Delayed-Neutron Studies from the Thermal-Neutron-Induced Fission of Pu^{241}†

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The delayed-neutron activity resulting from the thermal-neutron-induced fission of Pu^{241} has been studied. The measured total delayed-neutron yield, determined from a comparison measurement between Pu^{241} and U^{235}, is 0.0154±0.0015 neutron/fission. The measured values for the individual group yields and the associated half-lives are: 0.000154±0.00004 *n/f*, 54.0±1 sec; 0.00365±0.0001 *n/f*, 23.2±0.5 sec; 0.00275±0.0004 *n/f*, 5.6±0.6 sec; 0.0062±0.0008 *n/f*, 1.97±0.1 sec; 0.0029±0.0003 *n/f*, 0.43±0.04 sec. The systematic behavior of delayed-neutron emission is discussed. The systematic study suggests that the variation of delayed-neutron yield with both mass number and atomic number is influenced more by changes in the fission-product charge distribution than by changes in the fission-product mass distribution.

INTRODUCTION

EXTENSIVE delayed-neutron measurements have been made for many fissile nuclides ranging from Th^{232} to Cf^{252}.[1] In 1957 Keepin *et al.*[2] published results of their measurements for isotopes of thorium, uranium, and plutonium. Their studies included delayed neutrons from thermal-neutron-induced fission and from fission induced by fast fission neutrons from the Los Alamos bare critical assembly "Godiva." For a given nuclide no significant differences were observed in the delayed-neutron yields between thermal-neutron-induced fission and from fast-neutron-induced fission. Large differences were observed in both the total and individual neutron group[3] yields between the various nuclides; however, the group periods were in general agreement for all of the nuclides studied. Keepin[4] also made a comprehensive study of possible delayed-neutron precursors, and as a result he selected two small areas of the periodic table just above the neutron-magic numbers 50 and 82, which should contain all delayed neutron precursors of appreciable yield. In 1958 Cox *et al.*[5] reported measurements on the delayed-neutron activity following the spontaneous fission of Cf^{252}. Two features of the Cf^{252} results were not in accord with Keepin's suggested precursor scheme. The half-life of the second delayed-neutron group, which should have resulted almost entirely from the decay of 24-sec I^{137}, was measured to be 20 sec. The 6-sec activity which should have been present from the decay of I^{138} was not observed. Cox *et al.* suggested that the precursor scheme might be considerably different for a nuclide as heavy as Cf^{252} and that the strong contribution of the iodine precursors in the lighter fissile elements might be replaced by new precursors of higher mass number and/or higher atomic number.

The purpose of this paper is to extend the measurements on the plutonium isotopes in the hope that it may lead to a better understanding of delayed-neutron emission.

EXPERIMENTAL PROCEDURE

The experiment consisted of irradiating a fissile sample with thermal neutrons and then counting the delayed-neutron activity as a function of time after irradiation. The experimental arrangement is shown schematically in Fig. 1. The thermal neutrons were extracted from the Argonne research reactor through a concrete and iron shielded collimator hole 1.27 cm in diameter and 100 cm long. The collimated neutron beam passed through a pneumatically operated shutter, a shielded neutron detector tank which contained the fissile sample, and was finally stopped by a "beam catcher." The shutter was used to interrupt the neutron beam at the end of the irradiation period. In the closed position the shutter interposed one m of paraffin in the path of the collimated neutron beam. The neutron detector consisted of 10 BF_3 counters immersed in mineral oil. The geometrical arrangement of the BF_3 counters was chosen so that the variation of detection

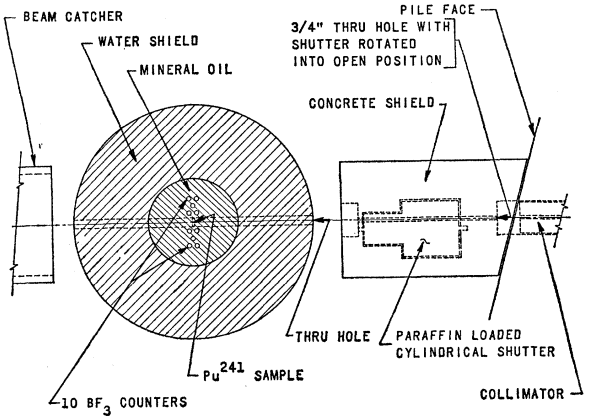

FIG. 1. Schematic diagram of the experimental arrangement.

