

# Prompt Fission Neutron Spectrum of $\text{Pu}^{241}$ †

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The energy distribution of prompt neutrons resulting from the thermal-neutron-induced fission of  $\text{Pu}^{241}$  is measured. Fast time-of-flight techniques are employed in the neutron energy range 0.3 to 6.0 Mev. Proton recoils in emulsions are utilized for the measurement of neutron energies from 1.6 to 7.0 Mev. The experimentally determined  $\text{Pu}^{241}$  fission neutron spectrum is well represented by the Maxwellian distribution,  $N(E) \propto E^{1/2} e^{-E/T}$ , where  $E$  is the neutron energy in Mev,  $N(E)$  the number of neutrons per unit energy interval, and  $T = 1.335 \pm 0.034$  Mev. The measured average  $\text{Pu}^{241}$  fission neutron energy is  $2.002 \pm 0.051$  Mev.

## INTRODUCTION

AS the result of a number of measurements the energy spectrum of prompt fission neutrons emitted following the fission of  $\text{U}^{235}$  is well known.<sup>1-7</sup> Similar experimental measurements of the fission neutron spectra of  $\text{U}^{233}$  and  $\text{Pu}^{239}$  have been reported.<sup>6,8-10</sup> In addition, the neutron spectrum resulting from the spontaneous fission of  $\text{Cf}^{252}$  has been measured.<sup>11,12</sup> The experimental results are, generally, consistent with the statistical theory of prompt fission neutron emission.<sup>13</sup> In order to obtain information that will extend our knowledge of the fission process and to provide data useful in applied work this study of the prompt fission neutron spectrum of  $\text{Pu}^{241}$  was undertaken.

## EXPERIMENTAL METHOD

Two sets of experiments, separated by about eighteen months, were carried out. The first group of measurements utilized proton recoils in nuclear emulsions to determine the spectrum of fission neutrons in the energy range 1.6 to 7.0 Mev. The fissile  $\text{Pu}^{241}$  sample consisted of  $\sim 3$  mg of metallic oxide. This sample was placed in a well collimated beam of thermal neutrons from the Argonne National Laboratory CP-5 reactor. The effective purity of the sample was such that  $>90\%$  of the fissions were due to  $\text{Pu}^{241}$ . The sample was surrounded by a set of Ilford C-2 emulsions. The fission neutrons leaving

the sample were incident on the emulsion surfaces at tangential angles of  $3-10^\circ$ . The emulsions were processed using temperature controlled development methods<sup>14</sup> and were impregnated with wood resin to reduce shrinkage. Proton recoil tracks within the emulsions were accepted for measurement if they fell within a pyramidal volume whose apex angle was  $\sim 20^\circ$  and whose axis was collinear with the fission neutron direction. Corrections were made for the probability of proton escape from the emulsion.<sup>15</sup> More than 2000 tracks resulting from  $\text{Pu}^{241}$  fission neutrons were measured. Under identical experimental conditions a similar number of tracks resulting from  $\text{U}^{235}$  fission neutrons were examined. Above a neutron energy of 1.6 Mev, the measured spectrum of  $\text{U}^{235}$  fission neutrons was in good agreement with the accepted distribution.<sup>1-7</sup>

The second set of measurements utilized fast time-of-flight techniques to determine the  $\text{Pu}^{241}$  fission neutron distribution throughout the neutron energy range 0.3 to 6.0 Mev.<sup>16</sup> Approximately 1 mg fissile samples of  $\text{U}^{235}$  and of  $\text{Pu}^{241}$ , both with an isotopic purity of  $>96\%$ , were placed within a gas scintillation counter.<sup>17</sup> This counter was constructed in such a manner that the  $\text{U}^{235}$  and  $\text{Pu}^{241}$  samples could be interchanged in the active volume without disturbing the geometric arrangement of the apparatus. A fission neutron flight path extended from the fissile source, through a conical collimator, 94 cm to a neutron detector. This detector consisted of a plastic scintillator coupled to a RCA 6342 photomultiplier tube.<sup>18,19</sup> Pulsed-beam time-of-flight studies had previously demonstrated that the collimator employed would cause essentially no distortion of the

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<sup>11</sup> Smith, Fields, and Roberts, Phys. Rev. **108**, 411 (1957).

<sup>12</sup> E. Hjalmar, H. Slatis, and S. Thompson, Phys. Rev. **100**, 1542 (1955).

<sup>13</sup> J. Terrell, Phys. Rev. **113**, 527 (1959).

