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LOS ALAMOS SCIENTIFIC LABORATORY
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University of California

# Pulsed Neutron Research for Nuclear Safeguards



Program Status Report April-June 1967

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

#### NUCLEAR SAFEGUARDS RESEARCH SERIES

G. Robert Keepin, Editor

This LA... MS report presents the status of the nuclear safeguards research program at Los Alamos. The previous report in this series is LA-3682-MS.

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# LOS ALAMOS SCIENTIFIC LABORATORY of the University of California

# Pulsed Neutron Research for Nuclear Safeguards



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Distributed July 17, 1967



#### PULSED NEUTRON RESEARCH FOR NUCLEAR SAFEGUARDS

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### NUCLEAR SAFEGUARDS RESEARCH AND DEVELOPMENT PROGRAM

The major effort of the LASL Pulsed Neutron Research Group during the 2nd quarter of 1967 has been concentrated on (1) basic measurements required in the application of delayed neutron kinetic response techniques to the detection, identification and analysis of fissionable materials and (2) the development of a compact, intense pulsed source for practical implementation of the Group's safeguards research and development program (cf. LA-3682-MS, LASL Nuclear Safeguards Research Series, Program Status Report, January-March 1967).

### ABSOLUTE DELAYED NEUTRON YIELD MEASUREMENTS

The determination of absolute delayed neutron yield per fission from each of the major fissioning species is basic to the kinetic response methods for nondestructive assay of fissionable materials being developed by Group N-6. Extension of accelerator pulsing capabilities and improvements in detector instrumentation have increased effective delayed neutron source intensity by a factor of 5, and decreased detector background by an order of magnitude. Also an absolute fission counter has been constructed and installed next to the accelerator neutron source (D or T target) to provide an independent absolute determination of the number of fissions in the sample under study. The present data on absolute yields for three fission species at two incident neutron energies (3 MeV and 14 MeV, corresponding to the D,D and D,T neutron reactions respectively) are summarized in Table I.

TABLE I  $\begin{tabular}{ll} ABSOLUTE DELAYED NEUTRON YIELDS IN \\ U^{235},\ U^{238} \ AND \ Th^{232} \ FISSION \end{tabular}$ 

Fission	Energy of Neutrons Inducing Fission		Yield Ratio.
Isotope	3 MeV	14 MeV	(3 MeV/14 MeV)
u <sup>235</sup>	0.0175 ± 0.003	0.010 ± 0.002	1,75 ± 0,18
U <sup>238</sup>	0.052 ± 0.010	0.033 ± 0.007	1.57 ± 0.16
Th <sup>232</sup>	0.062 ± 0.018	0.037 ± 0.011	1.67 ± 0.17

The appreciable decrease in yield at 14 MeV as compared to 3 MeV indicated in Table I is common to all fission species studied thus far (and is, in fact, expected for all species undergoing binary fission). ratios in the last column of Table I should be essentially free of systematic errors (e.g. arising from sample size effects and detector efficiency calibration), since the comparison of relative delayed neutron yields at 14 and 3 MeV involves only a change from the tritium to the deuterium target -- the entire experimental arrangement being otherwise identical. This accurately measured decrease of delayed neutron yield at 14 MeV is in accord with theoretical expectations based on the behavior of fission mass and charge distributions, and the known decrease in fission chain lengths with increasing energy of the neutron inducing fission. The yield data reported here are particularly significant inasmuch as they stand in direct contradiction to previous data (largely from the USSR, and some from the USA) which would indicate a strong increase -- roughly a doubling -- in delayed neutron yield as one progresses from low energy fission to 14 MeV fission.

In Table I the measured absolute yield for  ${\rm U}^{235}$  at 3 MeV is in agreement (to within 5%) with corresponding yield values reported in the literature. The new yield values measured at 3 MeV for the threshold fissioning species, U<sup>238</sup> and Th, are approximately 30% higher than previously reported yields for these species. A further measurement of delayed neutron yield from thermal-neutron-induced fission of U<sup>235</sup> was made using a 9-1/2" polyethylene sphere surrounding the accelerator target, to provide the source of thermal neutrons. The yield value thus obtained was in good agreement with the published value of 0.0158 delayed neutrons per U thermal fission. It should be noted that absolute delayed neutron yield per U 235 fission is essentially a constant over the neutron energy range from thermal to 3 MeV (and probably higher) -- a physical result of considerable significance to the practical application of delayed neutron response techniques to nondestructive isotopic assay. This same energy-independence of yield is expected to hold for the other fissile species; a planned series of measurements on the other major thermal-fissioning species will provide a precise check on this important point.

