target and detector positions (i. e., sample rotated thru 180°) and the measured response averaged. Thus simple rotation of the sample would give the results shown in Fig. 1 by the curve marked "Averaged Response". For this particularly simple arrangement of target and detector and choice of slab thickness, the response for all combinations of materials and material densities so far considered[c] is expected to lie between the averaged response and the data obtained with no hydrogenous material present--a nominal variation of less than a factor of two.

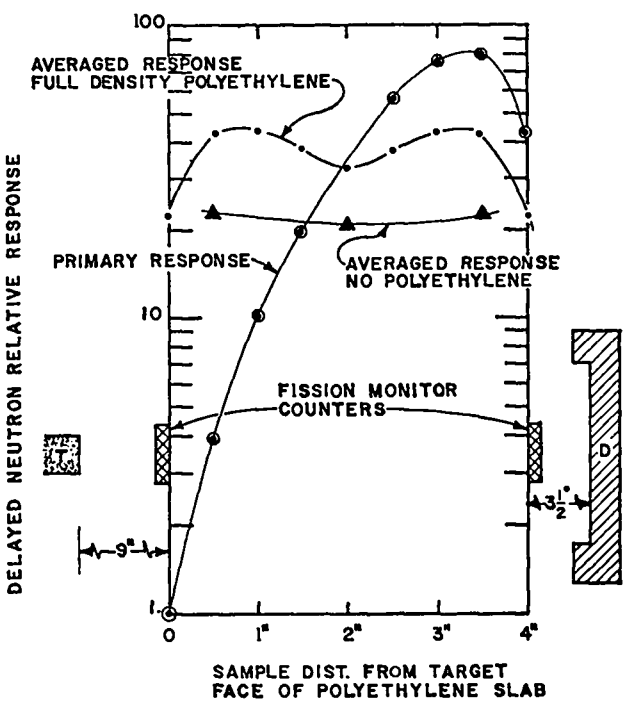


Fig. 1. Delayed Neutron Relative Response. (T - 14 MeV neutron source target; D - Cadmium covered slab neutron detector.)

[c]See "Neutron Response of Simulated Scrap Barrels," LA-3921-MS, January-March 1968.

Thus the normalization of delayed neutron response to an appropriate fission counter monitor provides a most effective means of compensating for the presence of hydrogenous materials which may be present in practical scrap configurations. At present, both calculations (to serve as a guide for further measurements) and experiments are being extended to more complex arrangements of sources and detectors, as well as different slab thicknesses, and to neutron source spectra other than 14 MeV. The direct goal of these studies is, of course, the development of reliable, accurate techniques for nondestructive quantitative assay of a wide range of practical scrap configurations.

Neutron Response of Simulated Scrap Barrels to Fast-Tailored-Spectrum Interrogation

The neutron transport study of simulated scrap barrels, begun early in 1968, has been extended to investigate the delayed neutron response to a fast tailored source. The tailored source spectrum used in the calculations was produced by tungsten moderator configuration No. 5 described in the last progress report (10 cm thick W around a 14 MeV source). This spectrum extends from $\sim 5 \times 10^{-4}$ MeV to ~ 5 MeV and peaks at ~ 0.1 MeV. All other parameters were the same as described in LA-3921-MS: one gram of ^{235}U was distributed at the center of the simulated barrel and also at r = 15 cm (barrel half-radius). In separate calculations the barrel was filled with CH_2, C, Fe, and Pb at several densities. The results of this new series of response calculations using a fast-tailored spectrum are shown in Fig. 2.

As in the case of the 14 MeV source, barrels containing almost any common scrap material other than hydrogenous material and weighing up to a few hundred pounds are essentially transparent to this fast-tailored-spectrum neutron source. The sensitivity of kinetic response to ^{235}U position is slightly less for the fast tailored source than for the 14 MeV source. Again it is most important to

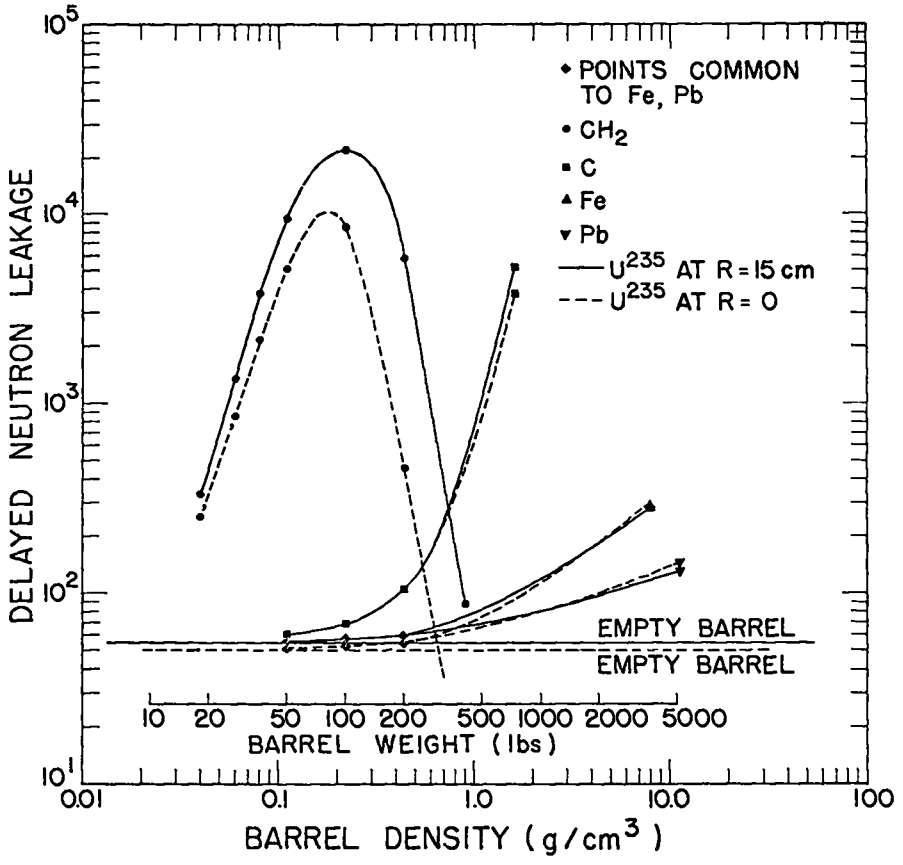


Fig. 2. Delayed neutron response of simulated scrap barrels to a "fast-tailored" neutron source. The simulated barrels contain one gram of ^{235}U distributed at indicated radii within representative moderator or matrix materials. (The base lines labeled "empty barrel" refer to one gram of ^{235}U in a barrel containing no other material.)

note that, except for hydrogenous material, measured delayed neutron response is relatively independent of the position or distribution of the ^{235}U in the barrel.

Figure 3 shows the response of one gram of ^{235}U at r = 15 cm in a matrix of CH_2 for the three types of neutron source spectra for which calculations have been performed: 14 MeV, fast tailored, and 1/E tailored. It is seen that the region of densities over which hydrogenous material produces increased response (compared to "empty" barrel response) is greater for the fast tailored source than for the 14 MeV source, and still greater for the 1/E tailored source. This is due to increased thermalization of source neutrons in the case of the two "softer" sources. The magnitude of the response for the tailored sources is also greater than for the 14 MeV source.

A somewhat "harder" tailored source (e.g., that produced by a lead moderator) may preserve more of the penetrability of the 14 MeV neutrons than does the present W-moderated fast tailored spectrum, and still suppress possible background reactions which can occur between ~8 and 14 MeV. Such alternative moderators for fast tailored spectra will be explored (both calculationally and experimentally) during the next quarter.