<sup>14</sup> C. C. Dilworth, G. P. S. Occhialini, and R. M. Payne, Nature **162**, 102 (1948).

<sup>15</sup> L. Rosen, Nucleonics **11**, 32 (1953).

<sup>16</sup> L. Cranberg, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 4, Paper P/577.

<sup>17</sup> C. Eggler and C. M. Huddleston, Nucleonics **14**, 34 (1956).

<sup>18</sup> Pilot "B", Pilot Chemical Company, Waltham, Massachusetts.

<sup>19</sup> Radio Corporation of America, Tube Division, Trenton, New Jersey.

fission neutron spectrum.<sup>20</sup> The experimental arrangement is shown schematically in Fig. 1.

A well-defined beam of thermal neutrons incident on the gas scintillator induced fissions in the  $\text{Pu}^{241}$  or  $\text{U}^{235}$  samples. Some of the resulting fission neutrons proceeded down the collimated flight path and interacted with the neutron detector. The time between the response of the fission counter and the detection of the neutron in the plastic scintillator was converted, in an analog manner, to a voltage pulse. The distribution of voltage pulses and therefore neutron flight times was recorded with the aid of a multichannel pulse height analyzer. By interchanging the  $\text{Pu}^{241}$  and  $\text{U}^{235}$  samples within the gas scintillator a direct comparison of the velocity spectra of prompt fission neutrons from the two isotopes was obtained. This comparison method was free from errors associated with the absolute sensitivity of the neutron detector. A typical experimental distribution of fission neutrons from  $\text{Pu}^{241}$  is shown in Fig. 2.

#### EXPERIMENTAL RESULTS AND CONCLUSIONS

Theory, based upon Weisskopf's nuclear evaporation model, predicts a Maxwellian distribution of fission neutron energies in the laboratory system.<sup>13,21</sup> This distribution is of the form

$$N(E) \propto E^{\frac{1}{2}} e^{-E/T}, \quad (1)$$

where  $E$  is the neutron energy in Mev,  $N(E)$  is the number of neutrons per unit energy interval and  $T$  is a "temperature" constant. This theoretically predicted distribution is in good agreement with experimentally measured fission neutron spectra.<sup>13</sup>

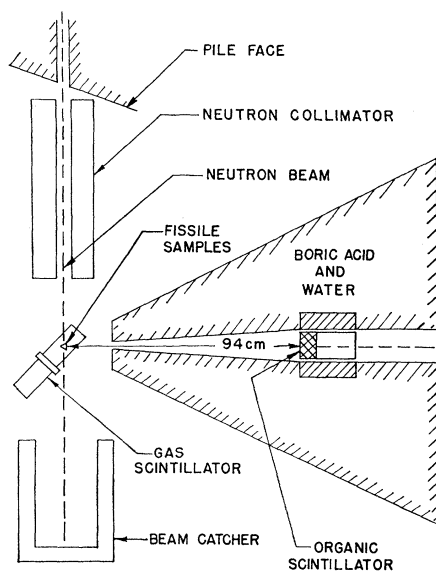


Fig. 1. Schematic diagram of the experimental apparatus.

<sup>20</sup> A. B. Smith and P. A. Moldauer, *Bull. Am. Phys. Soc.* **5**, 409 (1960).

<sup>21</sup> J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), pp. 365-374.

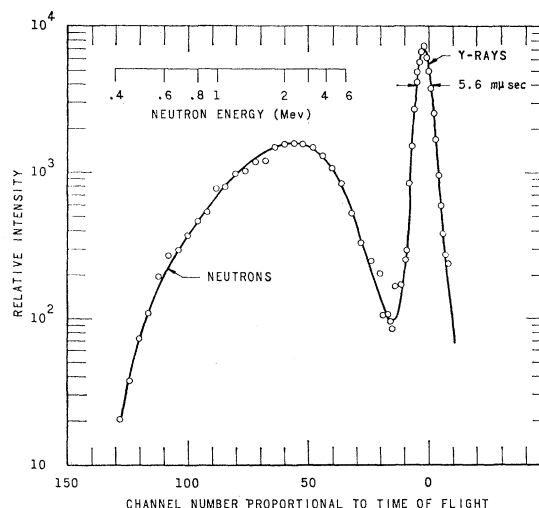


Fig. 2. Experimentally determined velocity spectrum of neutrons resulting from the thermal-neutron-induced fission of  $\text{Pu}^{241}$ .

