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Measurement of the Average Number of Prompt Neutrons Emitted per Fission of ^{235}U Relative to ^{252}Cf for the Energy Region 500 eV to 10 MeV

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R. Gwin, R. R. Spencer, R. W. Ingle,* J. H. Todd,* and H. Weaver

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*Instrumentation and Controls Division

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ABSTRACT

The average number of prompt neutrons emitted per fission, $\bar{\nu}_p(E)$, has been measured for ^{235}U relative to $\bar{\nu}_p$ for the spontaneous fission of ^{252}Cf over the neutron energy range from 500 eV to 10 MeV. The samples of ^{235}U and ^{252}Cf were contained in fission chambers located in the center of a large liquid scintillator. Fission neutrons were detected by the large liquid scintillator. The present values of $\bar{\nu}_p(E)$ for ^{235}U are about 0.8% larger than those measured by Boldeman. In earlier work with the present system, it was noted that Boldeman's value of $\bar{\nu}_p(E)$ for thermal energy neutrons was about 0.8% lower than obtained at ORELA. It is suggested that the thickness of the fission foil used in Boldeman's experiment may cause some of the discrepancy between his and the present values of $\bar{\nu}_p(E)$. For the energy region up to 700 keV, the present values of $\bar{\nu}_p(E)$ for ^{235}U agree, within the uncertainty, with those given in ENDF/B-V. Above 1 MeV the present results for $\bar{\nu}_p(E)$ range about the ENDF/B-V values with differences up to 1.3%.

I. INTRODUCTION

Measurements of the neutron energy dependence of $\bar{\nu}_p(E)$, the average number of prompt neutrons emitted per fission, have been made for ^{235}U over the energy range from 500 eV to 10 MeV. The normalization was made relative to $\bar{\nu}_p$ for spontaneous fission of ^{252}Cf; therefore, the result given in this work is $R_p(E) = [\bar{\nu}_p(E)^{235}U]/(\bar{\nu}_p{}^{252}Cf)$. The present work is a continuation of experiments previously reported. In ref. 1 values of $R_p(E)$ for ^{239}Pu and ^{235}U were given for the neutron energy range from about 0.01 eV to a few MeV. It was noted, however, in ref. 1 that a large systematic uncertainty of about 3% should be placed on the results for ^{235}U in those measurements for the high neutron energy region ($E > 1$ keV) because of an inappropriate method used for determining the background in the measurements on ^{235}U, as will be discussed later.

It was also noted in Ref. 1 that the value derived in the thermal neutron energy region for $R_p(E)$ for ^{239}Pu was in good agreement with that of Boldeman,[2] but for ^{235}U the value presented in Ref. 1 was 0.8% larger than that of Boldeman.[2]

The ^{239}Pu fission chamber used by Boldeman[2] was similar to the one used in ref. 1 in that the thickness of the fissile deposit was less than 1.5 g/m^2. Boldeman's experiment on R_p for ^{235}U in the thermal energy region was performed using a fissile deposit of ^{235}U of about 8 g/m^2. For a deposit of 8 g/m^2 of ^{235}U, approximately 10 to 15% of the fission fragments will not escape the deposit with sufficient energy to be detected with a pulse-ionization chamber.[3] Because of geometrical considerations the selective loss of fission fragments may cause a reduction in the observed $\bar{\nu}_p$.

In order to obtain experimental data on the foil-thickness effect on $\overline{\nu}_p$, measurements have been made using two fission chambers; one chamber had a ^{235}U coating thickness of about 1 g/m^2 and the other chamber had a coating thickness of 20 g/m^2. The measured value of $\overline{\nu}_p$ using the thick coating, 20 g/m^2, was about 3% less than that measured using the chamber having a ^{235}U coating of 1 g/m^2. Asplund-Nilsson, Conde, and Starfelt[4] estimated a correction factor of +1.1% for their measured $\overline{\nu}_p$ for ^{238}U where the coating thickness was 40 g/m^2. The above estimate was based upon measurements of $\overline{\nu}_p$ for ^{252}Cf covered with varying thickness of ^{238}U.

Although ^{235}U is not included in current designs of fast breeder reactors, the present data are useful in evaluations, ENDF/B-V,[5] for example, where both differential and integral data are used. Lemmel's[6] evaluation of nuclear parameters at 2200 m/s indicated a 2% difference between the results of evaluations of $\overline{\nu}_p$(.0253 eV) for ^{233}U and ^{235}U which used data from integral measurements and those which used only differential data obtained at 2200 m/s. An increase in the ratio R_p(E) for ^{235}U of 0.8% would aid in resolving some of the difficulties in establishing a consistent set of nuclear parameters for ^{235}U.

II. EXPERIMENTAL METHOD

The present experiments on $\overline{\nu}_p$(E) utilized a liquid scintillator containing 0.22% by weight natural gadolinium to detect fission neutrons (with an efficiency of about 79%) and a fission chamber (efficiency ~95%) to define fission events. A description of the basic experimental techniques and some corrections made on the data are given by Hopkins and

Diven[7] and by Mather et al.[8] Many auxiliary experiments used in the analysis of the present data are discussed by Spencer et al.[9] The present experiments were performed using a neutron flight path of 83.4 m. The liquid scintillator had a volume of about 0.910 m^3 and the diameter of the through tube was about 13.3 cm.

The fission chamber was located at the center of the liquid scintillator in a through tube which traverses the scintillation tank. Neutrons from a pulsed source produced by the Oak Ridge Electron Linear Accelerator, ORELA, were collimated to impinge only on the fission foils. A fission event was identified by the simultaneous detection of the prompt fission gamma rays by the large liquid scintillator and a pulse from the fission detector system.

A bias equivalent to a pulse-height of about 900 keV gamma-ray energy was applied to the neutron detector. Pulses above this low (900 keV) bias were used to define coincidence with pulses from the fission chamber and pulses above this (neutron detector) bias were recorded in an interval following fission. In addition, another bias equivalent to about 2.8-MeV gamma-ray energy was applied to the neutron detector and pulses above this high bias were also recorded during the counting gate. The purpose of this high bias was to eliminate or minimize the contribution of delayed gamma rays to the counts following fission.

Fission neutrons are moderated in the scintillator, diffuse, and are absorbed in the gadolinium with the neutron absorption rate in the scintillator increasing after fission, reaching a peak at about 8 μs, and then decreasing exponentially such that about 90% of the neutrons are absorbed in 32 μs. A fixed dead time of about 0.1 μs on the neutron detector system sets the minimum possible delay between fission and the starting of the neutron counting gate. The gate starting time used was selected on the basis of Boldeman's[2] analysis of the delayed gamma-ray data following fission, the measured time dependence of pulses following fission, and measurements of $R_p(E)$ for different starting times of the neutron counting gate.

Measurements of the time distributions after fission of pulses from the neutron detector showed a decrease in the count rate for times up to about 1 μs. This observation is consistent with Boldeman's[2] suggestion that delayed gamma rays effect large liquid scintillator measurements of $\bar{\nu}_p$. In Boldeman's[2] evaluation of delayed gamma-ray data, the half life of the delayed gamma-ray emitters ranges from 0.02 to 80 μs and one emitter has a half-life of 0.62 μs.