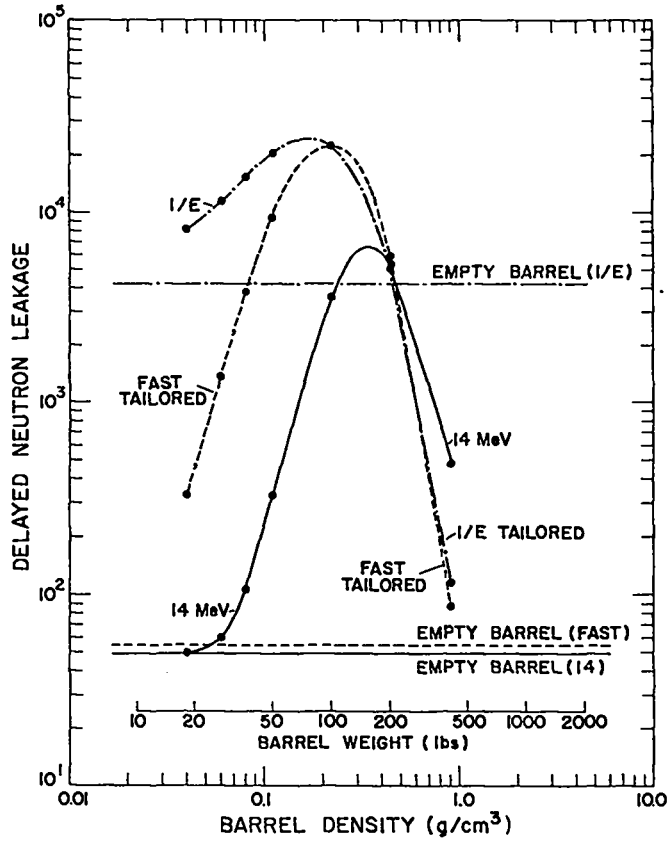


Fig. 3. Comparison of the delayed neutron response from one gram of ^{235}U distributed at r = 15 cm in a simulated scrap barrel containing CH_2 for three neutron source spectra.

NEUTRON MULTIPLICATION EFFECTS IN ACTIVE INTERROGATION METHODS

The neutron multiplication measurements described in the previous quarterly progress report (LA-3921-MS) have been extended to include samples of ^{239}Pu (94% enriched). In addition, attempts were made to calculate the delayed neutron multiplication curves presented in this and the previous progress report.

For the 14 MeV neutron irradiations, the 2" diameter disc-shaped samples were placed 3" from the (D, T) neutron source and a high efficiency long counter was positioned 11" from the samples to detect the delayed neutron response. The Cockcroft-Walton accelerator was repetitively pulsed (0.050 sec irradiations followed by 0.050 sec counting intervals), and the α-particles from the T(d, n)α reaction were counted to monitor the source neutron production rate. The sample thicknesses varied from 0.010" to 0.276". In order to determine the sensitivity of the multiplication effect to the angle of the incoming 14 MeV neutrons, the samples were also irradiated at a distance of 1" from the (D, T) neutron source. This change in position did not significantly alter the multiplication results.

The preliminary results of the measurements

for ^{239}Pu are shown in Fig. 4. For thicknesses less than approximately 0.080", the curve increases almost linearly. A least-squares fit of the initial slope to a straight line yields a slope of 0.38%/mil. This initial multiplication is roughly a factor of 1.5 greater than for ^{235}U and 4.4 greater than for ^{238}U.

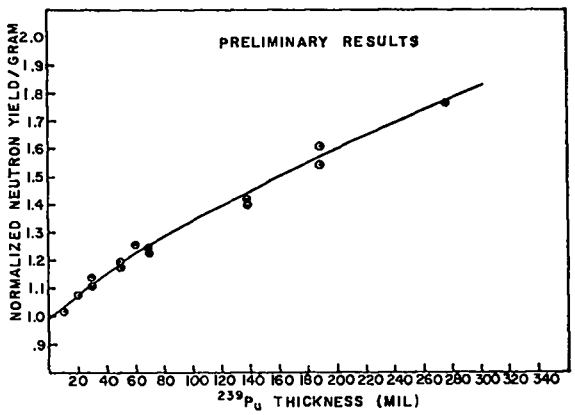


Fig. 4. Delayed neutron yield (multiplication) from 14 MeV neutron irradiation of ^{239}Pu (94% enriched).

Calculations of this multiplication effect have been performed using different approaches to investigate the relative accuracies and limitations of the methods. Of primary interest is the slope of the curves at small h/D (height-to-diameter) ratios where the disc sample can be approximated by a thin slab of infinite extent. The experimental data were analyzed by the method of Maximum Likelihood to determine the best fit of the initial slope and the uncertainty of the fit. These results are presented in Table IV.

TABLE IV

INITIAL SLOPES, DELAYED NEUTRON MULTIPLICATION

Isotope	Experimental Slope	"Collision Probability" Calculated Slope
^{238}U	.087 (1 ± .05)%/mil	.074 %/mil
^{235}U	.248 (1 ± .02)%/mil	.136 %/mil
^{239}Pu	.38 (1 ± .06)%/mil	.163 %/mil

A "collision probability" model has been developed to calculate the slope. This simple model utilizes only three energy groups and exponential expressions for escape probabilities. The formula derived for the slope, using energy groups at 14 MeV, 3 MeV, and 0.5 MeV, is

$$\text{Slope} = \left(\eta \Sigma_{ne}\right)_{14} \frac{\left(Y \Sigma_f\right)_3}{\left(Y \Sigma_f\right)_{14}} - \frac{\left(\Sigma_{ne}\right)_{14}}{2} + \left[\Sigma_f (\bar{\nu} - 1)\right]_{0.5}$$

where: η = number of neutrons per nonelastic collision, Σ_{ne} = macroscopic nonelastic cross section, Y = delayed neutron yield per fission, Σ_f = macroscopic fission cross section, $\bar{\nu}$ = number of prompt neutrons per fission, and the subscripts designate neutron energy. The last term in the formula, which is the multiplication of the delayed neutrons escaping the sample, is significant only for fissile material, e.g., ^{235}U and ^{239}Pu, since the average delayed neutron energy is 0.5 MeV. The results obtained using this formula are listed in the last column of Table IV. The discrepancies with the experimental data are probably due to the crude approximations involved in the derivation of the formula.

Another calculational approach has been to obtain the slope using the Zero Prompt Lifetime Approximation in conjunction with the discrete-ordinates transport theory code. These preliminary results have indicated a strong dependence of the calculated slope on the order of angular quadrature. More refined slope calculations are in progress.

Further studies will include an investigation of this angular quadrature effect as well as an extension of multiplication calculations and measurements to other isotopes, including ^{233}U and ^{232}Th.

2DF--A Two-Dimensional Neutron Transport Code

A two-dimensional version, called 2DF, of the Los Alamos S_n neutron transport code is now operational on the CDC-6600 computer. A program is being written to calculate the first collision source for a general two-dimensional problem and to punch source cards for use in 2DF. It is hoped that the use of this source will reduce the ray effects[3] which can occur in two-dimensional S_n calculations containing a point source.

ABSOLUTE DELAYED NEUTRON YIELDS

The program of absolute delayed neutron yield measurements from 3.1 and 14.9 MeV neutron-induced fission of the major fission species was completed during the second quarter of 1968. The experimental methods used have been described in detail in previous N-6 progress reports. The results for the five most common fissionable materials are summarized in Table V. The indicated yield uncertainties (standard deviations) in Table V are approximately 10%.

These measurements show clearly that the delayed neutron yield of every isotope studied decreases significantly in going from 3.1 MeV to 14.9 MeV neutron-induced fission--a result which is expected from the known behavior of fission mass and charge distributions as a function of incident neutron energy in neutron-induced fission. As noted previously, these results stand in direct contrast to previous measurements by other workers both in the U. S. A. and the U. S. S. R.

TABLE V

MEASURED ABSOLUTE DELAYED NEUTRON YIELDS

(Delayed Neutrons/Fission)

Indicated uncertainties are standard deviations. All yield values have been corrected to 100% isotopic purity, except in the case of ^{233}U (having the following sample composition: 98.5% ^{233}U; 1.2% ^{234}U; 0.3% ^{238}U).

Isotope	14.9 MeV Fission Yield	3.1 MeV Fission Yield	Yield Ratio: $\frac{\text{3.1 MeV}}{\text{14.9 MeV}}$
^{239}Pu	.0043 ± .0004	.0069 ± .0007	1.60 ± .09
^{233}U	.0043 ± .0004	.0077 ± .0008	1.77 ± .10
^{235}U	.0095 ± .0008	.018 ± .002	1.89 ± .11
^{238}U	.0286 ± .0025	.049 ± .005	1.71 ± .10
^{232}Th	.031 ± .003	.059 ± .006	1.92 ± .11

[3] K. D. LATHROP, "Ray Effects in Discrete Ordinates Equations," Nucl. Sci. Eng. 32, 357 (1968).