The time-of-flight method, as utilized in this experiment, determines the energy-dependent ratio

$$R(E) = N(E)\text{Pu}^{241}/N(E)\text{U}^{235}, \quad (2)$$

of the  $\text{Pu}^{241}$  fission neutron spectrum to that of  $\text{U}^{235}$ . In order to convert these ratio measurements to  $\text{Pu}^{241}$  fission spectra it was assumed that the  $\text{U}^{235}$  fission neutron spectrum was Maxwellian in shape, [Eq. (1) above] with a temperature constant  $T=1.290$  Mev. This assumption was based upon the analysis of the experimental measurements summarized in Table I.<sup>13</sup> Utilizing the above assumption of time-of-flight ratio measurements were converted to  $\text{Pu}^{241}$  fission neutron spectra. The results of a typical measurement are shown in Fig. 3. The figure also presents the results of the emulsion measurements. The solid curve in the figure corresponds to a least squares fit of a Maxwellian distribution [Eq. (1)] to the experimental data. As is indicated in Fig. 3, the Maxwellian distribution is an excellent description of the experimentally measured  $\text{Pu}^{241}$  fission neutron spectrum. The weighted average Maxwellian temperature obtained from nine sets of data is  $T=1.335 \pm 0.034$  Mev. It follows directly that the average  $\text{Pu}^{241}$  fission neutron energy  $\bar{E} \equiv \frac{3}{2}T = 2.002 \pm 0.051$  Mev. The errors quoted pertain to this work only and are relative to the assumed  $\text{U}^{235}$  "standard" value  $T=1.290$  Mev. The results of this experiment are compared with other known fission spectra in Table I.

Statistical arguments have been used to relate the average number of neutrons emitted per fission,  $\bar{\nu}$ , to the average fission neutron energy,  $\bar{E}$ . Terrell obtained the following correlation.<sup>13</sup>

$$\bar{E} = 0.78 + 0.62(\bar{\nu} + 1)^{\frac{1}{2}}. \quad (3)$$

This expression combined with the measured  $\bar{E}(\text{Pu}^{241}) = 2.002$  Mev leads to a value  $\bar{\nu}_{\text{th}}(\text{Pu}^{241}) = 2.88$ . The fis-

TABLE I. Characteristics of fission neutron spectra.

Fissile nuclide	Energy range (Mev)	Method	$\bar{E}$ (Mev) <sup>a</sup> average energy	$\bar{E}$ (Mev) <sup>b</sup> combined average	Maxwellian temperature in Mev $T = \frac{2}{3}\bar{E}$ . Combined average. See Eq. (1)	Reference
$U^{235} + n_{th}$	0.180-3.0	Time-of-flight	$1.916 \pm 0.04$	$1.935 \pm 0.05$	$1.290 \pm 0.032$	4
	0.35-12.0	Photoplate	$1.952 \pm 0.013$			4
	1.0-12.0	Photoplate	$1.91 \pm 0.040$			6
	0.4-7.0	Photoplate	$2.055 \pm 0.040$			2
$U^{235} + n_{th}$	3.3-17.0	Proportional counter	$1.854 \pm 0.010$	$1.93 \pm 0.06$	$1.287 \pm 0.040$	1
	>2.7	Threshold detector	$\bar{E}(U^{235}) + 0.02 \pm 0.01$			c
	1.3-11.0	Proportional counter	$2.17 \pm 0.10$			9
	2.0-8.0	Photoplate	$2.23 \pm 0.13$			9
	>2.7	Threshold detector	$\bar{E}(U^{235}) + 0.06 \pm 0.02$			23
	1.0-10.0	Photoplate	$2.04 \pm 0.06$			6
	0.30-8.0	Time of flight and photoplate	$1.87 \pm 0.04$			8
$Pu^{239} + n_{th}$	>2.7	Threshold detector	$\bar{E}(U^{235}) + 0.07 \pm 0.02$	$2.00 \pm 0.05$	$1.333 \pm 0.033$	c
	>2.7	Threshold detector	$\bar{E}(U^{235}) + 0.08 \pm 0.02$			d
	1.0-12.0	Photoplate	$1.87 \pm 0.05$			6
	0.6-8.0	Photoplate	$2.275 \pm 0.04$			10
$Pu^{241} + n_{th}$	0.3-9.0	Time of flight and photoplate	$2.002 \pm 0.051$	$2.002 \pm 0.051$	$1.335 \pm 0.034$	This experiment
$Cf^{252}$ spontaneous	2.0-10.0	Photoplate	$2.12 \pm 0.24$	$2.2 \pm 0.1$	$1.466 \pm 0.066$	12
	1.4-7.0	Photoplate	$2.35 \pm 0.08$			11
	0.3-4.0	Time of flight	$2.13 \pm 0.05$			11

<sup>a</sup> Data from Terrell<sup>13</sup> where applicable. All averages result from least-squares fit of Maxwellian to experimental data.