A set of auxiliary experiments was performed to measure $\bar{\nu}_p$ for spontaneous fission of ^{230}Pu relative to $\bar{\nu}_p$ for ^{252}Cf. Counting gate starting times of 0.5 and 2.0 μs were used in these experiments and the observed value for R_p (^{240}Pu) was 0.3% lower in the case where the starting gate was delayed 2.0 μs than observed when the gate was started 0.5 μs following fission. Also in the present work on ^{235}U gate starting times of 0.6 and 2.0 μs after fission were used and a statistically significant smaller (~0.2%) value of R_p was observed when the starting gate was delayed 2.0 μs compared to R_p observed for the gate delayed for 0.6 μs. Because of these above observations most of the measurements performed for ^{235}U were made using a counting gate starting 2.0 μs following fission.

Figure 1 shows the pulse-height distribution in the large liquid scintillator measured for ^{235}U and ^{252}Cf fission neutrons. Four μs after

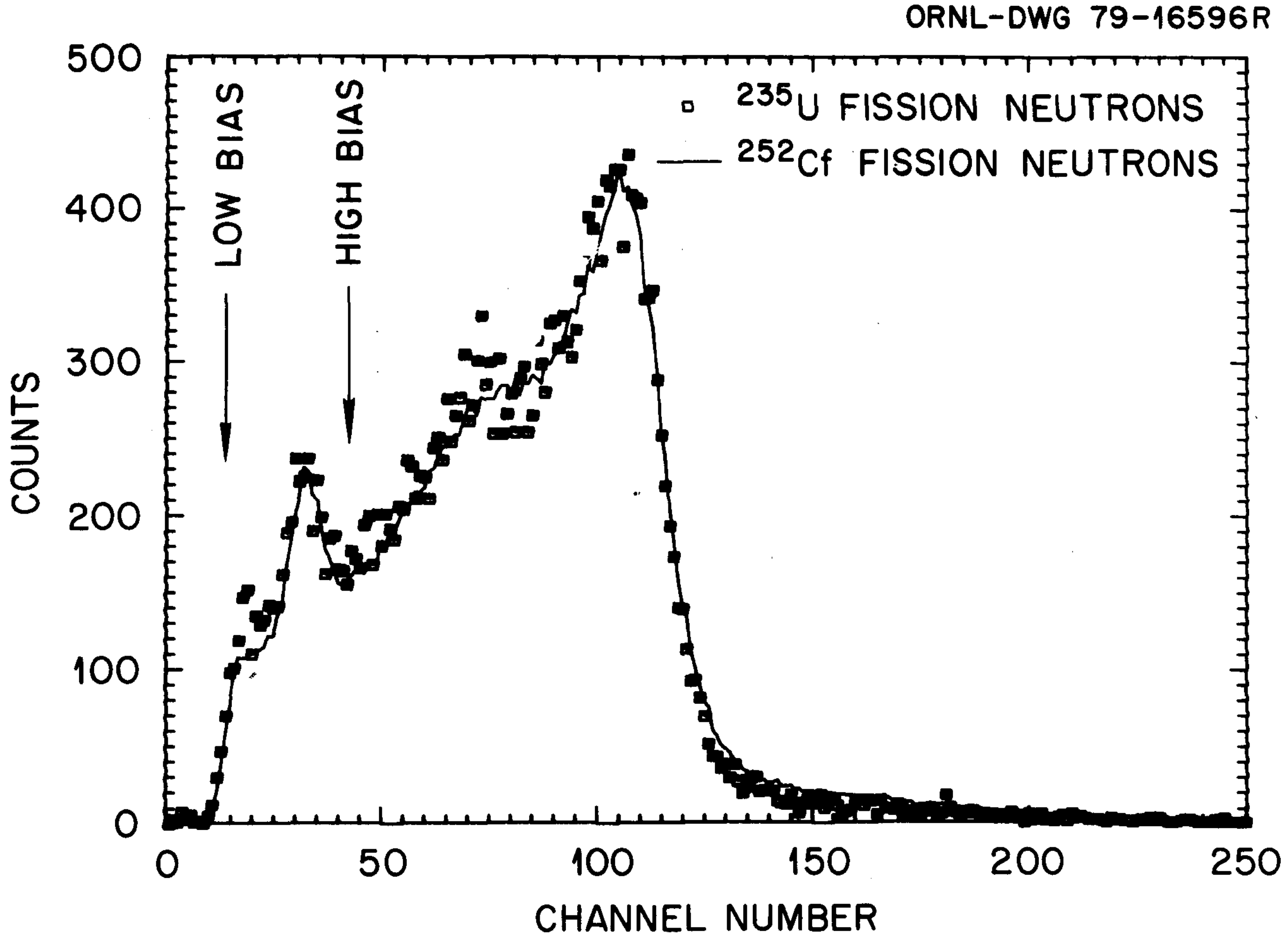

Figure 1. The measured pulse-height distribution from the large liquid scintillator system due to the absorbtion of neutrons from fission of ^{252}Cf and ^{235}U.

fission the pulse-height analyzer was enabled to record an event from the neutron detector. The integrating time was about 1 μs, and thus some pile up of events occurred. There is a larger probability that a background event will be recorded following ^{235}U fission than for ^{252}Cf fission since $\bar{\nu}_p$ for ^{235}U is less than $\bar{\nu}_p$ for ^{252}Cf, which can be seen in the lower pulse-height channels of Fig. 1. The small difference in the two pulse-height distributions above channel 125 may be due to the increased probability of neutron pulses piling up in 1 μs for ^{252}Cf fission compared with ^{235}U fission because of the difference in the two values of $\bar{\nu}_p$.

Two fission chambers containing ^{235}U were used in the experiments. One chamber, used to define $R_p(E)$ for ^{235}U, contained foils of aluminum 0.0025-cm thick coated with ^{235}U to a thickness of about 1 g/m^2. Experiments were also performed using a second chamber having plates coated to 20 g/m^2 of ^{235}U. Normalization of the $\bar{\nu}_p(E)$ data was made relative to $\bar{\nu}_p$ for ^{252}Cf, contained in a fission chamber called the primary standard in ref 1. A ^{252}Cf fission chamber used by Spencer et al.[9] in the measurements of absolute $\bar{\nu}_p$ for ^{252}Cf served as a secondary standard to monitor the present experiments. It was noted in ref. 1 that the observed $\bar{\nu}_p$ for the secondary standard was 0.25% greater than that observed for the primary standard. During the experiments the ^{252}Cf fission chamber, secondary standard, was positioned outside the neutron beam at one end of the ^{235}U fission chamber.

Two methods were used to estimate the background recorded in the foreground counting gates. In one technique a random pulse generated by a radioactive source started a 32 μs counting gate to measure the background.

In the second method a neutron detector positioned in the neutron beam was used to generate a counting gate. Neutron bursts generated by the ORELA may be expected to vary from pulse-to-pulse. Part of the background in the experiments is due to neutrons, for example, neutrons scattered from the fission chamber, and this background may be expected to be proportional to the neutron intensity. Since the probability of a fission is proportional to the neutron intensity, the use of a neutron detector to generate a gate for background analysis of the ^{235}U data is appropriate. For ^{252}Cf, a spontaneous fission source, the random background method is appropriate. A pulse ionization chamber filled with argon (80%) and BF_3 (20%) was placed about 82 m from the neutron source and used as a "beam weighted" background gate generator.

In the present experiments the neutron intensity at the sample position was adjusted such that the maximum background was about 0.5 counts per 32 μs. In the neutron energy region where the background was 0.5 counts per 32 μs, the corresponding number of neutrons detected from ^{235}U fission was in excess of 2.5 counts in the 32 μs counting gate.

III. UNCERTAINTIES IN AND CORRECTIONS TO THE DATA

Uncertainty in Using a ^{252}Cf Standard Sample

In ref. 1 it was noted that the observed value of $\bar{\nu}_p$ for ^{252}Cf was different by 0.25% for two fission chambers which had different geometry. This discrepancy has still not been explained and a systematic uncertainty of 0.25% should be applied to the present measurements because of this discrepancy.