The indicated decrease in yield at 14.9 MeV is further confirmed by accurate relative measurements of delayed neutron yields at 3.1 and 14.9 MeV incident neutron energies. Essentially all systematic errors are eliminated in this relative measuring technique, wherein only the accelerator target is changed (from D to T) to obtain a direct yield comparison at the two incident neutron energies. Reasonable agreement is obtained between the present yield data at 3.1 MeV and the corresponding yield data of other workers, as summarized in Keepin's book.[4] Although the 3.1 MeV absolute yield values in Table V are some 10 to 20% higher than previous values, this difference may be due, at least in part, to differences in effective energies of the neutrons inducing fission.

A summary of the LASL delayed neutron absolute yield measurements was reported at the June, 1968, meeting of the American Nuclear Society in Toronto.[5] A complete paper on the LASL delayed neutron yield measurements is being prepared for publication in Nuclear Science and Engineering.

As noted previously, accurate absolute delayed neutron yields as a function of incident neutron energy are essential to the practical application of delayed neutron response techniques to nondestructive isotopic assay.

SELF-INDICATION TECHNIQUES FOR DIA APPLICATIONS

During the past quarter, several measurements have been performed to investigate the possibility of using the resonance self-indication technique for nondestructive assay of fissile materials. This technique utilizes the characteristic resonance structure in the neutron fission cross sections of the fissile isotopes. The basic principles of the self-indication method and the experimental setup have been previously given (cf. LA-3859-MS).

In the measurement, a collimated neutron beam from the LASL Water Boiler Reactor is passed through a Gd foil (4.5 mil) or a Cd foil (30 mil) to remove the thermal neutrons. Gd is used since its neutron cutoff energy is just below the peak of the 0.3-eV ^{239}Pu giant resonance. The epithermal neutron beam is then passed through the fissile sample under investigation and thence through a sandwich of three parallel-plate ionization chambers containing thin evaporated deposits of ^{239}Pu, ^{235}U, and ^{10}B. The counting rates in all three detectors were recorded both with and without the various fissile samples and absorbers in the beam. The cadmium ratio was approximately 40 for each of the reactions recorded by the ionization detectors.

The response of the fission detectors to the variation in the thickness of the corresponding fissile samples is shown in Fig. 5. These curves represent the transmission of neutrons through the samples weighted by the fission cross sections of the detectors. If there is no extraneous material in a fissile sample, then a simple transmission measurement of this type could be used to determine the sample thickness. The assay accuracy under these conditions would be quite good, since

[4] G. ROBERT KEEPIN, Physics of Nuclear Kinetics (Addison Wesley Publishing Company, Inc., Reading, Massachusetts, 1965).

[5] C. F. MASTERS, M. M. THORPE, and D. B. SMITH, "Measurement of Absolute Delayed Neutron Yields from 14 MeV Fission," Trans. Am. Nucl. Soc., 11, 179 (1968). This paper was cited in the ANS publication, Nuclear News, as "probably the most impressive new work reported" in the ANS technical session on Nuclear Data and Doppler Broading at Toronto.

an error of 1% in the measured response would correspond to errors of approximately 1% and 2.6% in the thickness determinations (in the region of ~ 20 mil) of ^{239}Pu and ^{235}U, respectively. With the present experimental setup, a measurement time of 2 minutes yielded counting statistics of better than 0.7%.

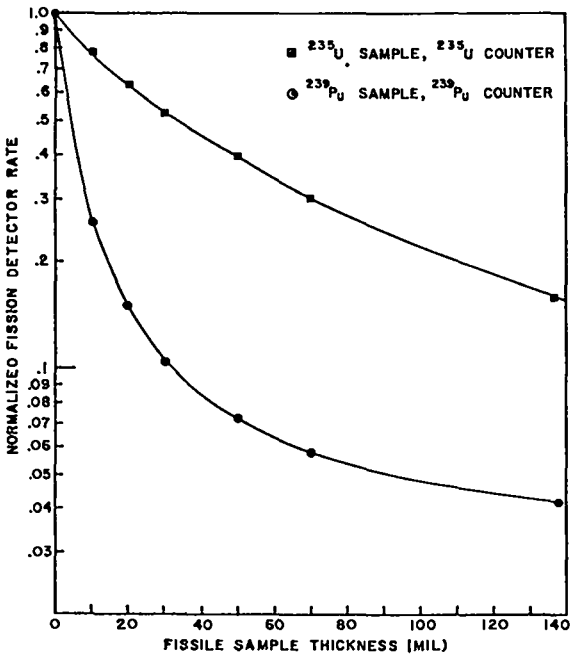


Fig. 5. Transmission curves for epi-gadolinium neutrons through ^{235}U and ^{239}Pu.

The counting rate ratios of the fission detectors to the ^{10}B detector for different thicknesses of Pu are shown in Fig. 6. A 4.5-mil Gd filter was used for this set of measurements. The upper curve corresponds to the response in the ^{235}U fission detector and the lower curve to the ^{239}Pu detector. Both curves have been normalized to the counting rate in the ^{10}B detector which makes the measured response relatively insensitive to extraneous materials mixed in with fissile sample, since both the ^{10}B and the fission response rates are decreased roughly the same amount by the extraneous material.

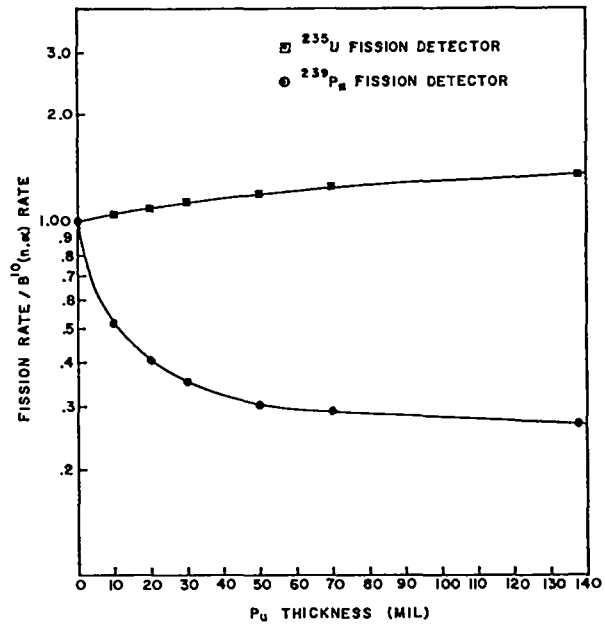


Fig. 6. Counting rate ratio of fission detectors to ^{10}B detector as a function of Pu sample thickness (using a Gd filter).

In order to investigate the effect that various extraneous materials have on the fission detector responses, a series of measurements were made wherein a Pu sample (0.010" thick) was sandwiched between relatively thick (~0.5") layers of common materials (Al, Pb, ^{238}U, ^{235}U, and CH_2). Table VI gives the thickness of the added materials along with the changes in the ^{239}Pu fission detector response and the ($^{235}U/^{239}Pu$) ratio response. It can be seen that the ordinary transmission measurement is quite sensitive to the added material (factor-of-2 changes are typical), whereas the changes in the detector ratios are typically only a few percent. Polyethylene has a large scattering cross section for low energy neutrons, and hence tends to mask the self-indication effect.

TABLE VI

EFFECTS OF ADDING EXTRANEOUS MATERIALS TO FISSILE SAMPLE

Fissile Sample and Absorber[a]	Change in ^{239}Pu Detector Response[b]	Change in Fission Detector Ratio ($^{235}U/^{239}Pu$)[b]
^{239}Pu in Al (1.0")	1.41	- 0.3%
^{239}Pu in Pb (0.50")	1.73	- 2.4%
^{239}Pu in ^{238}U (0.50")	2.28	- 2.6%
^{239}Pu in ^{235}U (0.060")	3.39	+ 3.2%
^{239}Pu in CH_2 (1.0")	10.02	- 36%

[a]The 0.010"-thick ^{239}Pu sample was sandwiched in the center of the absorbers. The neutron beam was filtered by 4-mil Gd.

[b]Represents the change caused by the absorber compared with the pure Pu measurement.