<sup>b</sup>  $U^{235}$ ,  $Pu^{239}$ , and  $Cf^{252}$  values are from Terrell.<sup>13</sup> Other averages formed by weighting at present authors' discretion.

<sup>c</sup> J. A. Grundl and J. R. Neuer, Bull. Am. Phys. Soc. 1, 95 (1956).

<sup>d</sup> V. P. Kovalev, V. N. Andreev, M. N. Nilolaev, and A. G. Guseinov, J. Exptl. Theoret. Phys. (U.S.S.R.) 33, 1069 (1957).

sion neutron spectra of  $Pu^{241}$  and  $U^{235}$  measured by the time-of-flight method, as described above, were normalized to the same number of fission events recorded in the fission counter for the respective samples. The resulting normalized  $Pu^{241}$  and  $U^{235}$  spectra were inte-

grated to obtain the ratio

$$\bar{\nu}(Pu^{241})/\bar{\nu}(U^{235}) = 1.185 \pm 0.074. \quad (4)$$

Assuming  $\bar{\nu}(U^{235}) = 2.43 \pm 0.03$ , it follows that  $\bar{\nu}(Pu^{241})$  equals  $2.88 \pm 0.18$ .<sup>22</sup> This value is in fortuitously good agreement with the predictions of Eq. (3) above. Indeed, other work indicates that  $\bar{\nu}(Pu^{241})$  is  $\sim 2.95$ .<sup>23-26</sup> A critical test of Eq. (3) must await more precise experimental studies of more widely varying  $\bar{E}$  and  $\bar{\nu}$  values than have thus far been completed.<sup>27,28</sup>

#### ACKNOWLEDGMENTS

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<sup>22</sup> D. J. Hughes and R. B. Schwartz, *Neutron Cross Sections*, compiled by D. J. Hughes and R. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), Suppl. No. 1.

<sup>23</sup> M. G. Sowerby, Fast Reactor Data Conference, Harwell, England, 1960 (unpublished).

<sup>24</sup> Average U.K., U. S. A., and U.S.S.R. value; Atomic Energy Research Establishment, Harwell (unpublished report). (See also reference 28.)

<sup>25</sup> A. H. Jaffey, Argonne National Laboratory Report ANL-5396 (unpublished).

<sup>26</sup> G. de Saussure and E. Silver, Applied Nuclear Physics Division Annual Report for Period Ending September 10, 1956, ORNL-2081 (unpublished), p. 105.

<sup>27</sup> Argonne National Laboratory Report ANL-5800, Revision 1, 1961 (to be published).

<sup>28</sup> E. K. Hyde, University of California Radiation Laboratory Report UCRL-9036 (unpublished), Sec. 11.7, p. 241ff.

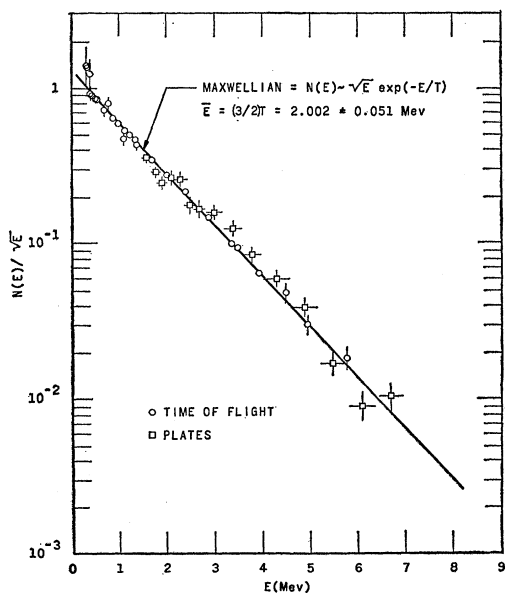


FIG. 3. Typical experimental measurement of the  $Pu^{241}$  fission neutron energy spectrum. Circular points are from time-of-flight measurements; square points are the result of the measurement of proton recoil tracks in nuclear emulsions. The solid curve represents a least-squares fit of a Maxwellian distribution to the experimental points.