Correction for Displacement of the Fission Samples from the Center of the Large Liquid Scintillator

Measurements of the average neutron detection efficiency for ^{252}Cf fission neutrons were made for various displacements of the ^{252}Cf standard sample from the center of the through tube. The results of these measurements were used to correct the measured values of $R_p(E)$ for displacement of the fissile isotope and the ^{252}Cf from the center of the through tube. A correction to $R_p(E)$ of -0.3 ± 0.06% was established for the low bias and -0.5 ± 0.10% for the high bias for the geometry used in the present work. For comparison, a 10-cm displacement of the ^{252}Cf source from the center reduced the observed values of $\overline{\nu}_p$ by about 0.5% and 0.9% for the low and high bias, respectively, and the corresponding values for a 25-cm displacement were 6% and 8.4% for the low and high bias, respectively.

Figure 2 shows the pulse-height distributions from the large liquid scintillator system due to neutron absorption. Two cases are shown; in one case the neutron source was located at the center of the through tube and in the other one the source was displaced about 23 cm from the center. Note that for an electronic bias around channel 40 ($\sim$2.8 MeV equivalent pulse height) the fraction of the pulses above that bias is less in the case where the neutron source is displaced from the center of the through tube.

The uncertainty on the correction to the high bias results for displacement of the ^{252}Cf from the center of the through tube is about the same size as the correction to $R_p(E)$ for delayed gamma rays when the counting gate start time is 2 μs after fission.

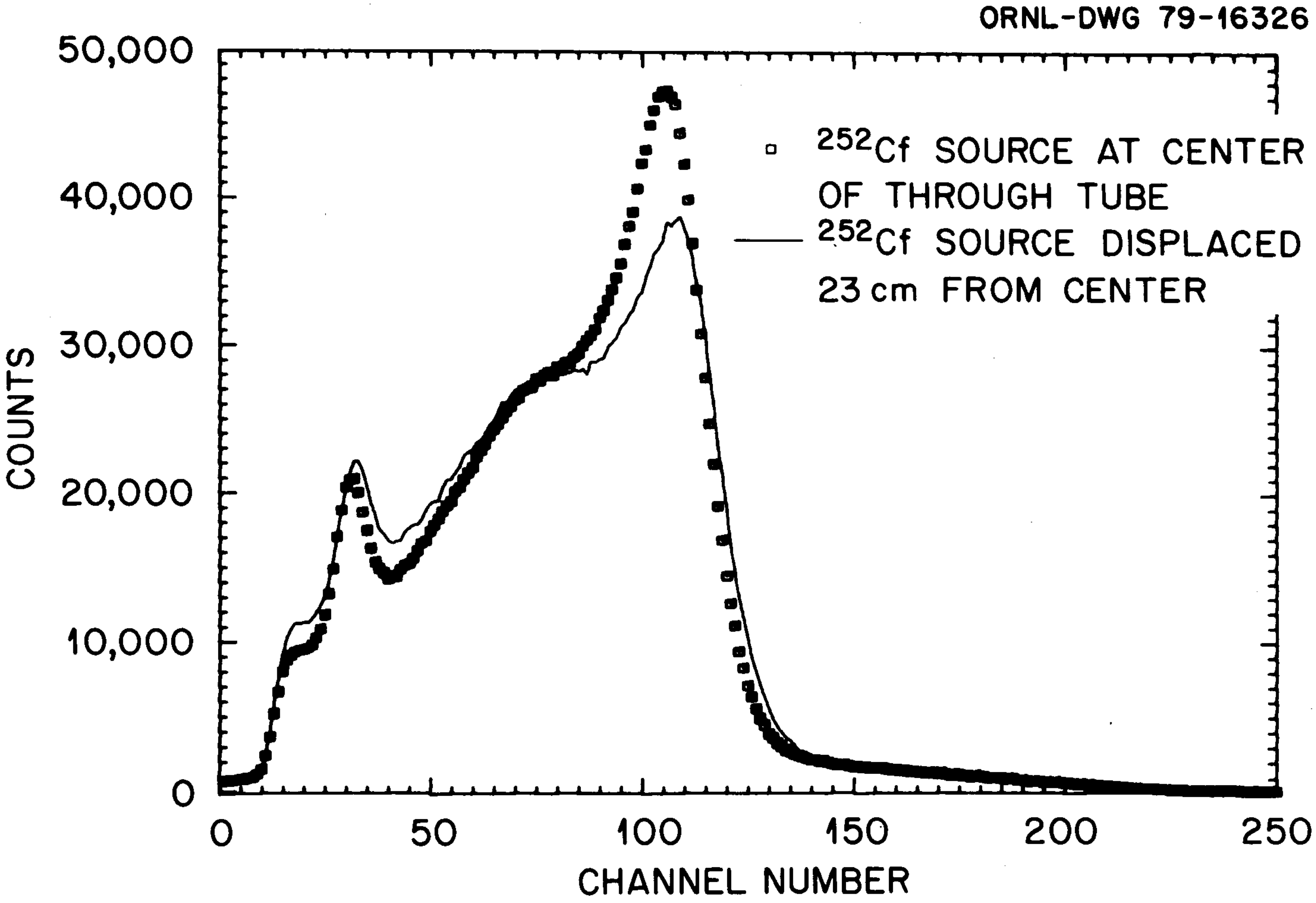

Figure 2. The effect of the fission neutron source location on the measured pulse-height distribution from the large liquid scintillator.

Correction for False Fissions

The random coincidence of a pulse from the fission chamber caused by the known alpha particle activity or noise with a pulse from the neutron detector initiates a counting gate which is called a false fission. The false fission rate was estimated by generating a random signal (mock alpha) and treating it in the electronic system as a fission chamber pulse. A coincidence of this mock alpha pulse and a scintillator pulse permitted a measure of the false fission rate as a function of time after the neutron burst. The required correction increases rapidly as the neutron intensity decreases at low neutron energy due to the ^{10}B placed in the neutron beam to serve as a neutron overlap filter. At 500 eV the correction factor for false fissions was about 2.5% and at 2.5 MeV it was about 0.3%. An uncertainty of 10% of the correction has been incorporated in the uncertainty analysis.

Corrections for Backgrounds in the Neutron Detector

Backgrounds from the large liquid scintillator during the counting gates started by fission events are due to local radioactivity, counts introduced by operation of the ORELA (scattered neutrons for example) to delayed gamma rays, and to delayed neutrons. The effect of delayed neutrons on the present results is negligible because of the low yield of delayed neutrons (1%) and the long half lives ($>10^{-3}$ s) of their precursors.

Delayed gamma rays are not measured in the methods of estimating the background used in the present work. Data on the half lives and cascade energies of delayed gamma rays from fission suggested by Boldeman were folded with the measured response of the present liquid scintillator to gamma rays to obtain correction factors for the present work. A correction factor of -0.1% was calculated for $R_p(E)$ for ^{235}U (gate starting 2.0 μs after fission) and an uncertainty of ±50% of the correction has been assumed. An optional high electronic bias was also used in the neutron detector system which was sufficiently high to reject pulses resulting from known delayed gamma rays.

Correction for Pulse Pile Up

The recovery time of the detector system for the large liquid scintillator following a pulse was about 0.075 μs. A fixed deadtime of 0.095 μs was imposed upon the neutron detector system. Correction of the data for pulse pile up (deadtime) was made in the manner given by Ribrag et al.[10] The correction factor obtained for the ratio $R_p(E)$ for ^{235}U was about 0.3% in the low-energy region ($E < 100$ keV) of the present work. Boldeman[2] obtained a correction of -0.3% for $R_p(E)^{2235}U$ in the thermal energy region. An uncertainty of 30% has been assumed for the deadtime correction factors.