To determine the effect of neutron cutoff energy on the fission detector responses, the measurements were carried out using different neutron filters of Gd and Cd; the results are shown in Fig. 7. The differences exhibited by the four curves in Fig. 7 are due primarily to the 0.3-eV resonance in ^{239}Pu. The 30-mil Cd filter absorbs most of the neutrons in the energy region containing this resonance, whereas the 20-mil Cd filter transmits an appreciable fraction of the neutrons in the resonance wing. Thus the fission response ratio ($^{235}U/^{239}Pu$) increases, with Pu sample thickness, faster for the 20-mil filter than for the 30-mil filter. The 4.5-mil Gd filter attenuates the neutron spectrum on the low energy side of the 0.3-eV resonance, whereas the 1-mil Gd filter transmits an appreciable number of lower energy neutrons which in turn tends to decrease the ($^{235}U/^{239}Pu$) response.

In future work on the self-indication technique it is planned to include the isotope ^{233}U. A ^{233}U fission chamber was recently built for this purpose. Also, an appropriate moderating assembly to obtain an epithermal 1/E spectrum of neutrons

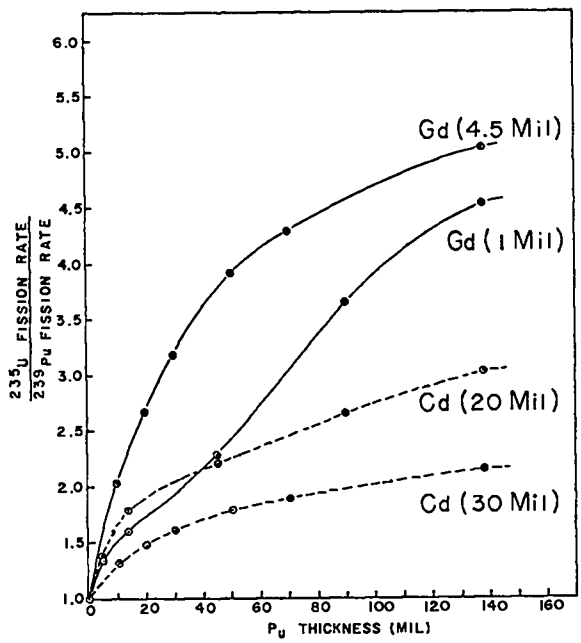


Fig. 7. Fission detector ratio ($^{235}U/^{239}Pu$) versus Pu sample thickness for different neutron filters (Gd or Cd).

from a 14 MeV (D, T) source is being developed. The potential use of a radioactive ^{252}Cf source of neutrons for this assay technique is also under investigation. Preliminary calculations have indicated that a ^{252}Cf source strength in the range 10^9 - 10^{10} neutrons/sec would be sufficient for most self-indication applications.

PASSIVE NEUTRON COUNTING TECHNIQUES

Passive Neutron Counting of a Cold MTR Fuel Element

For reactor elements containing alloys, mixtures, or chemical compounds of fissionable materials and low Z elements, counting of neutrons produced by (α, n) reactions initiated by α-particles from the fissionable materials, as well as neutrons from spontaneous fission, may provide a useful means for monitoring fuel content and material uniformity of cold fuel elements. An experiment was performed to determine if a detectable number of neutrons are emitted from a cold MTR fuel element which contains approximately 340 grams of uranium ($\sim$93% ^{235}U, $\sim$5-1/2% ^{238}U, $\sim$1% ^{234}U, $\sim$1/4% ^{236}U) alloyed with aluminum. For the neutron count the element was placed in close geometry between two N-6 slab detectors which in this configuration had a combined absolute neutron detection efficiency of $\sim$5% as determined by neutron source calibration. The counting rate with the fuel element in place was a factor of two greater than background, yielding a net counting rate for the fuel element of 132/min. This rate corresponds to an absolute neutron emission rate of $\sim$45 neutrons/sec. These neutrons are believed to arise mainly from $Al(\alpha, n)$ reactions initiated by the α decay of ^{234}U and ^{235}U. These results show that ample neutron counts could be obtained within a few minutes for MTR elements, if a high efficiency ($\sim$70%) 4π detector is used.

Coincidence Counting of Neutrons from the Spontaneous Fission of ^{240}Pu

To measure the sensitivity of the N-6 slab detectors for coincidence counting neutrons from the spontaneous fission of ^{240}Pu, a 2.356-gram Pu sample (94.15% ^{239}Pu, $\sim$5.46% ^{240}Pu, .36% ^{241}Pu) was centered between the slab detectors which were positioned face-to-face in close geometry. The coincidence rate was measured using an 80 μsec coincidence gate width and the background (random coincidence rate) was measured by inserting a delay of 500 μsec in the output circuit of one of the detectors. The neutron coincidence counting rate for this sample (attributed to the 0.13 gm ^{240}Pu content) was observed to be 14.4 counts/minute and the random coincidence background was 0.4 counts/minute. Thus the minimum amount of ^{240}Pu detectable with the slab detectors is the order of 5 milligrams. Considerably greater sensitivity for the detection of ^{240}Pu by neutron coincidence counting is expected for the N-6 4π detector (cf. LA-3921-MS), whose detection efficiency is much greater than that of the two-slab configuration. In the case of the 4π counter, spontaneous fission events are differentiated from other neutron sources by electronically requiring that two or more neutrons be detected within the characteristic neutron die-away time of the detector ($\sim$60 μsec).

To check that the above measurement of ^{240}Pu with the slab detectors is reliable in a large

background of single neutron counts,[d] a ^{238}Pu-LiF neutron source (which has a negligible spontaneous fission rate) was placed on the center line between the detectors at a distance such that the singles counting rate in each slab detector was ~13,000 counts/min. This singles background gave a random coincidence rate almost equal to the true coincidence rate from spontaneous fission of ^{240}Pu in the sample, in agreement with the rate calculated from the singles rate and the coincidence gate width. This result indicates that the present system should be adequate for the quantitative measurement of ^{240}Pu concentrations in typical oxide fuels containing ^{235}U, ^{238}U, ^{239}Pu and normal concentrations of the associated minor isotopes.

Neutrons from Spent Fuel Elements

The possible use of neutron counting techniques to determine the Pu buildup in spent reactor fuel elements has been investigated in some detail. In order to estimate the neutron production from spontaneous fission and (α, n) reactions due to elements other than ^{239}Pu and ^{240}Pu present in an irradiated fuel element, a computer code has been written which calculates the concentration of any desired element from multiple neutron capture and subsequent decay chains as a function of time during and after neutron irradiation. Calculations of transuranic nuclide concentrations were performed for Yankee Core I because this core is reasonably typical of present-day thermal power reactors and the plutonium production in this core has been measured. Using a self-consistent set of cross sections and resonance integrals, it is possible to closely match the U-burnup and Pu production that had been experimentally determined for Yankee Core I[6] without having to take into account the detailed reactor operating history. The calculated Pu isotopic abundances are compared to the measured values in Table VII.

TABLE VII

ATOMIC FRACTIONS OF Pu ISOTOPES RELATIVE TO INITIAL ^{238}U LOADING

Pu Isotope	Calculated	Yankee Core I
239	.00533	.00401
240	.00046	.00051
241	.00021	.00024
242	.00001	.00002
Total	.00601	.00478

Calculations have shown that neutron production from spontaneous fission of ^{242}Cm would be about an order of magnitude greater than from ^{240}Pu spontaneous fission for freshly irradiated fuel elements. In addition, the neutron production from the spontaneous fission of ^{244}Cm would be of the same order of magnitude as from ^{240}Pu. The principal source of production of these isotopes is through the following neutron capture and radioactive decay chains:

$$^{238}\text{U} + n \longrightarrow {}^{239}\text{U} \xrightarrow{\beta^-} {}^{239}\text{Np} \xrightarrow{\beta^-} {}^{239}\text{Pu}$$

$$+ n \longrightarrow {}^{240}\text{Pu} + n \longrightarrow {}^{241}\text{Pu} \xrightarrow{\beta^-} {}^{241}\text{Am}$$

$$+ n \longrightarrow {}^{242}\text{Am} \xrightarrow{\beta^-} {}^{242}\text{Cm}$$

for ^{242}Cm, and

$$^{241}\text{Pu} + n \longrightarrow {}^{242}\text{Pu} + n \longrightarrow {}^{243}\text{Pu} \xrightarrow{\beta^-} {}^{243}\text{Am}$$

$$+ n \longrightarrow {}^{244}\text{Am} \xrightarrow{\beta^-} {}^{244}\text{Cm}$$

for ^{244}Cm.

