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Pulsed Neutron Research for Nuclear Safeguards

Program Status Report

July-September 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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PULSED NEUTRON RESEARCH FOR NUCLEAR SAFEGUARDS

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DELAYED NEUTRON KINETIC RESPONSE TECHNIQUES FOR NONDESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS

A general experimental program is now underway on testing of delayed neutron kinetic response techniques for nuclear safeguards applications to fissionable material assay. Preliminary measurements have been carried out on U^{235}, U^{238} and Pu^{239} using D, T neutrons from the N-6 Cockcroft-Walton accelerator. Neutron production is monitored by an associated particle detector which counts alpha particles from the T(D, n)He^4 reaction. The samples of U^{235}, U^{238} and Pu^{239} were in the form of metal discs weighing approximately 100 grams each. Samples were irradiated for 1 sec and delayed neutron time-decay spectra were observed for 4 sec following irradiation. The time spectra were measured using a multichannel pulse height analyzer in conjunction with a time mark generator adjusted to provide 50 millisec channel widths.

Figure 1 shows the delayed neutron time response from the U^{238} and the U^{235} samples, and Figure 2 presents similar data for U^{238} and Pu^{239} as well as for a composite sample of 54% U^{238} and 46% Pu^{239}. These data are preliminary; approximately eight minutes total running time was required to obtain each decay curve in Figures 1 and 2. The delayed neutron detector used thus far has been a flat-energy-response "long counter" with an intrinsic efficiency the order of 1%. A high-intrinsic-efficiency (~16%) flat-response detector has now been constructed and tested for use in future kinetic response studies (see Detector Development).

Although it will be possible in some instances to use the spontaneous fission neutrons from Pu^{240} as a direct indicator of the total amount of plutonium in an unknown system, in the present kinetic measurements Pu^{240} contributed a constant background of less than 5% of the total delayed neutron count. Clearly the use of repetitive pulsing enhances the delayed neutron response relative to background, thus providing a considerable advantage in the assay of high neutron background systems (e.g. those containing Pu). The data in Figures 1 and 2 have been normalized to the same total number of counts during the last second of observation time to emphasize decay slope differences at earlier times.

As suggested by the minimum-running-time optimization studies described below, ratios of counts in the first second to those in the remaining observation time were calculated and are shown in Table I. The counting statistics in these ratios are about 1%. The resulting "isotope discrimination ratio" between U^{238} and U^{235} is 1.25, and the corresponding ratio for U^{238} and Pu^{239} is 1.26. It should be noted that these ratios apply to experimental parameters (irradiation, observation times, etc.) corresponding to overall minimum running time. With some increase in running time, considerably greater isotope discrimination factors can be expected under other experimental conditions. (Cf. N-6 Program Status Report, January-March 1967, LA-3686-MS, Table I.)

For the more difficult problem of discrimination between U^{235} and Pu^{239}, auxiliary methods based on the relatively large (factor of 3) difference in delayed neutron fractions between

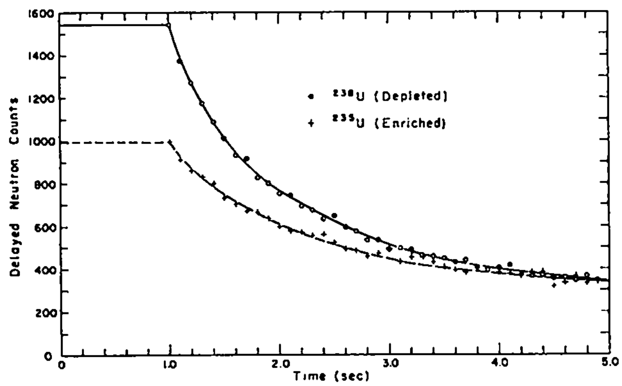

Fig. 1. Delayed neutron time response spectra for U^{238} and U^{235}.