Correction for Fission Spectrum Differences

A calculation of the relative efficiency of the liquid scintillator for capturing fission neutrons from ^{252}Cf and ^{235}U was made by Ullo.[11,1] These calculations yield a correction factor for $R_p(E)$ of -0.13 ± 0.04% for the present work using average fission neutron energies E of 2.11 and 1.99 MeV for ^{252}Cf and ^{235}U, respectively. A correction factor of -0.16% was estimated for $R_p(E)$ using values of E given in ENDF/B-V[5] of 2.18 and

2.03 for ^{252}Cf and ^{235}U, respectively. The correction factor of -0.13 ± .04% has been used in the present work.

Correction for Missed Fissions

Fission events may be missed because either the fission chamber did not detect the fragments or because the large liquid scintillator did not detect the prompt gamma rays from fission. Fission fragments may not be detected because they do not escape the source deposit or the bias on the fission chamber is higher than the pulse produced by the fragments which do escape the deposit.

Measurements by Spencer et al.[9] for ^{252}Cf show that for the bias used on the large liquid scintillator in the present work the requirement of detecting the prompt fission gamma rays results in an increase of about 0.03% in the observed $\overline{\nu}_p$. No uncertainty has been assigned to $R_p(E)$ for ^{235}U due to fissions missed by the neutron detector. Corrections to $R_p(E)$ for fissions not detected by the fission chamber are more complex than for the case where the prompt fission gamma rays are missed.

Neutrons entering the large liquid scintillator have probabilities of being absorbed in the scintillator which depend upon both the energy of the neutron and the angle with respect to the through tube from which the neutrons enter the scintillator. Cramer[12] performed Monte Carlo calculations with neutrons representative of the ^{252}Cf fission spectrum originating at the center of the scintillator and entering the tank at various angles with respect to the axis of the through tube. These calculations showed that the absorption probability varied from a value of 99.5% at 90° to 93% at 18°. For an isotropic source the probability of neutron absorption was 97.3%. The above calculations did not include the possible variation in the escape of the neutron capture gamma rays nor the possible change in the light collection efficiency of the detector with angle. Figure 3 shows the results of Cramer's[12] calculations along with a schematic drawing of the neutron detector and the fission chamber system. An exploded view in Fig. 3 of a fission foil shows how a finite deposit of the fissile isotopes results in an anisotropic distribution of fission fragments escaping the deposit and, therefore, an anisotropic distribution of neutrons about the fission source for detected fission events if the path of fission neutrons is correlated with the path of the fission fragments. Experimental data obtained by Fraser[13] on ^{239}Pu shows a correlation of the neutron path with the path of the fission fragment and about a factor of four more neutrons traveling along the path of the fragment than at 90° to the path of the fragment.

Since the ^{252}Cf fission chamber used to normalize the present data on $\overline{\nu}_p(E)$ for ^{235}U has a high efficiency, about 98%, it is expected that the fission neutron distribution will be close to isotropic. Usually fission foils are oriented such that the normal to the surface of the fissile foil is parallel to the axis of the through tube. For foils of finite thickness fission fragments moving with angles between 90° and some smaller angle (Fig. 3) will not escape from the foil with sufficient energy to be detected. Therefore, gates for counting fission neutrons will not be generated for those fissions in which the probability of detecting the neutrons is the largest.

For a fissile deposit of 1 g/m^2 of ^{235}U, it has been estimated that about 5% of the fission events are missed. If 5% of the fission

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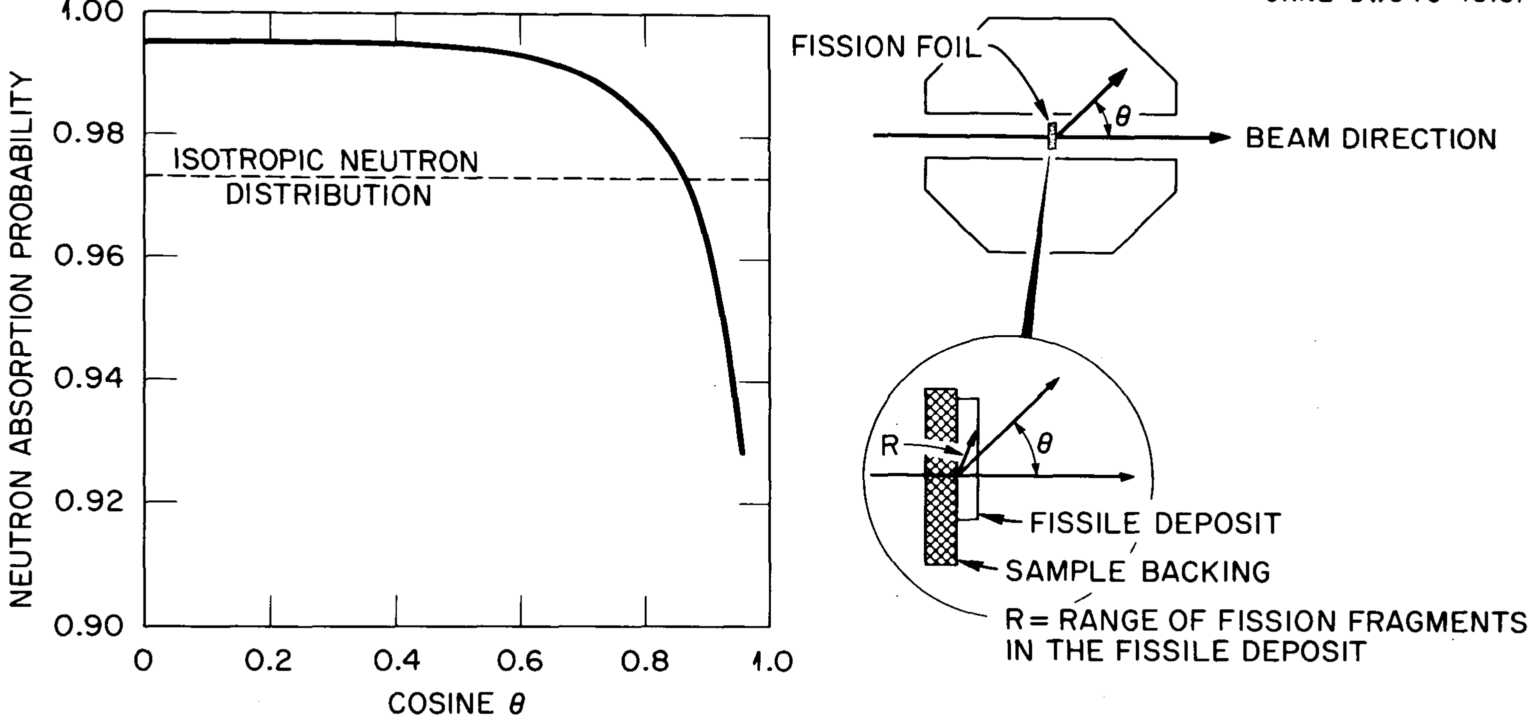

Neutron Absorption Probability for ^{252}Cf Fission Neutrons vs the Angle of Entry θ into the Scintillator.