The computed buildup of ^{240}Pu, ^{242}Cm, and ^{244}Cm during a typical fuel element irradiation,

[d] As might arise, in practice, from (α, n) reactions in the sample.

[6] J. JEDRUCH and R. J. NODVIK, "Experimentally Determined Burnup and Spent Fuel Composition of Yankee Core I," Westinghouse Atomic Power Division Report, WCAP-6071 (1965).

and their decay after irradiation is shown in Figs. 8, 9, and 10. From the relative spontaneous fission activities vs. time after irradiation shown in Fig. 11, it is apparent that ^{242}Cm, which decays with a half-life of 163 days, is the dominant spontaneous fission source for the period up to ~1.5 years after termination of the irradiation. In oxide fuel elements, additional neutrons can result from $^{18}O(\alpha, n)$ reactions initiated by alpha decay of the transuranic elements. As in the case of spontaneous fission activities, the dominant source of (α, n) neutrons arises from ^{242}Cm. After ^{242}Cm has decayed for ~1.5 years, (α, n) neutrons arise primarily from ^{239}Pu, ^{240}Pu, and ^{241}Am alpha activity (in order of decreasing source strength). At the time of removal of a Yankee Core I fuel element from the reactor, the total neutron intensity would be ~4×10^6 neutrons/sec-element, of which $\gtrsim 85\%$ are spontaneous fission neutrons with the remainder being (α, n) neutrons.

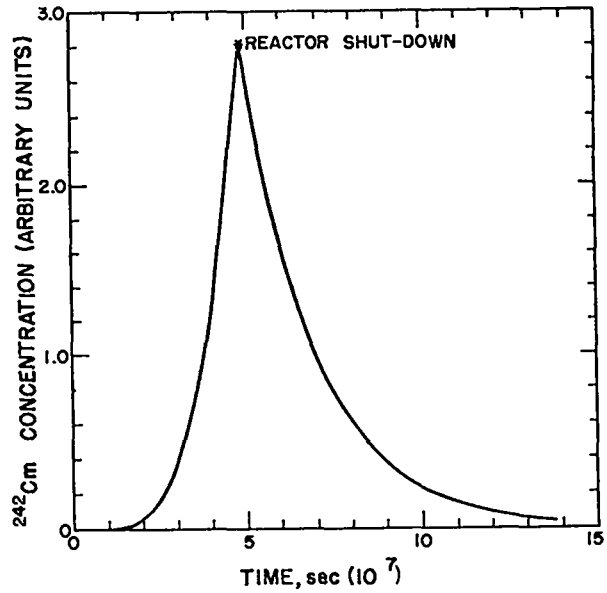


Fig. 9. Calculated ^{242}Cm concentration vs. time in a Yankee Core I fuel element.

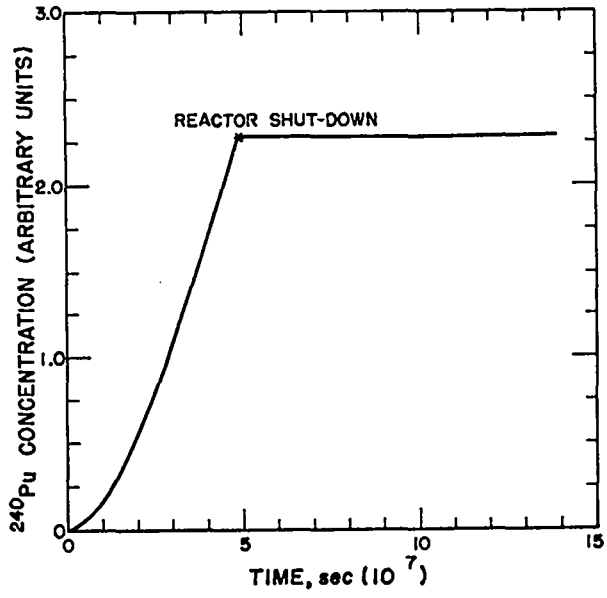


Fig. 8. Calculated ^{240}Pu concentration vs. time in a Yankee Core I fuel element.

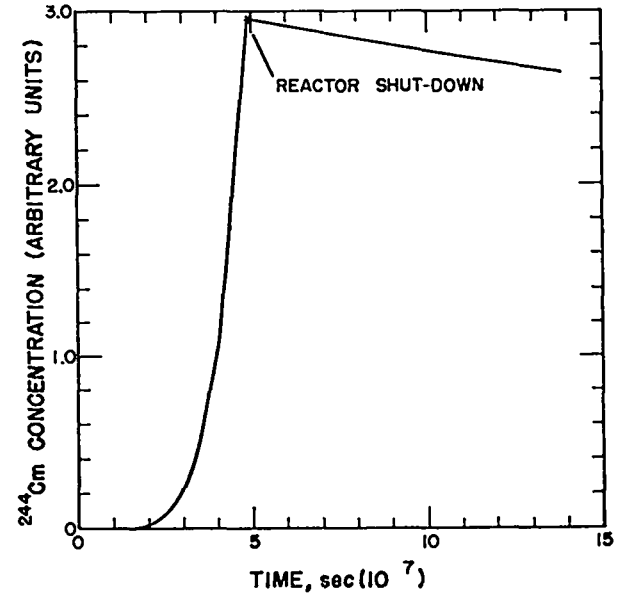


Fig. 10. Calculated ^{244}Cm concentration vs. time in a Yankee Core I fuel element.

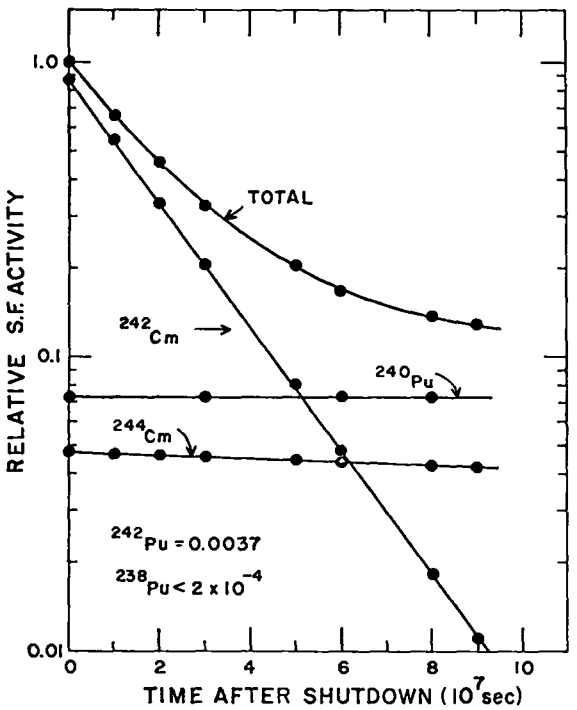


Fig. 11. Calculated spontaneous fission activities in a Yankee Core I fuel element following reactor shutdown.

It is concluded from these calculations that the expected neutron yield from a spent fuel element is sufficiently high to obtain an accurate count with existing neutron detectors. However, since ^{242}Cm, the primary source of the neutrons, has a half-life which is short compared with typical in-pile times, quantitative information on the ^{239}Pu could be inferred from neutron measurements only if detailed fuel irradiation history is known. Conversely, passive neutron counting of fuel elements which have been irradiated under controlled conditions could be used to verify, nondestructively, the predictions of burnup and transuranic production codes (such as ISOCHECK).

DELAYED GAMMA RAYS FROM FISSION

The potential of early (~ 1 sec to several minutes) delayed gamma rays for detection, identification, and assay of fissionable materials was discussed briefly in the previous progress report (LA-3921-MS). For active interrogation assay methods utilizing fission delayed gamma rays, the early time domain is expected to be most important because: (a) the largest isotopic differences in delayed gamma-ray characteristics should be found at early times; (b) the gamma-ray energy spectra should be richer in distinct high-energy (penetrating) gamma rays which can be resolved with Ge(Li) detectors. As discussed previously, analysis of gross delayed gamma-ray activity (the relevant fundamental data for which are known) may complement delayed neutron assay techniques, particularly for applications in which the environment of the nuclear material is unknown, e. g., high density scrap.