COCKCROFT-WALTON ACCELERATOR; EXPERIMENTAL USE AND FACILITY DEVELOPMENT

During the second quarter of 1967, operating time on the N-6 Cockcroft-Walton accelerator was divided roughly equally between N-6 nuclear safeguards work and the

work of other LASL technical groups, mainly in the Weapons and Test Divisions. In view of the significant amount of N-6 accelerator time being devoted to nuclear weapons research, and the growing needs for accelerator use in the safeguards research program, it is now clear that N-6's new Dense Plasma Focus source (and associated safeguards technology development) cannot be located, even temporarily, in the already-fully-scheduled Cockcroft-Walton accelerator bay at TA-18. Fortunately an alternative location for the Dense Plasma Focus source has now been provided at another site in Los Alamos (see later section on Dense Plasma Focus).

Beam modulation capabilities of the Cockcroft-Walton accelerator have been increased from 5% to 60% duty factor for use in delayed neutron yield and kinetic response measurements. The new beam deflection system is completely solid state, utilizing state-of-the-art secondary breakdown phenomenon of silicon transistors. Design studies are underway on a new light link for fully remote control and increased flexibility during all modes of pulsed operation. New additions to the accelerator control system include a constant current power supply for the beam analyzer magnet, and a remote control system for the electrostatic beam shutter. To provide some temporary alleviation of the critical shortage of electronics and instrumentation work space at the Cockcroft-Walton facility, a 500-square-foot trailer has been located near the control room and is presently being outfitted for electronics support of the Cockcroft-Walton accelerator bay area.

DENSE PLASMA FOCUS (DPF) SOURCE

The search for a compact, intense pulsed neutron source has been narrowed down to devices employing the Z-pinch or Dense Plasma Focus principle developed in the course of the Sherwood program at Los Alamos. A similar Z-pinch device (cf. E. Beckner, Rev. Sci. Inst. 38, 507 [1967]) incorporates strip-line power transmission and dielectric-switch firing in place of the usual coaxial cables and air or vacuum spark gaps. The strip-line configuration offers the advantages of lower inductance (~7 nH for the condenser bank and strip line) and lower fabrication cost. The dielectric switch, a one-shot expendable item which costs \$3.50 and can be replaced in 15 seconds, offers distinct advantages in fabrication and maintenance costs over either vacuum spark gaps or atmospheric-pressure spark gaps.

Design drawings are presently being prepared for a 30-kilojoule DPF discharge tube ("gun") which can be coupled to the energy storage bank either by strip lines or by coaxial cables.

Consideration is being given to means of measuring the neutron yield of the DPF device. A silver activation counter (cf. LA-3498-MS) will be used to measure total absolute yield. A liquid scintillator-plus-fast photomultiplier system is being developed to study the yield vs. time profile of the DPF neutron pulse.

The DPF device will be built, tested and operated in the high bay area of the Fast Reactor Core Test Facility at Ten Site, LASL. Excellent shielding accommodations for full power DPF operation are already

provided at the Ten Site facility, which is generally very well suited for LASL's planned safeguards research and development program using Dense Plasma Focus sources.

MATHEMATICAL SIMULATION STUDIES OF KINETIC RESPONSE MEASUREMENTS

A series of theoretical analyses (T-Division) and projected computer studies (Tand N-6) are directed toward mathematical simulation of delayed neutron kinetic response to neutron irradiations (pulsed or modulated) on various types of composite and heterogeneous systems. This theoretical program should provide valuable guidance for experimental programs (at both the Cockcroft-Walton and DPF facilities) and should help to determine the precision of basic data required to achieve a given accuracy in isotope assay applications. Three approaches are being developed.

(1) Zero prompt lifetime approximation Within a few prompt neutron lifetimes after pulsed neutron irradiation of any (subcritical) fission system, the neutron level will adjust to a nearly constant value determined by the residual neutron multiplication of the system, and will remain at this "quasistatic" level until it is ultimately decreased by delayed neutron decay. The time rate of change of neutron density is approximately zero during this quasistatic time interval (which need only be short compared to the delayed neutron periods). The time behavior of the delayed neutrons is then determined by a Neumann

series, the coefficients of which are the solutions to a succession of steady-state distributed source calculations. The distributed source calculations are performed with the latest neutron transport codes DTF-IV and 2DF (one and two dimensional, multiregion, multigroup S<sub>p</sub> calculations).

#### (2) Monte Carlo calculations

Model pulse shapes in time, energy and angle will serve as sources for Monte Carlo calculations. Delayed neutron emission (six period groups) will be taken into account in conjunction with the fission process. The neutron response (energy, time and space) will then be determined for all times of interest on the delayed neutron time scale.