Figure 3. The neutron absorption probability of the large liquid scintillator versus angle of neutron entry and the effect of thick fission foils on the angle of entry.

events are missed and if the neutrons in these events were moving at angles near 90°, see Fig. 3, then the corresponding neutron absorption probability estimated from Cramer's[12] calculations was 97.2% in contrast to the 97.3% obtained for the isotropic distribution of neutrons. Because of the uncertainty in the above estimate no correction to the present data has been made. A precise calculation of the effect of foil thickness on $\bar{\nu}_p$ would require a description of the fragment losses in the foil, a description of the actual neutron angular distribution about the path of the fragment (such as given by Fraser[13] for ^{239}Pu), and a detailed calibration of the neutron detector.

Auxiliary measurements in the present work have shown that for ^{252}Cf fission chambers the pulse-height distribution varies, as would be expected, with the angle the path of the fragment makes with respect to the normal to the foil. The fragment angle in the experiment was selected by mounting a NE-213, pulse-shape discriminator neutron detector about 1 m from a ^{252}Cf fission chamber and varying the normal to the plane of the fission foil with respect to the line joining the neutron detector and the fission foil. Figure 4 shows the pulse-height distribution measured for the fission chamber system for angles of 0°, 60°, and 90° and these three distributions have been normalized to the same number of observed counts. The three pulse-height distributions shown in Fig. 4 have a similar shape but the data do suggest that the application of a bias to the fission chamber can change the neutron distribution, for detected fissions, about the source and therefore may change the observed $\bar{\nu}_p$. It is noted in Fig. 4 that more pulses are observed in the low pulse-height region, between channel 6 and 20, for the distributions measured

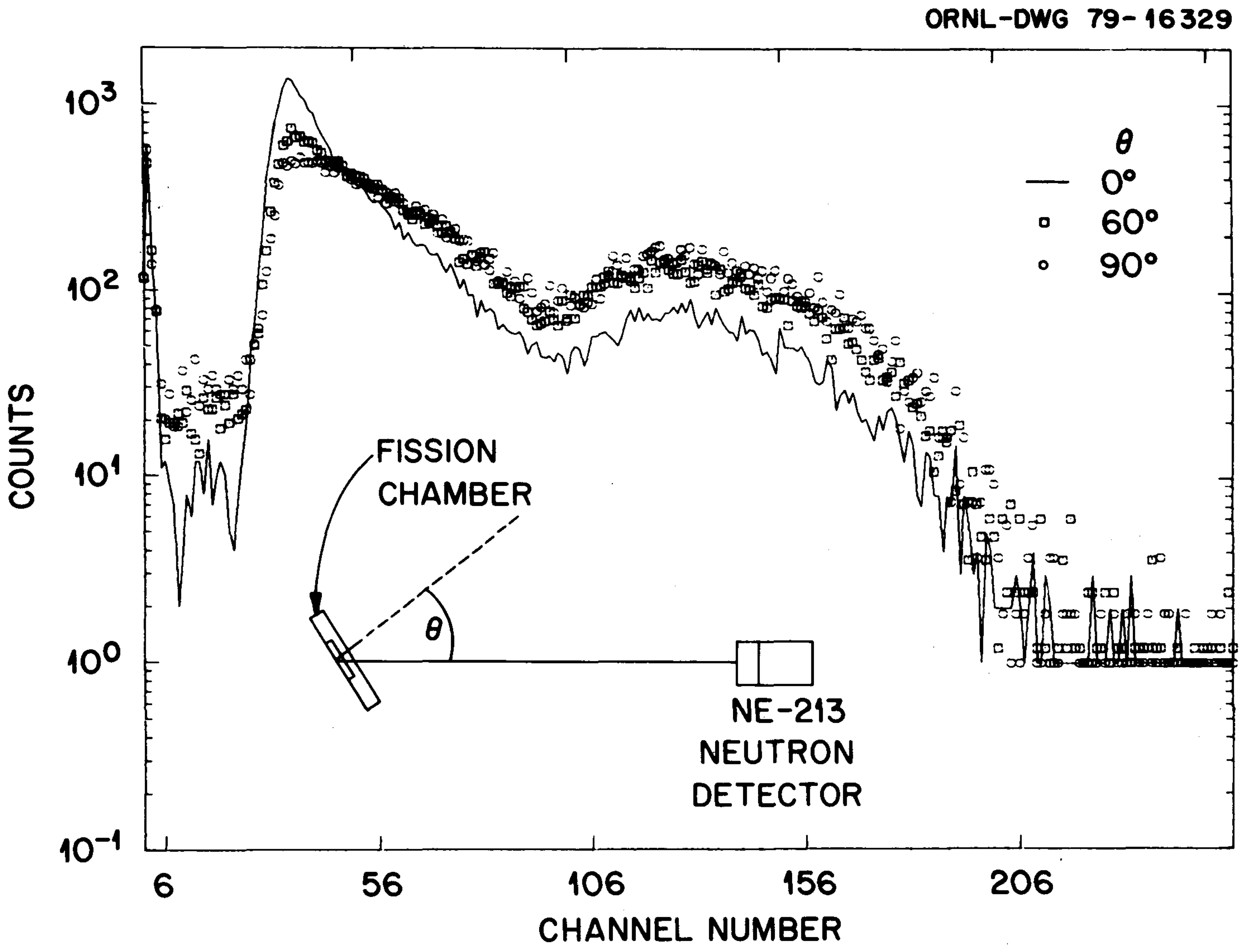

Figure 4. The measured pulse-height distribution from the ^{252}Cf fission chamber system for selected fission neutron directions.

at 60° and 90° than for the one measured at 0°. This above observation may be due to the surface condition of the fission foil. For a bias on the fission chamber, at approximately channel 12 in Fig. 4, a pulse from the chamber was observed 97.1 ± .1% of the times a neutron was detected for an angle of 0°, and the corresponding value for an angle of 90° was 96.1 ± .1%.

Variations in $\bar{\nu}_p$ for ^{252}Cf with fission chamber bias have been measured by Boldeman,[2] Mather et al.,[8] and in the present experiments. In describing the results of the experiments $\bar{\nu}_p$ will be given relative to that measured in the particular experiment at "zero" bias on the fission chamber. Mather et al.[8] observed a monotonic decrease in $\bar{\nu}_p$ as the bias on the fission chamber was increased and for a bias equivalent to 0.8 of the maximum pulse-height the observed $\bar{\nu}_p$ decreased by 2%. Boldeman observed a 0.2% increase in $\bar{\nu}_p$ when 50% of the fissions were below the bias and for 60% or more, up to 95%, of the fissions below the bias $\bar{\nu}_p$ was the same as observed with "zero" bias. In the present work $\bar{\nu}_p$ decreased 0.1% when the bias on the fission chamber system eliminated 10% of the fission events, $\bar{\nu}_p$ increased by 0.2% when 50% of the fissions were detected, and for 80% of the fission events below the bias $\bar{\nu}_p$ decreased by 1%.

The results of the above experiments on the variation of $\bar{\nu}_p$ for ^{252}Cf with the electronic bias applied to the fission chamber detector system may overlap within their respective uncertainties. Mather et al.[8] suggest that the variation of $\bar{\nu}_p$ with fission chamber bias is probably a consequence of the relation between the fragment kinetic energy and the number of neutrons emitted by that fragment. If the above suggestion by

Mather et al.[8] is responsible for the observed variation in $\bar{\nu}_p$ with fission chamber bias then the experimental results above should agree since the measured quantity was the source strength $\bar{\nu}_p$ for a given bias. It seems established that the fissile deposit in fission chambers and the bias on the fission chambers can influence the observed $\bar{\nu}_p$. The size of the perturbation will depend upon the properties of the particular large liquid scintillator and the fission chamber.