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[1] For a resume of delayed-neutron measurements prior to 1956, see G. R. Keepin, *Progress in Nuclear Energy* (Pergamon Press, New York, 1956), Vol. I.

[2] G. R. Keepin, T. F. Wimett, and R. K. Zeigler, Phys. Rev. **107**, 1044 (1957).

[3] A delayed-nuetron group is defined as one component of a complex exponential decay. The ith group is expressed as $A_i e^{-\lambda_i t}$. The total yield is

$$N(t)=\Sigma_{i=1}^{n} A_i e^{-\lambda_i t}.$$

[4] G. R. Keepin, J. Nuclear Energy **7**, 13 (1958).

[5] S. Cox, P. Fields, A. Friedman, R. Sjoblom, and A. Smith, Phys. Rev. **112**, 960 (1958).

efficiency with neutron energy was within 15% of a "long counter" response from 30 kev to 1 Mev. The over-all neutron detection efficiency was approximately 5%. The neutron detector assembly was totally surrounded by 50 cm of water for shielding against the background neutrons in the reactor room. The fissile sample was enclosed in a cylindrical tube with thin aluminum windows and was placed in fixed position in the center of the neutron detector assembly. With the sample in position and with the shutter closed, the total background counting rate was approximately 150 counts/min with the reactor operating at a power level of 2 megawatts. The initial counting rate following an "infinite" irradiation of the fissile sample was approximately 50 times background. At the end of a 2.5-min irradiation the shutter was closed in approximately 0.2 sec and the decay of the delayed-neutron activity was followed by a 256-channel analyzer modified to serve as a time analyzer. The channel widths used were 0.1–1.5–2.0 and 4.0 sec. The long channel widths were used to determine the long half-life components and the background. When the multichannel analyzer reached channel 256, it automatically reset itself to channel zero and simultaneously opened the shutter to begin a new irradiation. Since the background could be determined only with the long channel widths, the short and long channel widths were used alternately on successive days. In this way the error due to any systematic change in the background was minimized. In practice there was a small change in the background counting rate over the four-week period during which all of the data were taken.

The delayed-neutron decay was measured for both Pu^{241} and U^{235}. The U^{235} measurement was made to check for any systematic error due to counter response. A comparison of the U^{235} results of this experiment with those of Keepin *et al.*[2] showed very good agreement.

The Pu^{241} sample, which was in the form of a thin 3.17-cm diameter electrodeposited film on thin platinum backing, was obtained from the Hanford laboratory. The total mass of the Pu^{241} sample was measured at this laboratory[6] by alpha counting to be 2.55 mg. Our measurement of the Pu^{241} mass and that reported to us by Hanford agreed within 1%, after correction for the decay of Pu^{241}.

Since the fissile samples were $1\frac{1}{4}$ in. in diameter, exposures were taken with x-ray film at various positions along the path of the neutron beam to insure that the entire sample was uniformly illuminated. The photographs showed that this was indeed the case.

Finally, the total number of delayed-neutrons per fission from the thermal-neutron-induced fission of Pu^{241} was measured relative to the known delayed-neutron yield of U^{235}.[2] Many measurements were made with each sample rotated to various positions to minimize any error due to nonuniformity of the fissile deposits.

[6] The author is grateful to Dr. D. K. Butler for the Pu^{241} mass determination.

RESULTS AND CONCLUSIONS

If it is assumed that only one beta-decay transition precedes the emission of a delayed neutron, then the distribution in time of the measured delayed-neutron activity following an "infinite irradiation" is given by

$$N(t)=\sum_{i=1}^{n} A_i \exp(-\lambda_i t), \tag{1}$$

where A_i is the relative abundance of the ith group, λ_i is the decay constant of the ith group, and n is the total number of delayed neutron groups. Equation (1) was fitted to the experimental data by the method of least squares with the aid of a large digital computer. The long-lived component was fitted to a single exponential decay and the result was subtracted from the total decay curve to give the contribution of the remaining delayed-neutron periods. Each of these were in turn fitted to a single exponential decay. In all, five delayed-neutron periods were observed. The 0.2-sec component measured by Keepin *et al.* for other nuclides was not observed in this experiment because the data could not be analyzed until after 0.5 sec decay time had elapsed.