An active neutron interrogation system based on individual early delayed gamma-ray (line) yields may prove useful for safeguards if the fundamental high-resolution gamma-ray spectra (which are not yet available) show large differences among the isotopes. Of particular interest for neutron interrogation would be expected large differences in the yields of high energy gamma rays from $^{235}U(n, F)$ and $^{239}Pu(n, F)$. In this case interrogation with subthreshold neutrons could be used to obtain concentrations of ^{239}Pu and ^{235}U, separately, in the presence of a large quantity of ^{238}U, as occurs, for example, in power reactor fuels. The merits

of gamma-line signatures for fissile-material assay, in contrast to the delayed neutron yield method already under development at LASL, will be evaluated as fundamental high resolution gamma-ray data become available.

DETECTOR AND INSTRUMENTATION DEVELOPMENT

Slab Detectors

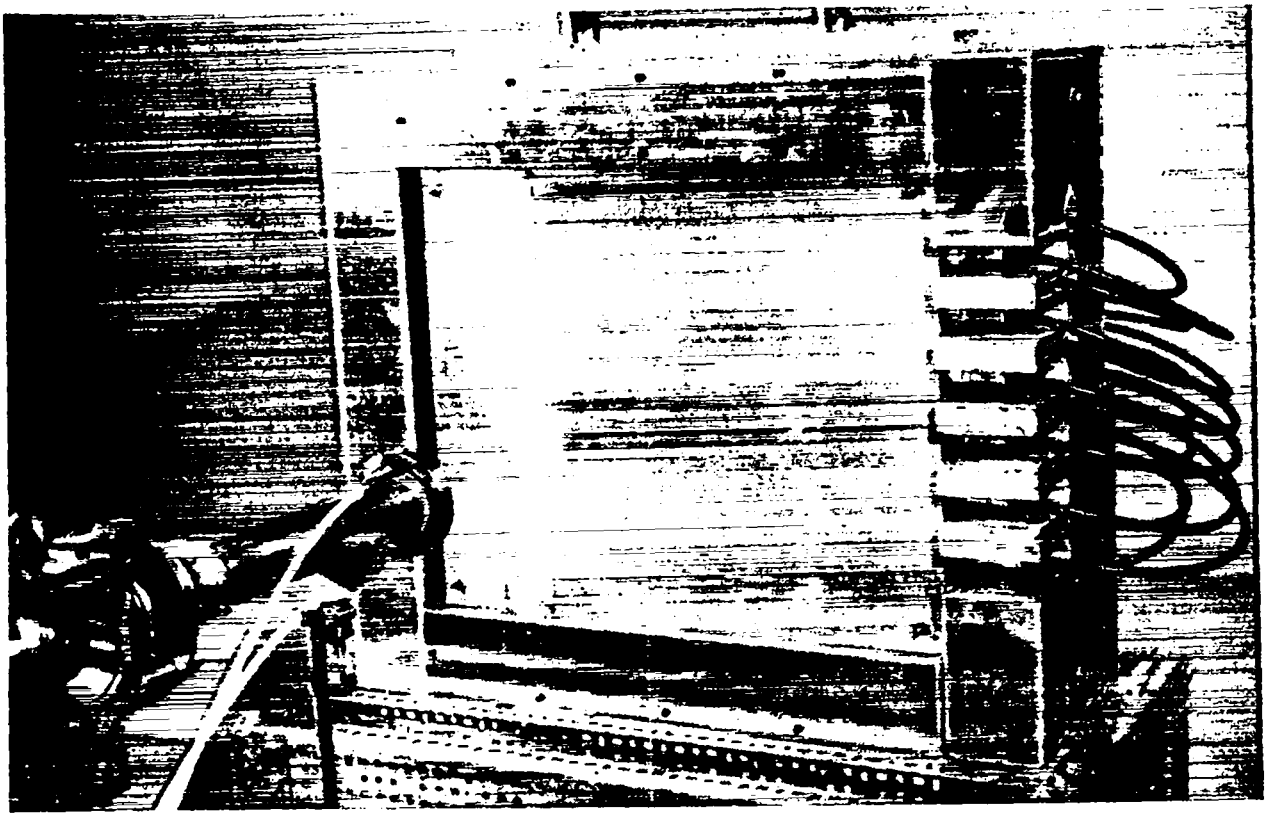
The N-6 slab detectors described previously (LA-3921-MS, LA-3859-MS) have proven to be very useful in various aspects of nuclear safeguards research. A photograph of one of these detectors located near the beam target of Accelerator I is shown in the upper part of Fig. 12. Each slab detector consists of thirteen 1-in. diam., 20-in. long, 6 atmos. ^{3}He proportional counters imbedded in two 24" x 20" x 2" slabs of polyethylene. These slabs are positioned in tandem inside a B_4C + Paraffin shield with an open face for neutron entry. Additional neutron moderating and absorbing materials may be inserted in front of and between the slabs in order to vary the energy dependence of the detector. Since neutron attenuation in CH_2 is a strong function of neutron energy, the ratio of the response of the front counter bank to the rear counter bank with CH_2 and Cd absorbers between the banks can be used to determine the "effective" average energy of incident neutrons. Optimization of the energy discrimination mode and its applications have been described previously (LA-3921-MS).

For most neutron counting applications it is desirable to use a detector which has a response independent of neutron energy. This "flat response" characteristic is particularly important for delayed neutron nondestructive-assay applications, since fission delayed neutrons have a continuous spectrum that is time dependent and is subject to energy degradation by surrounding moderator materials, particularly those containing hydrogen. Therefore, a detailed investigation of the energy response of the N-6 slab detectors has been carried out as a function of thickness and position of CH_2 moderating slabs. The "flattest" energy response was obtained using one-inch slabs of polyethylene in front, between, and behind the ^{3}He counter banks, and by removing the inner layer of Cd and the 1"-diameter polyethylene shim rods in the front counter bank. The resulting energy response of this configuration is shown in the lower portion of Fig. 12. The total energy response of the detector is essentially independent of neutron energy from a few keV to greater than 2 MeV. (Typical delayed neutron spectra lie well within this range.) Note in Fig. 12 that some information on the average energy of the incident neutrons is also obtainable from the ratio of responses of the front and rear counter banks when the detector is used in the "flat" configuration. Assuming that the active area of the slab detector is 20" x 24", the intrinsic efficiency for detecting a neutron entering this area is 13%.

Total Absorption Ge(Li) Gamma Ray Spectrometer

A coaxial Ge(Li) "duode" detector of the type developed by Kraner and Chase at Brookhaven[7] is presently being fabricated. One crystal with an active volume of ~ 16 cm^3 has been lithium drifted and is now ready to undergo preliminary testing. Special preamplifiers for use with this detector have also been designed and built. A detector of

[7] H. W. KRANER and R. L. CHASE, "A Total Absorption Ge(Li) Gamma Ray Spectrometer," Brookhaven National Laboratory Report, BNL-12332.