Correction Due to Anisotropy of Fission Fragment Emission

Davis et al.[14] made an estimate of the effect that anisotropy of the fission fragments has on the efficiency of ^{235}U fission chambers. In this analysis[13] the anisotropy factor A was estimated to increase from near unity at 400 keV to more than 1.1 at 936 keV. A value of 1.1 for A predicts 10% more fissions with an angle of 0° with respect to the beam than at 90° with respect to the beam. This change in anisotropy has an effect on $\bar{\nu}_p$ which is similar to that discussed for missed fissions. Mather et al.[8] give a discussion of the effect of anisotropy of the fission fragments on liquid scintillator measurements, and estimate that a correction of +0.2% should be made to their data at 7 MeV because of changes in anisotropy of the fission fragments. Below 7 MeV Mather et al.[8] considered that no correction for fission fragment anisotropy was needed. Walsh and Boldeman[15] assumed that no correction was needed for anisotropy of the fission fragments over the range of their experiments up to about 2 MeV.

In the present experiments different values for $R_p(E)$ are obtained for neutron energies above 5 MeV using the low and the high bias on the neutron detector. As will be discussed later, this above discrepancy may be electronic in origin; however it may be due to physical differences in the fissioning nucleus including anisotropy of the fission fragments or to delayed gamma rays. No corrections for anisotropy have been made for the present data.

The systematic uncertainties combine to yield an uncertainty of about 0.3%.

IV. PRESENTATION OF THE DATA

Table 1 lists measured values of $R_p(E)$, using the low bias on the neutron detector, for ^{235}U for neutron energy intervals in the range 500 eV to 10 MeV. Unless specifically stated otherwise the values of $R_p(E)$ pertain to data obtained using the low bias on the neutron detector. The uncertainty shown for each value of $R_p(E)$ is comprised of the statistical uncertainty as well as that due to false fissions. The 0.3% systematic uncertainty should be folded with that given in Table 1 to obtain an estimate of the total uncertainty. For the lowest energy interval in Table 1, the uncertainty in $R_p(E)$ due to false fissions is about 0.25%, while at about 600 keV the corresponding uncertainty on $R_p(E)$ is 0.02%.

Figure 5 shows the present experimental values for $R_p(E)$ for ^{235}U as a function of neutron energy over the range 500 eV to 2 MeV. Also shown in Fig. 5. are values of $R_p(E)$ given by Boldeman, Frehaut, and Walsh.[16] This latter work[16] includes the results of measurements by Walsh and Boldeman,[15] and Soleilhac et al.[17] The experimental values of $R_p(E)$ from ref. 16, which are shown in Fig. 5., have been multiplied by the factor 1.008 as explained below. Figure 6 shows the present values for

Table 1. Experimental Values of $\bar{\nu}_p(E)$ for ^{235}U Relative to $\bar{\nu}_p$ for ^{252}Cf.

E1 (MeV)	E2 (MeV)	$R_p(E)$	Delta $R_p(E)$*
0.0005	0.0011	0.6398	0.0030
0.0011	0.0051	0.6395	0.0012
0.0051	0.0101	0.6417	0.0017
0.0101	0.0516	0.6412	0.0023
0.0516	0.1041	0.6384	0.0032
0.1041	0.2116	0.6472	0.0017
0.2116	0.3177	0.6571	0.0029
0.3177	0.4201	0.6501	0.0023
0.4201	0.5293	0.6557	0.0027
0.5293	0.6206	0.6616	0.0034
0.6206	0.7379	0.6621	0.0051
0.7379	0.8574	0.6668	0.0052
0.8574	0.9672	0.6714	0.0111
0.9672	1.0525	0.6579	0.0087
1.0525	2.1628	0.6876	0.0031
2.1628	3.2637	0.7244	0.0045
3.2637	4.5430	0.7670	0.0058
4.5430	6.0730	0.8345	0.0069
6.0730	8.5330	0.9263	0.0060
8.5330	11.1150	1.0116	0.0081

*A systematic uncertainty of 0.3% must be folded with delta $R_p(E)$ to obtain the total uncertainty.

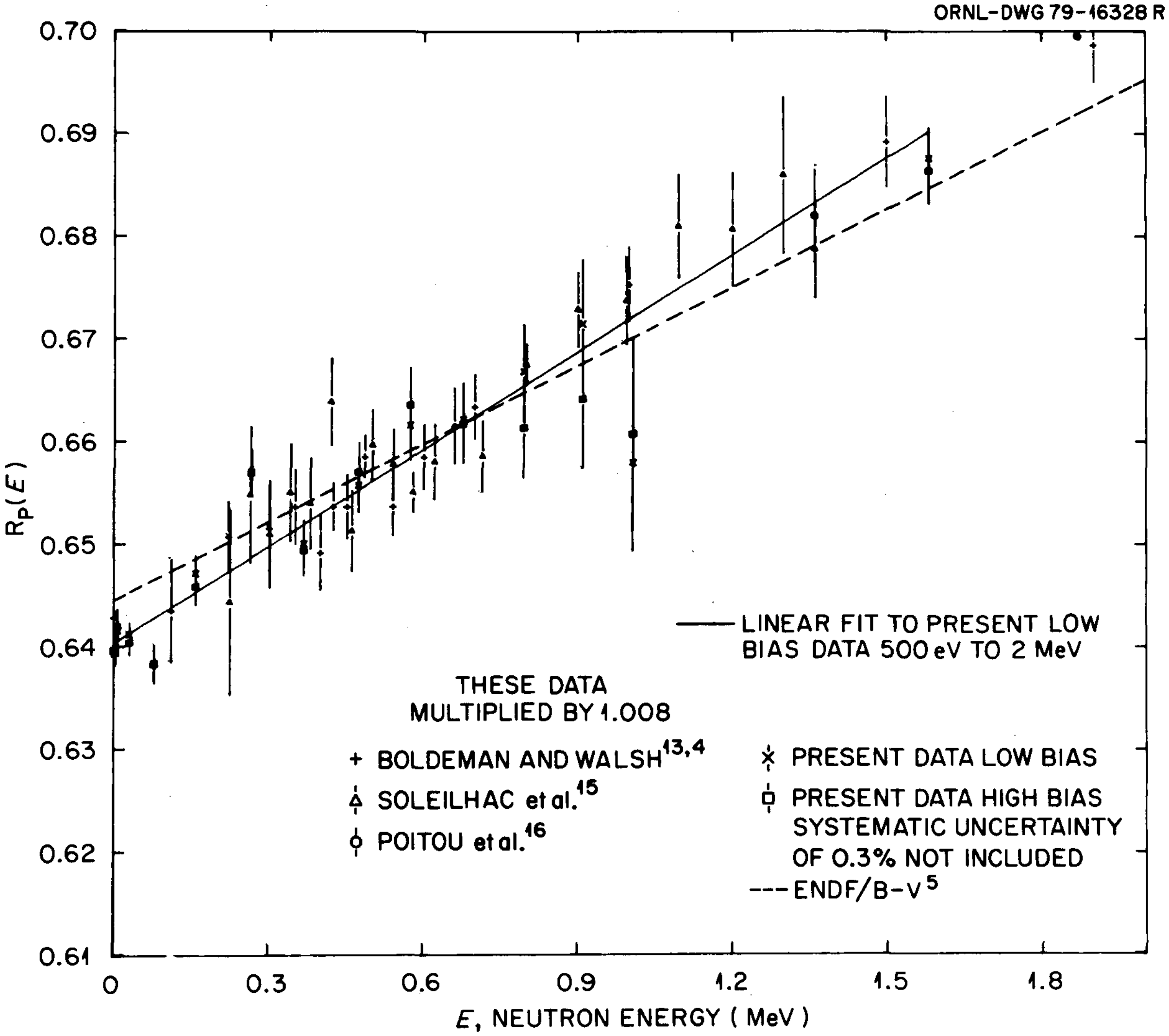

Figure 5. Experimental values of $R_p(E)$ for ^{235}U for the energy range 0 to 2 MeV. The results of ENDF/B-V are also shown.