The combined results of two independent runs for the delayed-neutron emission from Pu^{241} are given in Table I together with the results of Keepin *et al.*[2] for the thermal-neutron-induced fission of Pu^{239} and the fast-neutron-induced fission of Pu^{240}. The data are corrected for finite irradiation time and finite decay time between irradiations. The corrections are appreciable only for the 23-sec and 54-sec delayed-neutron groups. It is evident from the table that, within the stated statistical errors, the decay time for a given delayed-neutron group is not significantly different for the three isotopes. The absolute yield however exhibits very large changes. This behavior is in agreement with the empirical rule, first pointed out by Keepin,[4] that

TABLE I. Plutonium delayed-neutron emission.

Group index	Delayed-neutron yields[a] Pu^{239}[b]	Pu^{240}[b]	Pu^{241}
1	2.1 ±0.6	2.2 ±0.3	1.54± 0.4
2	18.2 ±2.3	23.8 ±1.6	36.5 ± 1
3	12.9 ±3.0	16.2 ±4.4	27.5 ± 4.0
4	19.9 ±2.2	31.5 ±2.7	62.0 ± 8.0
5	5.2 ±1.8	11.9 ±1.8	29.0 ± 3.0
6	2.7 ±1.0	2.4 ±0.5	···
Total	61 ±3	88 ±6	154 ±15
Group index	Delayed-neutron periods (seconds) Pu^{239}[b]	Pu^{240}[b]	Pu^{241}
1	54.28 ±2.34	53.56 ±1.21	54.0 ± 1
2	23.04 ±1.67	22.14 ±0.38	23.2 ± 0.5
3	5.60 ±0.4	5.14 ±0.42	5.6 ± 0.6
4	2.13 ±0.24	2.08 ±0.19	1.97± 0.1
5	0.618±0.213	0.511±0.077	0.43± 0.04
6	0.257±0.045	0.172±0.033	

[a] All values are (absolute number of delayed neutrons/fission) $\times 10^4$.
[b] The Pu^{239} and Pu^{240} data are from Keepin *et al.* (reference 2).

the absolute yield of the delayed-neutron groups increases with mass number for a given fissile element, while the decay times of the neutron groups show no significant change.

A graphical presentation of the absolute delayed-neutron group yields as a function of mass number from isotopes of Th, U, Pu, and Cf is given in Fig. 2. The data of Keepin *et al.*[2] are used for Th^{232}, $U^{233,235,238}$, and $Pu^{239,240}$. The Pu^{241} data are from this experiment and the Cf^{252} data are from Cox *et al.*[5] The total absolute delayed-neutron yields are presented in Fig. 3 for the same nuclides. The curves drawn through the experimental points are for purposes of emphasizing the systematic behavior. Several empirical observations

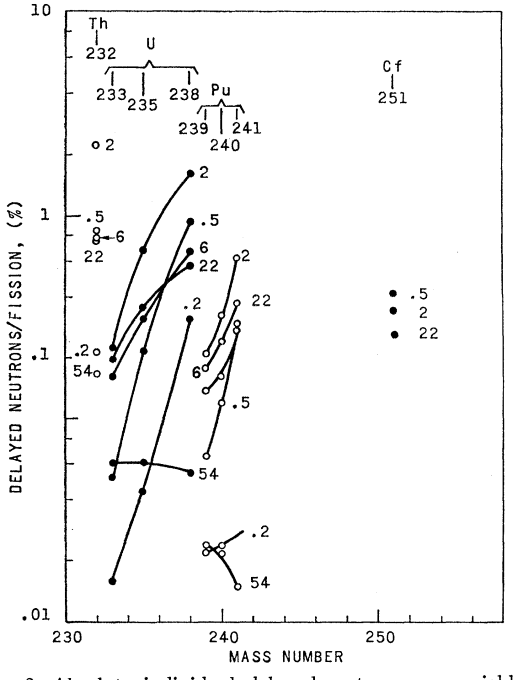

FIG. 2. Absolute individual delayed-neutron group yield for isotopes of Th, U, Pu, and Cf.[12] The numbers beside the experimental points and curves represent the approximate half-life for the associated delayed-neutron group. The curves drawn through the experimental points for U and Pu are for the purpose of emphasizing the systematic behavior.