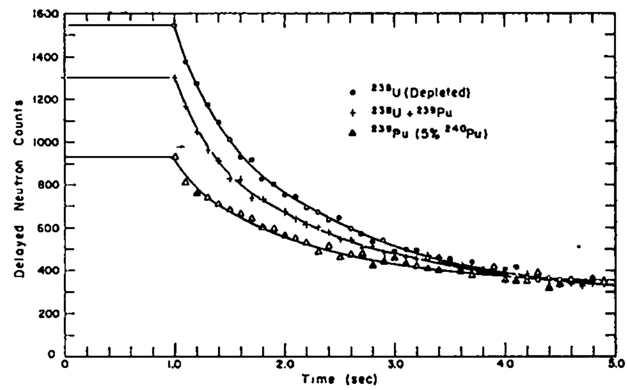

Fig. 2. Delayed neutron time response spectra for U^{238}, Pu^{239} and composite sample (54% U^{238} and 46% Pu).

these fissile species are being considered.

TABLE I

ISOTOPE DISCRIMINATION RATIOS FOR REPETITIVE PULSING UNDER MINIMUM RUNNING-TIME CONDITIONS[a]

Fission Isotopes	Measured Discrimination Ratio	Calculated Discrimination Ratio
U^{238}, U^{235}	1.25	1.31
U^{238}, Pu^{239}	1.26	1.38

[a] sample irradiation time 1 sec, observation time 4 sec, counting time fiducial 1 sec, total cycle time 5 sec.

The discrepancy between measured and calculated discrimination ratios in Table I suggests that delayed neutron periods and/or abundances are different for 14 MeV neutron-induced fission than for fission-spectrum-induced (~ 3 MeV) fission. An experiment to measure delayed neutron periods and abundances from 14 MeV fission is planned and is now being implemented.

In addition to experimental measurements, parallel analytical and computational studies are underway to investigate optimum conditions for nondestructive assay of fissionable materials by delayed neutron kinetic response techniques. The sensitivity of relative isotopic abundance determination to the various input parameters in kinetic response experiments has been calculated for different combinations of materials, assuming the previously measured LASL delayed neutron data for fast (fission-spectrum induced) fission. By proper choice of irradiation pulse width, observation time and pulse repetition frequency, it has been shown that delayed neutron response can be adjusted so as to obtain a given assay accuracy in minimum overall measurement time. Such calculations provide very useful guidance for kinetic response measurements such as those described above.

To provide an alternative method of sample irradiation for DIA (detection, identification and analysis) applications to safeguards, arrangements are being made, thru the AEC Transplutonium Committee, to obtain a 1 mg Cf^{252} spontaneous fission neutron source. A transfer system and shielding for this intense neutron source are being designed. Present calculations indicate a shield of laminated lead and borated polyethylene 50-100 cm thick will reduce the neutron intensity to acceptable levels (both biologically and experimentally, the latter being a considerably more stringent requirement).

Calculations on moderated Cf^{252} and other "tailored spectrum" radioactive (α, n) neutron sources are being pursued in connection with DIA applications. Cf^{252} in particular appears extremely promising for future nuclear safeguards applications.

ABSOLUTE DELAYED NEUTRON YIELD MEASUREMENTS

One of the major parameters required for the measurement of absolute delayed neutron yield per fission is the number of fissions in the sample. Two methods have been used in the N-6 absolute yield experiments:

a) Radiochemical analysis whereby the sample is dissolved and the fission product Mo^{99} is extracted and counted. The number of fissions is obtained from an independently-evaluated "K" factor used to convert Mo^{99} β-counts to fissions. The "K" factors are well established for thermal and fission-spectrum fission, but are somewhat less certain for 14-MeV fission.

b) A small (few hundred microgram) known-weight foil of the same material as the sample is counted in a fission chamber. The sample is placed as close as possible to the foil. Since the efficiency of the fission counter is nearly unity, the number of fissions in the sample is then, to first approximation, the observed fission count multiplied by the ratio of the weights of the foil and sample. However, because of differences in position between sample and foil and neutron attenuation effects, corrections must be applied. An alternative is to "sandwich" the sample between two foils. This arrangement virtually eliminates the correction factors, but decreases considerably the neutron intensity at the sample.