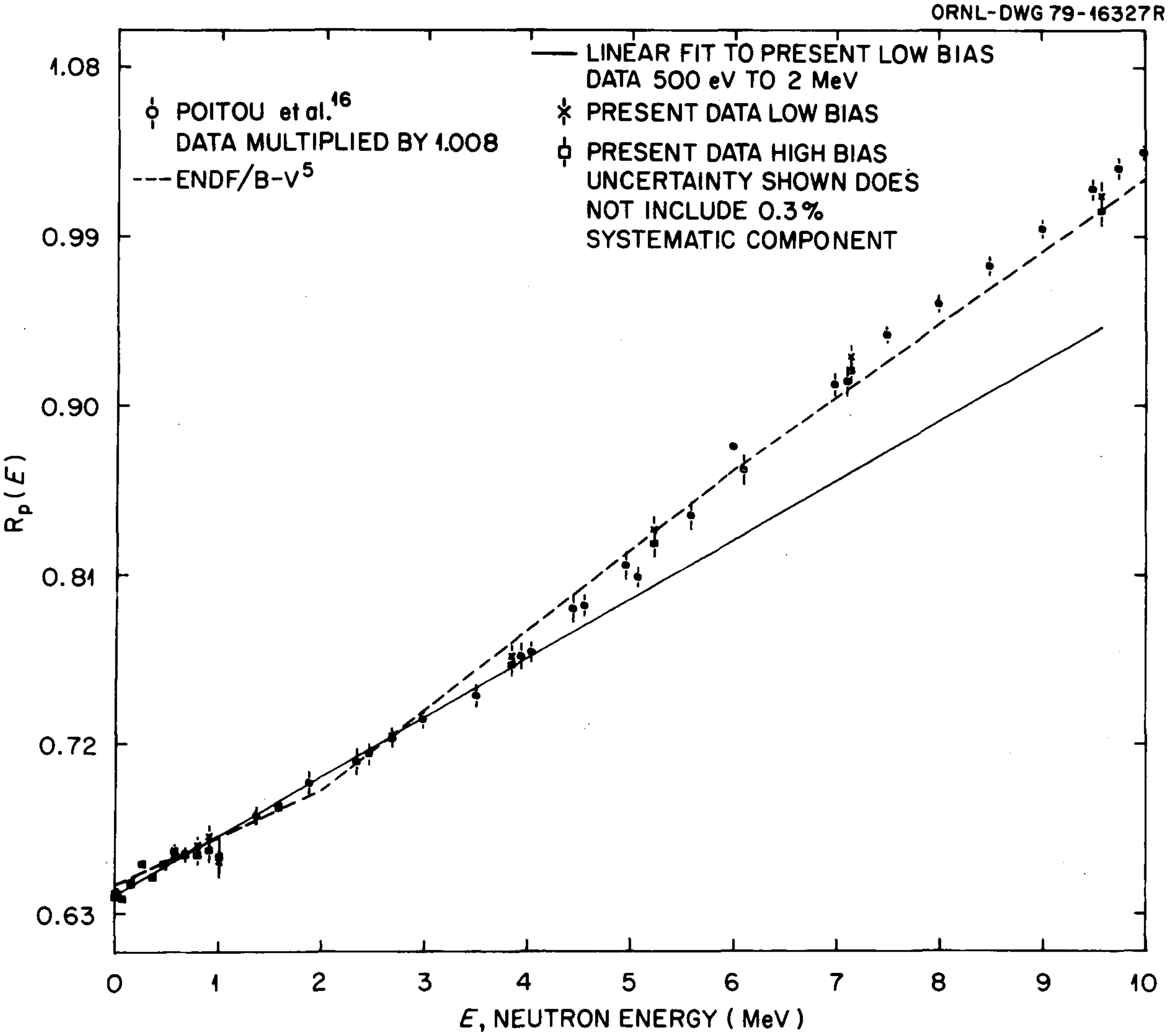

Figure 6. Experimental values of $R_p(E)$ for ^{235}U over the energy range 0 to 10 MeV. The results of ENDF/B-V are also shown.

$R_p(E)$ over the neutron energy range 500 eV to 10 MeV. Also shown in Fig. 6 are the experimental values of Poitou et al.[18] and these latter results have also been multiplied by the factor 1.008.

Values of $R_p(E)$ obtained from ENDF/B-V[5] are also shown in Figs. 5 and 6 as the dashed line. For the neutron energy region up to about 300 keV the present experimental data shown in Figs. 5 and 6 agree within their uncertainties with the values of $R_p(E)$ obtained from ENDF/B-V. For neutron energies above 700 keV, the present data differ from ENDF/B-V values by more than the experimental uncertainties around 1.5, 4, and 7 MeV. The solid straight line shown in Figs. 5 and 6 represent the results of the weighted least squares fitting of the data given in Table 1 up to 2 MeV and is given by the equation

$$R_p(E) = 0.6407 \pm 0.0008 + (0.0307 \pm 0.0021) \times E.$$

Including the thermal value for $R_p(E)$ of 0.6441 ± 0.0006 given in ref. 1 to the data of Table 1 and fitting from thermal to 2 MeV gives the relation

$$R_p(E) = 0.6427 \pm 0.0007 + (0.0282 \pm -.0024) \times E.$$

It was observed that the parameters of the linear fit varied as the energy range of the fit was changed. A linear fit to the data of Soleilhac et al.[15,17] up to 2 MeV gave the relation

$$R_p(E) = 0.6366 \pm 0.002 + (0.0301 \pm 0.0028)*E \quad ,$$

and the work of Walsh and Boldeman[15,16] including the thermal value of Boldeman[2] yields

$$R_p(E) = 0.6374 \pm 0.0008 + (0.0293 \pm 0.0015) \times E \quad .$$

At 1 MeV the value of $R_p(E)$ as given by the equation for the present data was about 1.008 larger than that given by the other two relations which is the reason for the upward normalization of the data given in ref. 15. The difference of 0.8% between the present values of $R_p(E)$ and that of Boldeman[2] is about twice the combined uncertainty of the experimental results. The uncertainties shown in Figs. 5 and 6 do not include the systematic uncertainty of 0.3% for the present work. It should be recalled that the results obtained at thermal neutron energy, as given in ref. 1, were about 0.8% larger than measured by Boldeman. The fission foil (^{235}U) used by Walsh and Boldeman[16] was the same as used in the measurements at thermal energy by Boldeman.[2] A fission chamber different from that used in ref. 1. was used in the present work, but the thickness of the ^{235}U foil coating (1 g/m^2) was the same. The ^{235}U coating thickness used in the fission chambers of Walsh and Boldeman,[16] Soleilhac et al.,[17,18] was 8 g/m^2, 7.5 g/m^2, and 7.5 g/m^2, respectively. It is suggested that the measured values of $R_p(E)$ are influenced by the thickness of the source foil and that this effect may account for the systematic difference between the present data for $R_p(E)$ for ^{235}U and that given in ref. 16.

As seen in Fig. 6 the present results for the energy dependence $R_p(E)$ for ^{235}U are consistent with that of Soleilhac et al.[18] which have been renormalized by the factor 1.008. Recall that the systematic uncertainty of 0.3% has not been included for the present results.