(3) Time dependent S<sub>n</sub> calculations
One and two-dimensional time-dependent S<sub>n</sub> calculational codes, multiregion, multigroup, with detailed (six period group) delayed neutron effects are being organized and programmed.

#### DETECTOR DEVELOPMENT

A survey of high efficiency, "flat"-energy-response neutron detection schemes is presently being conducted. Such detectors are needed for the measurement of delayed neutron group periods and abundances from fission induced by 14 MeV neutrons, as well as for general applications to safeguards technology. It is expected that the performance of several detection schemes and detector

geometries will be investigated using Monte Carlo computer techniques.

A Cf spontaneous fission neutron source was obtained in the form of a small amount of californium deposited on a platinum disc; the source was calibrated by absolute counting of source fissions and multiplying by the total (prompt plus delayed) neutron yield for  $Cf^{252}$ ,  $\overline{v} = 3.78 \pm 0.03$ , to obtain absolute neutron source strength. This calibrated source was then used to obtain an independent check of existing calibrations of other neutron sources used for detector calibration, etc., in the delayed neutron program of Group N-6. Neutrons from a Cf source are emitted with an accuratelyknown fission-spectrum energy distribution; because of the great usefulness and convenience of such sources in absolute detector calibration and other safeguards applications, a more intense Cf source has been ordered from Oak Ridge.

OTHER CONTRIBUTIONS TO NUCLEAR SAFEGUARDS RESEARCH AT LASL

#### Radiochemical Analysis (J-11)

The total number of fissions in four samples of  $U^{238}$  and four samples of  $Th^{232}$  irradiated at the CW accelerator was determined by standard counting of the 67 hour  $\beta$ -activity from Mo  $^{99}$  which was chemically separated from each irradiated sample. Measured  $\beta$ -yield is converted to number of fissions by an appropriate "K" factor determined from absolute fission counting measurements.

#### Alpha Counting of Thorium Foils (J-11)

Thorium foils (used in the fission counter for absolute delayed neutron yield measurements) were counted in a  $2\pi$  alpha counter in an attempt at accurate determination of foil masses. Unfortunately, decay product contamination has prevented definitive interpretation of the data.

#### Fission-Counter Foil Preparation (CMF-4)

Three foils each of U<sup>238</sup>, U<sup>235</sup>, U<sup>233</sup>, Th<sup>232</sup> and two foils of Pu<sup>239</sup> were prepared in the form of 1" diameter evaporated deposits on a platinum backing disc. The mass of material on each foil varied from 100 to ~300 micrograms.

## Thermal Neutron "Weighing" of Fissile Foils (P-2)

Thermal neutrons from the LASL Water Boiler reactor were used to "weigh," (by thermal-fission comparison counting relative to a  $U^{235}$  standard foil) the  $U^{235}$ ,  $Pu^{239}$  and  $U^{233}$  foils prepared by CMF-4.

#### Fission Counting of Cf<sup>252</sup> Source (N-2)

A low-geometry alpha counter was used to determine the spontaneous fission rate (and thereby total neutron emission) from a  $Cf^{252}$  source.

# Mass Spectrometric and Chemical Analysis (CMB-1, W-7)

Samples of U<sup>235</sup> were analyzed by mass spectrometry for uranium isotopic composition. Also samples of U<sup>235</sup>, U<sup>238</sup> and Th<sup>232</sup> were spectrochemically analyzed for trace concentrations of impurities (no significant impurities found).

#### Plutonium Sample Preparation (CMB-11)

Five of the fourteen plutonium samples being prepared by CMB-II have now been received by N-6. Samples are in the form of discs, 0.005" or 0.010" thick by 1" diameter, hermetically canned in aluminum.

#### **PUBLICATIONS**

- 1. "Nondestructive Detection, Identification and Analysis of Fissionable Materials," invited paper, USAEC Symposium on Safeguards Research and Development (unclassified and classified sessions), Argonne National Laboratory, June 26, 27, 1967.
- 2. "Decay of the 9.49-, 9.53- and 9.60-MeV Levels of O<sup>15</sup>," Phys. Rev. <u>155</u>, 1047-1053 (March 20, 1967).
- 3. "Delayed Fission Neutron Data in Reactor Physics and Design," invited paper, IAEA Panel on Delayed Fission Neutrons, International Atomic Energy Agency, Vienna, Austria, April 24-27, 1967.
- 4. "Nuclear Energy -- Its Future, and Its Control," panelist on program "Topic" (produced in conjunction with American Nuclear Society 1967 Annual Meeting in San Diego, California), KFMB-Radio-TV, San Diego, 2-3:00 p.m., June 15, 1967.