In order to compare the number of sample fissions as determined by the different methods, twenty-one samples were irradiated at the Cockcroft-Walton accelerator and sent to the LASL Radiochemistry Group, J-11, for radiochemical analysis. At this time, Group J-11 re-examined the K factor used for 14-MeV fission and concluded that the factor should be increased by approximately 10%. Direct fission measurements using the sandwich technique are in good agreement (± 5%) with radiochemical analysis (using the revised K factor). However, the single foil method, used previously for 3-MeV and 14-MeV yield measurements, gave approximately 10% lower fission yields. Accordingly, the absolute delayed neutron yield values reported in the previous progress report (LA-3732-MS) should be decreased by 10%, thus providing better agreement at 3 MeV with earlier results obtained by others. The yield ratios and thermal fission yields are unaffected.

A new high-efficiency "flat" neutron detector has been constructed (see Detector Development), and the neutron intensity from the Cockcroft-Walton has again been increased. These two developments will hopefully enable general use of the more precise sandwich technique for future fission-yield evaluations.

ABUNDANCES AND PERIODS OF DELAYED NEUTRONS FROM 14-MeV NEUTRON FISSION

Preparations to measure and analyze the intensity of delayed neutrons as a function of time following 14-MeV neutron-induced fission are progressing. In conjunction with the Cockcroft-Walton accelerator as the source of pulsed, 14-MeV neutrons, a highly efficient 4π neutron detector is being developed for this experiment. Because of the relatively high intensity of delayed gamma-rays which are emitted in the time region of interest for delayed neutron studies, a hydrogen-moderated plus BF_3 and He^3 counter system was selected as the detector for this experiment in preference to other high-efficiency neutron detectors, e. g. conventional large liquid scintillators.

Design and procurement is underway on a pneumatic transfer system which will transport each fission sample from the irradiation position (at the CW target) to the center of the 4π detector, and then, after a preselected counting time, on to a feeder hopper where the samples will "cool" before being recycled. A sufficient number of samples (~ 20) must be employed so that the data collection cycle will not be limited by "cooling" time of the irradiated samples. It is anticipated that times as short as 50 ms for transfer of the sample from point of irradiation to point of detection can be achieved with the planned 100 psi pneumatic transfer system.

CW ACCELERATOR; EXPERIMENTAL USE AND FACILITY DEVELOPMENT

During the third quarter of 1967, operating time on the N-6 Cockcroft-Walton accelerator was devoted to N-6 nuclear safeguards research, including neutron spectrometer and detector development, and to the work of other LASL technical groups, mainly in the Weapons and Weapons Test Divisions. Several needed maintenance functions and improvements to the accelerator were carried out during this reporting period. These included upgrading of the vacuum system, target changing facilities, beam focusing, and alignment. For DIA kinetic response measurements the maximum beam pulse width was extended to one second while maintaining the capability of a 50% maximum duty cycle.

Outfitting of the N-6 electronics and instrumentation work trailer located near the accelerator control room is essentially complete; this trailer provides urgently needed provisional work space for support of accelerator operations.

DENSE PLASMA FOCUS SOURCE

Design of the N-6 Dense Plasma Focus (DPF) source has been completed, and fabrication and procurement of components is largely complete as of the close of this reporting period. This device incorporates the DPF technology developed at Los Alamos and the stripline plus dielectric-switch method of power transmission recently reported by Beckner

(Rev. Sci. Inst. 38, 507 (1967)). Assembly of the 20-kilovolt charging circuitry and of the vacuum system for the 30-kilojoule discharge tube ("gun") is underway. It is hoped that the device will provide maximum pulse yields in excess of 10^{10} D, D neutrons or 10^{12} D, T neutrons per burst at pulse repetition rates of 2-3 per minute.