The linear least squares fit up to 2 MeV of the data given in Table 1 and shown as the solid line in Fig. 5 resulted in a chi-squared divided by degrees of freedom of 1.33. Three of the measured values of $R_p(E)$

deviated from the results of the fit by more than 1.5 σ, where σ is the standard deviation. For the energy intervals centered at about 0.08 0.26, and 1.0 MeV, the measured values were 1.5, 2.9, and 1.6 standard deviations respectively from the results of the fit. Boldeman et al.[16] state that the data of Walsh and Boldeman[15] and Soleilhac et al.[17] shown in Fig. 5 are inconsistent with fine structure or broad step-like structure in $\bar{\nu}_p(E)$ for ^{235}U but that there may be a slight flattening of the data between 0.25 and 0.60 MeV. No evidence of instability in the present neutron detector system was indicated by the measured values of $\bar{\nu}_p$ for ^{252}Cf in the time intervals following the neutron burst corresponding to the energy intervals of Table 1 which could account for deviations in $R_p(E)$ of more than 0.15%.

The evaluation of $\bar{\nu}_p(E)$ for ^{235}U by Manero and Konshin[19] shows a nonlinear dependence of $\bar{\nu}_p(E)$ on the neutron energy. From 0.05 MeV to about 0.3 MeV $\bar{\nu}_p(E)$ for ^{235}U from the above evaluation[19] rises more steeply than the linear representations shown in Fig. 5 and the present results correspond within the uncertainties to those of the evaluation in this energy interval. The present value of $R_p(E)$ at 0.36 MeV falls about 3 σ below that given by the evaluation of Manero and Konshin.[19]

Calculations were performed to investigate the difference in neutron production in a particular reactor spectrum using the measured values of $R_p(E)$ given in Table 1 and in using the results calculated from the fit to the those data. The neutron flux $\phi(E)$ determined[20] for ZPR-6/7 Fast Reactor Benchmark No. 12,[21] the ^{235}U fission cross section $\sigma_f(E)$ and $R_p(E)$ were used to calculate the quantitiy

$$A(R_p) = \sum_I \phi(I)\sigma_f(I)\, R_p(I) \quad ,$$

where the summation is over the energy groups of the reactor calculation ; $R_p(I)$ was set to 0.0 for groups having neutron energies larger than measured in the present work. In one case $A(R_p)$ was determined using the present experimental values of $R_p(E)$ (Table 1) and in another case values of $R_p(E)$ for neutron energies below 2 MeV were derived from the fit to the present data and experimental values used above 2 MeV. There was a 0.02% difference in the value of $A(R_p)$ calculated for the two cases cited above and the uncertainty on $A(R_p)$ due to the statistical uncertainties on $R_p(E)$ was 0.1%.

Measurements of $R_p(E)$ were also made in the present work using a ^{235}U fission chamber having a coating thickness of 20 g/m^2. The value of $R_p(E)$ using these foils (20 g/m^2) was about 3% less than observed using the thin (1 g/m^2) foils. Using Cramer's[12] results, as described earlier for the efficiency of the present neutron detector as a function of the angle neutrons enter the scintillator and assuming that the fission neutrons travel in the direction of the fission fragments a reduction of about 2% in $\bar{\nu}_p$ is calculated for thick (20 g/m^2) foils. Considerations concerning the selective discrimination against particular fission fragments was not made in the above application of Cramer's[12] calculations.

Figures 5 and 6 show two values obtained in the present work, one for the low bias (~900 keV) setting on the neutron detector and the other for a high (2.8 MeV) bias on the neutron detector. The high- and low-bias data were recorded simultaneously and the <u>difference</u> between the two values of $R_p(E)$ due to counting statistics is no greater than 0.1% A high bias was used on the neutron detector to minimize, or eliminate, the correction to $R_p(E)$ for delayed gammas following fission.[2,15] For the neutron energy region below about 400 keV the present data (counting gate starting 2.0 s after fission) for the measurements with the high and low biases are consistent with Boldeman's[2,15] interpretation of delayed gammas following fission of ^{252}Cf and ^{235}U as reflected in $R_p(E)$.

The present experiments using the high bias were not understood sufficiently to use directly, therefore the data obtained with the low bias were used and correction of those data for the effect of delayed gamma rays was made using the decay schemes suggested by Boldeman.[2]

In Fig. 6 it is noted that there are significant differences (>0.1%) in the results obtained for $R_p(E)$ using the low and high bias in the energy region around 700 keV and above 5 MeV. In the energy region above 5 MeV the difference between low- and high-bias values for $R_p(E)$ reaches 1%. This observed difference between low- and high-bias values of $R_p(E)$ in the present work may be due to an increasing number of delayed gamma rays from fission, or to electronic problems, such as lack of stability. One check on the relative stability of the neutron detector for the low and high biases is the ratio RHL of the observed neutron counts following ^{252}Cf fission for the high bias to those observed for the low bias. The maximum difference observed in RHL (^{252}Cf) for the time intervals after the neutron burst which correspond to the energy intervals of the analysis and shown in Table 1 was 0.15% from the average value for all intervals. It is suggested that anisotropy of fission fragment angular distributions produces the divergence of the values of $R_p(E)$ as measured in the present work using the high and low biases on the neutron detector. As stated previously the probability for detecting ^{252}Cf fission neutrons decreases more rapidly for the high-bias results than for the low-bias results when the ^{252}Cf source is displaced from the center of the large liquid scintillator. Displacement of the fission source along the through tube results in changing the spatial distribution of the neutron flux in the large liquid scintillator as does a change in the anisotropy of the fission fragments. It may be that experimental evidence of physical structure in $\overline{\nu}_p(E)$ for ^{235}U is due in part to incomplete correction of experimental data for anisotropy of the fission fragments. The same changes in the fissioning ^{236}U nucleus that lead to increasing anisotropy of the fission fragments may introduce other factors which influence the measured values of $\overline{\nu}_p(E)$, for example changes in the fission neutron energy spectrum. A detailed correction of experimental data on $R_p(E)$ for anisotropy may require an experimental calibration of the neutron detection efficiency as a function of the position the neutron was absorbed in the scintillator.

V. CONCLUSIONS

The main point derived from these experiments is that the neutron energy dependence of $R_p(E)$ for ^{235}U as measured in the present work is consistent with that of Boldeman et al.[15] up to 2 MeV. The normalization difference of 0.8 ± 0.4% is about the same as observed for the measurements in the thermal energy region (ref. 1). Experiments using coatings of ^{235}U of 20 g/m^2 for fission foils have shown a reduction (~3%) in $R_p(E)$ compared to that observed using foils with deposits of 1 g/m^2.

The results for $R_p(E)$ using the high bias on the neutron detector were obtained to check Boldeman's[2,15] suggestion that delayed gamma's following fission perturb the liquid scintillator method of measuring $\overline{\nu}_p$ and the results of the present experiments in the energy region below

400 keV are consistent with Boldeman's[2,15] suggestion. Above 500 keV the divergence of the results for $R_p(E)$ using the two biases has not been satisfactorily explained. It has been tentatively suggested that changes in the angular distribution of the fission fragments, and thus the fission neutrons, may explain this observed discrepancy.

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J. G. Craven wrote the computer programs used for data acquisition and assisted in formulating the data analysis procedures. Extensive use was made of a weighted least squares analysis program written by R. W. Peelle. R. Q. Wright and M. Westfall provided nuclear data and results of calculations used in the present work.

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L. W. Weston participated in design of the present experiments and has contributed to the continuing analysis of the experiments.

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