can be made from the systematics of Fig. 2 and Fig. 3. For a given atomic number the delayed-neutron yield increases strongly with mass number except for the 54-sec period. For a given mass number there is a sharp decrease in delayed-neutron yield with increasing atomic number. The dependence on atomic number appears to be much stronger than the dependence on mass number. For example, the extrapolated Pu^{238} yields are roughly an order of magnitude smaller than the U^{238} yields. For an addition of two mass units however, the delayed-neutron yield increases only about a factor of three. An examination of the fission mass distributions[7] for the three Uranium isotopes represented here shows immediately that the change in the

[7] Seymour Katcoff, Nucleonics **16**, 78, April (1958).

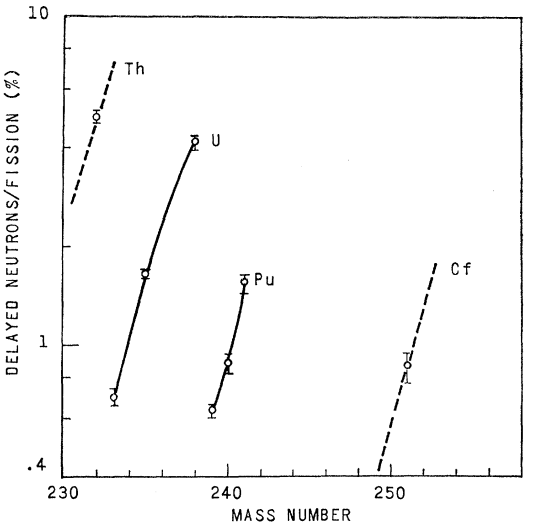

FIG. 3. Absolute total delayed-neutron yield for isotopes of Th, U, Pu, and Cf. The dashed curves through Th and Cf represent the possible behavior of the delayed-neutron yield of Th and Cf as suggested by the behavior of the delayed-neutron yield for U and Pu.

mass distribution between U^{233} and U^{238} is inadequate to account for the large increase in the delayed-neutron yield. A study of the mass distributions suggests that the mass yield in the region of the iodine precursors should remain relatively constant for the U isotopes while the yield in the region of the bromine precursors should be considerably smaller for U^{238} than for U^{233}.

The above discussion implies that the delayed neutron yield for a given fissile element is influenced primarily by changes in the fission-product charge distribution,[8–11] and that the sharp decrease in yield between U^{238} and Pu^{239}, and Th^{232} and U^{233} is due to an abrupt shift in the charge distribution resulting from the addition of two charges to the fissile nucleus.

The systematic trends of the delayed neutron yields from the U and Pu isotopes appear to be clear enough to justify approximate determination of the yields of unmeasured isotopes by interpolation and extrapolation. Such determination of delayed-neutron yields could be of importance in nuclear reactor studies.

The empirical systematic behavior of the delayed-neutron yields seems fairly well established. An accurate explanation of the delayed-neutron systematics must await a more precise determination of the fission-product mass distributions and, especially much better determination of the fission-product charge distribution.

[8] A. C. Wahl, J. Inorg. Nuclear Chem. **6**, 263 (1958).
[9] T. J. Kenneth and N. G. Thode, Phys. Rev. **103**, 323 (1956).
[10] E. P. Steinberg and L. E. Glendenin, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, Geneva, 1956), Vol. 7, Paper P/614.
[11] L. E. Glendenin, C. D. Coryell, and R. R. Edwards, *Radiochemical Studies: The Fission Products* edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 52, National Nuclear Energy Series, Plutonium Project Record, Vol. 9.
[12] The data for Figs. 2 and 3 were taken from references 2, 5, and this paper.