Various neutron detectors for measuring integrated total yield and yield-vs-time profile of the DPF source are being tested and calibrated. A silver activation counter (LA-3498-MS) has been constructed and calibrated for D, T neutrons on the CW accelerator; the desired linear response to total yield is indicated over a yield range of 4.5×10^8 to 2.5×10^{10} neutrons per burst. This Ag activation detector is limited to a maximum burst repetition rate of ~1 per 5 minutes by the 2.5 minute β-activity from Ag^{107}. For higher burst repetition rates, consideration is being given to the $F^{19}(n,\alpha)$ $N^{16} \xrightarrow{\beta} O^{16} + \gamma$ reaction with a 7 sec β-decay period and a 6.5 MeV γ from O^{16}. The γ sensor planned for these activation detectors is a 3" x 3" NaI crystal with matched photomultiplier tube.

The yield-vs-time profile of the DPF neutron pulse will be measured with a fast liquid scintillator-photomultiplier system which has been tested using intense neutron bursts from the CW accelerator. It has been shown that the output of this detector is proportional to the neutron yield-vs-time profile of the burst provided the average time separation of individual neutron pulses lies within the time resolution of the detector.

A laboratory has now been set up in the high-bay area of the Fast Reactor Core Test Facility where final assembly, testing and operation of the DPF source will be carried out as part of the N-6 safeguards research and development program.

DETECTOR DEVELOPMENT

High Efficiency "Long-Counter"

Development of high efficiency neutron detectors to be used in place of conventional long counters has been conducted using multiple He^3 proportional counters in polyethylene moderators. The most promising detector built to date consists of four two-inch diameter, 10 atmosphere He^3 counters placed in an eight-inch diameter polyethylene cylinder having removable sections for "tailoring" the detector's energy response. The detector can be adjusted to provide an essentially flat response for neutron energies up to ~4 MeV with an intrinsic efficiency of 14% (see Figure 3). Intrinsic efficiencies up to 16% can be obtained with a slight loss in flatness of response.

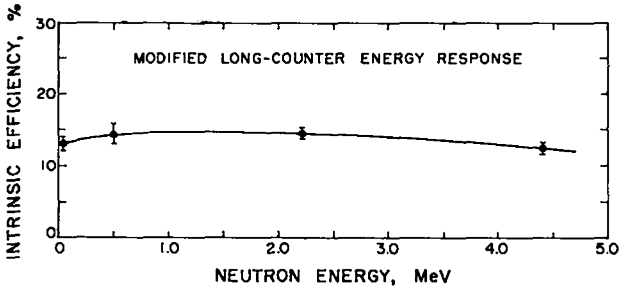

Fig. 3. Efficiency of modified long counter versus neutron energy.

4π Neutron Detector

As a result of an extensive survey of high efficiency, flat-energy-response, neutron detection schemes, N-6 will build a large 4π detector consisting of four concentric rings of high-pressure BF_3 proportional counters in a moderating material having an axial sample hole. In an earlier design of this type of detector (cf. J. T. Caldwell, UCRL-50287 (1967)), each ring contained 12 one-inch diameter, 20-inch long BF_3 counters. By increasing the number of BF_3 counters in the outer ring and/or using He^3 counters, the detection efficiency can be increased, particularly for neutrons above ~1 MeV. The total detection efficiency for delayed neutrons from fission is expected to be about 50%. This is less than could be obtained with a Cd- or Gd-loaded large liquid scintillator; however, the BF_3-plus-moderator detector has, for many experiments, the overwhelming advantage of being insensitive to gamma rays. In addition, crude neutron energy information ("spectral indices") can be obtained from the ratio of detected neutrons in the inner and outer counter rings (see Figure 4). High-pressure BF_3 and He^3 counters of various types are presently being evaluated for use in this detector.