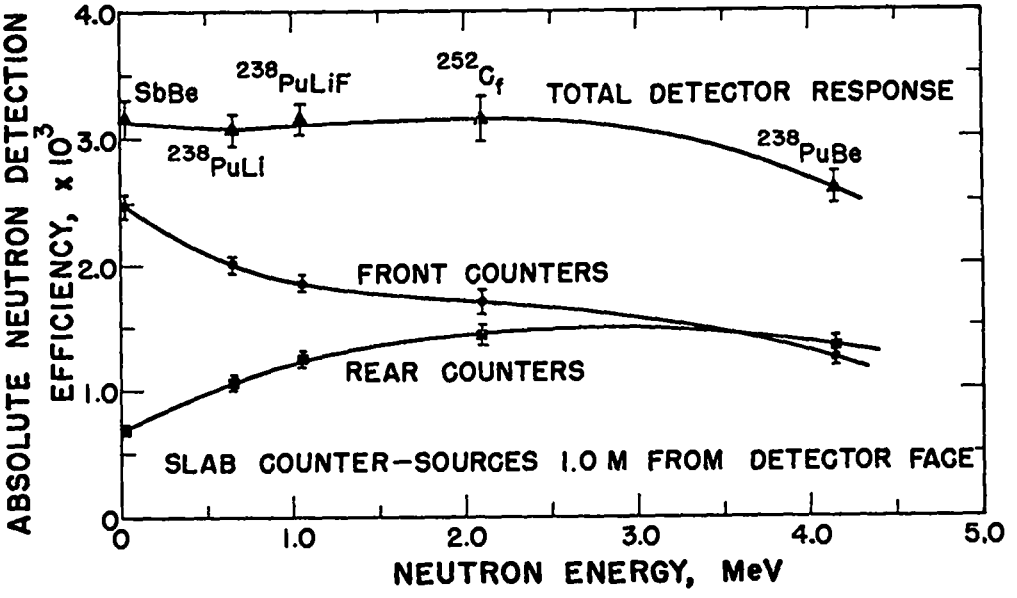


Fig. 12. The photograph shows an N-6 high efficiency (~13%) slab detector positioned near the Accelerator I beam target. This neutron source and detector combination is used extensively for delayed neutron assay techniques being developed at LASL. The lower portion of the figure shows the efficiency of the detector versus neutron energy as measured with absolute neutron sources located 1 meter from the detector. The flat response as a function of energy for the sum of the front and back counter banks (i. e., the total detector response) is necessary for most applications involving assay of large complex arrays of nuclear material such as fuel elements, nuclear devices, scrap, etc.

this type should provide a valuable laboratory tool for general γ-ray counting, identification of γ-rays from (α, n) reactions (notably from U and Pu α's), and on-line γ-ray counting of fissile materials for isotope identification and practical assay applications.

DENSE PLASMA FOCUS SOURCE

The intensive program of testing, debugging, and modification of the N-6 Dense Plasma Focus (DPF) pulsed neutron source has now achieved the desired neutron yield range ($\sim 10^{10}$ D, D neutrons/pulse) with adequate reproducibility and reliability.

The previously reported problems with crack-

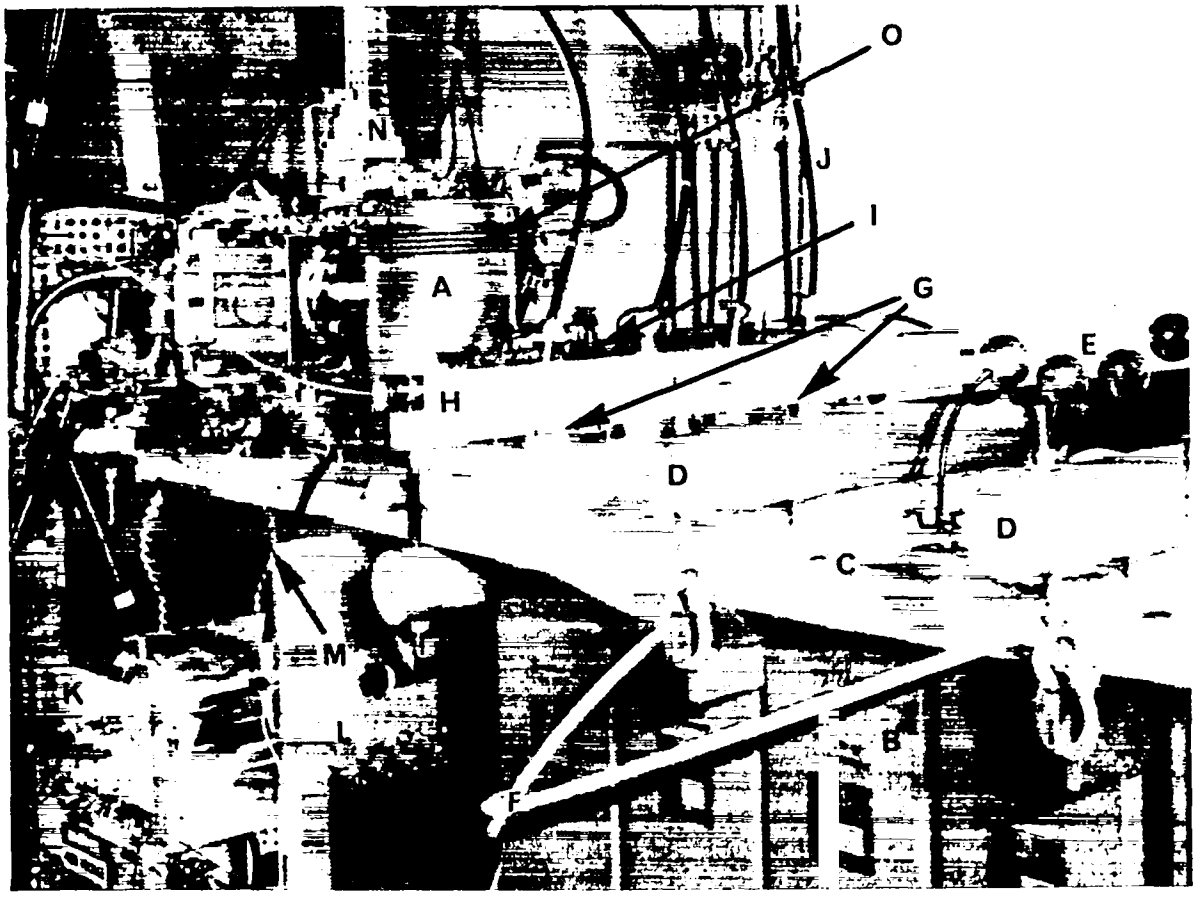


Fig. 13. Dense Plasma Focus Neutron Source.

A - Plasma discharge chamber
B - Capacitor bank
C - Strip lines
D - Strip-line clamping yokes
E - Strip-line protective spark gaps
F - Capacitor charging lines
G - Vacuum spark gaps
H - Spark-gap clamping yoke
I - Spark-gap vacuum and SF_6 manifolds
J - Spark-gap initiator cables
K - Spark-gap initiator
L - Energy dump resistors
M - High vacuum pump
N - Chamber safety pressure interlock
O - Chamber cooling coils

ing of the glass insulators in the vacuum switches have been shown to be due to nonuniformity of current load among the switches. Minute differences in firing time caused momentary massive overload of one switch, with consequent explosive destruction of the switch insulators. In order to correct this, the total storage bank of twelve capacitors has been divided into four sub-banks of three capacitors each, with an individual vacuum switch on each sub-bank. At the same time it was found necessary to isolate the spark plugs in each of the switches from one another by using separate cables from the firing pulses to each of the spark plugs. This change considerably improved the simultaneity of switching. Recent measurements indicate that all four vacuum switches now fire within less than 20 nanoseconds of each other. Also, to prevent the extensive damage caused by inadvertently firing with a broken glass insulator in the plasma gun, a diaphragm-operated vacuum-interlock switch has been installed directly on the DPF discharge chamber.

The above-mentioned improvements plus several minor alterations in insulation and vacuum protective circuitry have virtually eliminated shutdown of the DPF due to massive failures of the type experienced in the past. The DPF source, with the new vacuum switching system, is shown in Fig. 13.

Intensive testing of the improved DPF system has demonstrated that the Dense Plasma Focus is a practical, reliable neutron source, capable of producing neutron pulses of width 50 to 100 nanoseconds, with (D, D) neutron yields the order of 10^{10}--the original design objective. It will be recalled that this corresponds to a yield of 10^{12} (D, T) neutrons per pulse for a deuterium-tritium gas mixture replacing the present deuterium charge. Thus far maximum D, D neutron yield observed from a single DPF burst is $\sim 2 \times 10^{10}$. A repetition rate of one burst per minute is readily maintained, and shot-to-shot yield variations are, in general, quite nominal and acceptable. Thus the DPF pulsed neutron source is now ready for use in appropriate safeguards research problems; the first problem will be a measurement of delayed gamma rays from fission in the interval from 0.10 to a few seconds.

N-6 ACCELERATORS AND NEUTRON SOURCES

The N-6 mobile neutron source, "Accelerator I," (see Fig. 14) continues to perform with full reliability. Two new quick-change target-holders have been designed and fabricated. The alternate use of two target-holders allows a target "cooling time" for the induced radioactivity in the Cu-backed tritium target to decay to a reasonable level before reuse.