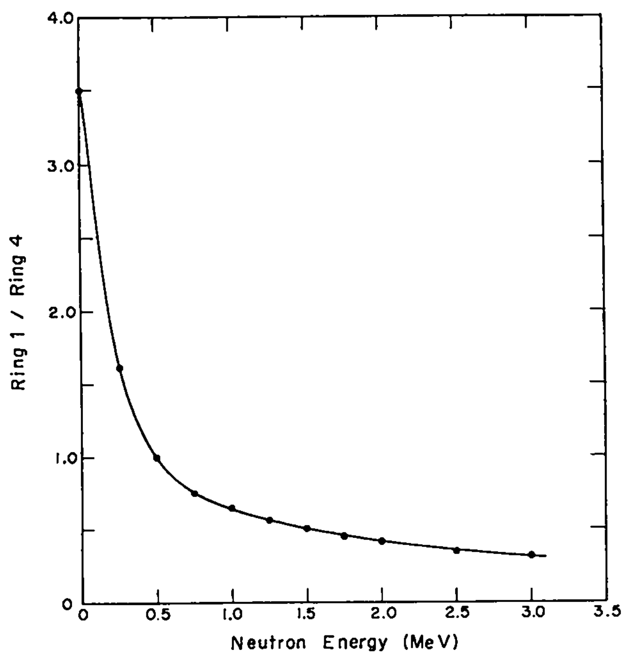

Fig. 4. The ratio of efficiency of detectors in ring 1 to that for detectors in ring 4 versus neutron energy. This ratio has been normalized to unity at E_n = 0.5 MeV, a representative average energy of the delayed neutron spectrum.

Large Liquid Scintillator

Consideration is being given to updating the large liquid scintillation detector used by Diven and Hopkins for measuring the number of prompt neutrons from fission. Such a very-high-efficiency detector could be very useful as a neutron source calibration facility, as well as in future neutron experiments.

Detector Recovery-Time from Intense Neutron Bursts

A simple preamplifier gating circuit for use with proportional counters has been developed which eliminates amplifier base-line shift following a neutron burst from the CW accelerator. Using pulse differentiation at the input of a charge-sensitive preamplifier in conjunction with gating allows complete recovery of a long counter within 200 μs of an intense neutron burst.

OTHER CONTRIBUTIONS TO NUCLEAR SAFEGUARDS RESEARCH AT LASL

Monoenergetic Neutron Source for Yield versus Energy Measurements (P-9, Van de Graaff)

Apparatus for measuring delayed neutron yields as a function of neutron energy was set up at the Los Alamos Van de Graaff accelerator (Group P-9). It was found that measurements could not be pursued because of neutron background, largely from T(p, n) neutrons originating at the post-acceleration beam-deflection slits. It is hoped that the introduction of a retractable slit, plus additional shielding, will reduce background to a tolerable level.

Radiochemical Analysis (J-11)

The total number of fissions in twelve samples of U^{235}, six samples of natural uranium, and three samples of thorium were evaluated by Mo^{99} radiochemical analysis.

Fission-Counter Foil Preparation (CMF-4)

Three U^{235} and six thorium foils were prepared in the form of 1" diameter evaporated deposits on platinum backing discs.

Sample Preparation (CMB-11)

All of the fourteen plutonium samples requested by Group N-6 have been fabricated by CMB-11 and received by N-6. In addition, fourteen samples of U^{233} have been received. The U^{233} samples are in the form of discs 1" diameter by .005" or .010" thick, hermetically sealed in copper.

PUBLICATIONS

1. A Procedure for Evaluating Modified Pulsed Neutron Source Experiments in Subcritical Nuclear Reactors, Nuclear Science and Engineering 29, 272 (1967).
2. Comparative Measurements of Uranium Atom Emission from Fissioning Surfaces, Nuclear Science and Engineering 29, 425 (1967).