Two new "long targets" (cf. Fig. 6, LA-3921-MS) were assembled and used on the 300 keV Cockcroft-Walton for the recently completed measurements on absolute delayed neutron yields. This improvement reduced the amount of scattering material in the immediate vicinity of the sample and fission monitors. One target was loaded with tritium and one with deuterium; with this arrangement the operator could change readily from (D, T) neutrons to (D, D) neutrons in approximately 20 minutes without disturbing the experimental setup (i. e., samples, detectors, etc.).

The 300 keV Cockcroft-Walton was down three days during the quarter for routine checking of the rectifiers, for diffusion pump maintenance, and for general cleanup.

Another new modification on the Cockcroft-Walton accelerator now permits remote switching,

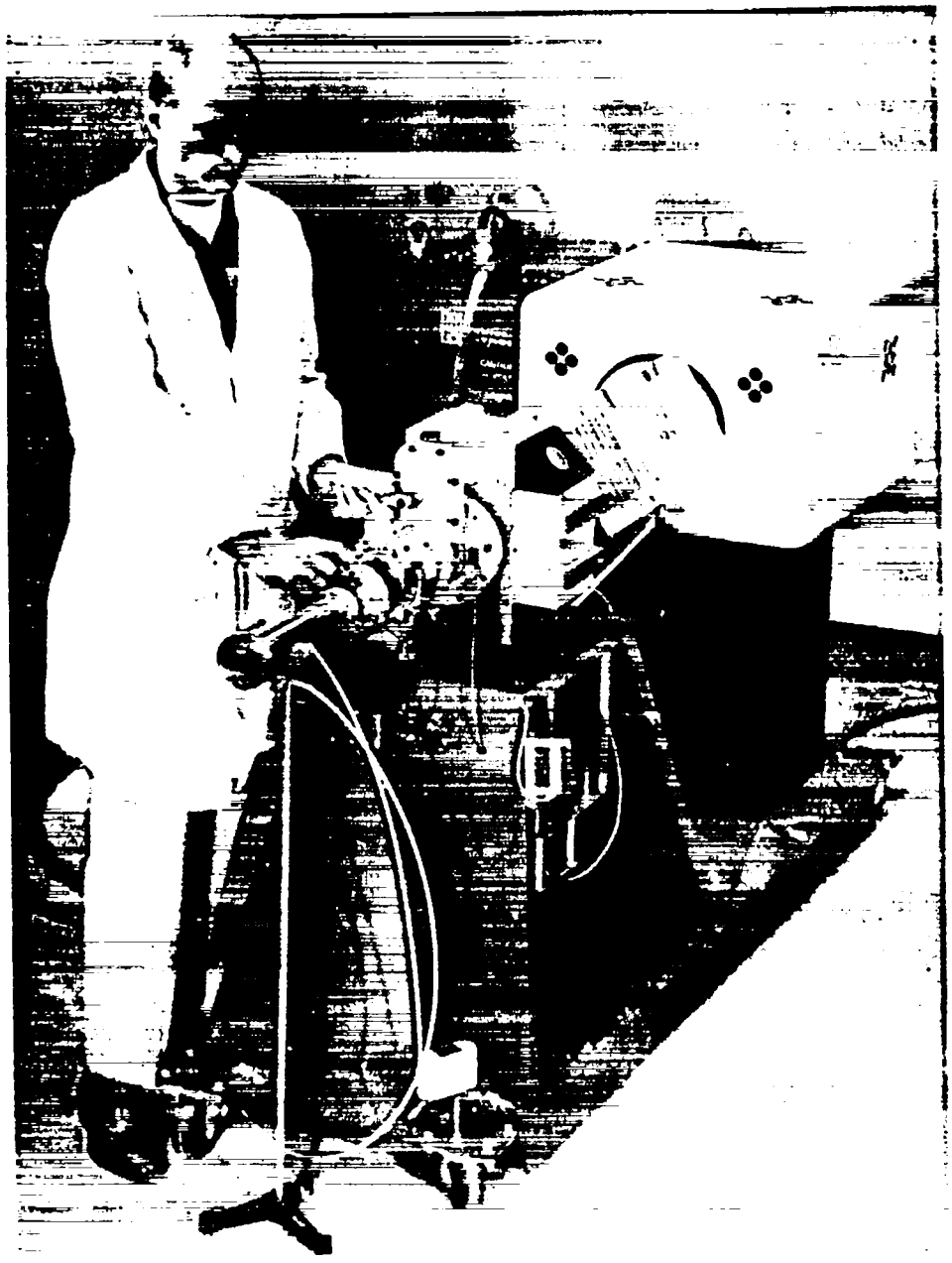


Fig. 14. Accelerator I - A Compact, High Intensity Source for Neutron Interrogation and Nondestructive Assay. (Accelerator potential: 150 kV; maximum beam current: 3.5 ma; beam pulsing by pre- and post-deflection; beam modulation by gating the RF ion source.) Maximum source intensity is $\sim 4 \times 10^{11}$ D, T neutrons/sec; the source can be gated on and off as required for optimum neutron interrogation applications. Two new quick-change target holders have been adapted to Accelerator I for fast, convenient target replacement in laboratory and field installations.

from the control console, of the electrostatic strong focusing from the left port to the right port of the accelerator. A versatile timing and light-link system for the C-W accelerator has been designed and is now under construction.

Both Accelerator I and the Cockcroft-Walton were in almost continuous use during the second quarter of 1968, and both continue to perform very satisfactorily in either the DC or pulsed mode of operation.

OTHER CONTRIBUTIONS TO NUCLEAR SAFEGUARDS RESEARCH AT LASL

Foil Preparation (CMF-4)

Several deposits of ^{233}U were evaporated on stainless steel and platinum backing discs for use in fission chamber detectors.

Californium Source Calibration (N-2)

The spontaneous fission rate of a thin deposit of ^{252}Cf was determined by absolute counting of the fission fragments in Group N-2's low-geometry alpha chamber.

^{238}Pu-Li Source Fabrication and Calibration (GMX-1, P-6, CMB-11)

Fabrication of Group N-6's new fluorine-free ^{238}Pu-Li source has now been completed. Contributing groups (during the second quarter of 1968) were:

- P-6 : Absolute calibration of the source.
- CMB-11: Major responsibility for preparation and containment of the source.
- GMX-1 : Radiographic inspections of the source containers.

Uranium Sample Preparation (CMB-11)

Several disc-shaped samples of ^{233}U were prepared and sealed in 0.07"-wall copper cans.

Enriched ^{17}O Sample (CMF-2)

Two samples of H_2O enriched to 3.8% (7 grams) and 34% (2.23 grams) in ^{17}O were supplied for $^{17}O(n, p)^{17}N$ cross section measurements.

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

Development of Group-Averaged Cross Sections (C-DO)

Several group-averaged cross sections have been calculated from the data in the LASL Evaluated Nuclear Data Library for use in DTF-IV transport calculations.

Ge(Li) Detector Fabrication (P-1)

The lithium drifting of a large (16 cm^3) germanium crystal has been completed.

Self-Indication Measurements (P-2)

The Water Boiler Reactor facilities at Group P-2 were used to perform the resonance self-indication experiments.

PUBLICATIONS

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2. G. R. KEEPIN, "Nuclear Safeguards Applications of ^{252}Cf Spontaneous Fission Neutron Sources," presented at Meeting of USAEC Transplutonium Committee, USAEC Headquarters, Washington, D.C. (April, 1968).

3. L. V. EAST and H. M. MURPHY, JR., "Decay of ^{109}Cd," Nuclear Physics A107, 382-384 (1968).

4. H. O. MENLOVE and W. P. POENITZ, "Absolute Radiative Capture Cross Section for Fast Neutrons in ^{238}U," Nucl. Sci. Eng. 33, 24 (1968).

5. H. A. GRENCH and H. O. MENLOVE, "$In^{113}(n,\gamma)In^{114m,114g}$, $In^{113}(n,n')In^{113m}$, $In^{115}(n,\gamma)In^{116m,116g}$, and $In^{115}(n,n')In^{115m}$ Activation Cross Sections Between 0.36 and 1.02 MeV," Phys. Rev. 165, 1298 (1968).