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Abstract

The energy dependence of the average number of prompt fission neutrons emitted per fission, $\bar{\nu}_p(E)$, has been measured for ^{233}U relative to $\bar{\nu}_p$ for ^{252}Cf over the neutron energy ranges 500 eV to 10 MeV and below 0.3 eV. A large Gd-loaded liquid scintillator was used to detect neutrons and the samples of ^{233}U and ^{252}Cf were contained in fission chambers. The present results for $\bar{\nu}_p(E)$ for ^{233}U are in accord with the experimental results of Boldeman and the evaluated results of Lemmel in the thermal energy range, but in the neutron energy region between 100 keV and 1 MeV the present data are 1% or more larger than other experimental values.

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 ^{233}U over the neutron-energy range from 500 eV to 10 MeV. The normalization of the data was made relative to $\bar{\nu}_p$ for the spontaneous fission of ^{252}Cf; therefore the result of the present work is the ratio $\bar{R}_p(E) = \bar{\nu}_p(E)^{233}U/\bar{\nu}_p{}^{252}Cf$. Another set of measurements was made which yields an estimate of $\bar{\nu}_p$ for ^{233}U in the neutron-energy region below 0.3 eV.

The present work is a continuation of an experimental program to measure $\bar{R}_p(E)$ for ^{233}U, ^{235}U, and ^{239}Pu in the thermal neutron energy region and in the energy region of the neutron spectrum of fast breeder reactors. A large liquid scintillator is used to detect fission neutrons and a fission chamber containing the fissile isotope is used to define fission events. Values of $\bar{R}_p(E)$ obtained for ^{235}U and ^{239}Pu in the thermal-energy range and from about 500 eV to 10 MeV have been reported previously.[1,2]

The value for $\bar{R}_p(E)$ obtained in the present work for the neutron-energy region below 0.3 eV was

$$\bar{R}_p(E < 0.3\ \text{eV}) = 0.663 \pm 0.002,$$

which is in agreement with the value

$$R_p(E_{\text{thermal}}) = 0.6601 \pm 0.0015$$

obtained by Boldeman and Dalton[3] and subsequently revised.[4,5] Boldeman[4] revised the results of Boldeman and Dalton to account for more recent data on delayed gamma rays from fission and on fission-neutron energy spectra. Boldeman and Frehaut[5] further revised those results[3] to account for the effect of foil thickness on $\bar{R}_p(E)$ measurements. Because of a lack of data on ^{233}U, Boldeman[4] based the parameters for delayed gamma rays from ^{233}U fission on the evaluated parameters for ^{235}U. Measurements in the present work of the time distribution of events following ^{233}U and ^{252}Cf fission suggest significantly more (factor of 5) events following ^{233}U fission than ^{252}Cf fission for a delayed gamma-ray emitter having a half life of ~0.6 μs. This will be discussed in a section giving the effect on $\bar{\nu}_p$ of delayed gamma rays. Both the present value $\bar{R}_p(E < 0.3\ eV) = 0.663 \pm 0.002$ and the revised value $\bar{R}_p(E_{th}) = 0.6601 \pm 0.0015$ of Boldeman and Dalton are in accord with that derived in the evaluation by Lemmel;[6] $\bar{R}_p(E = 0.0253\ eV) = 0.662 \pm 0.002$. The uncertainty of 0.002 given by Lemmel is the standard deviation resulting from the least-squares analysis of nuclear data obtained using monoenergetic neutrons (2200 m/s) as well as data obtained with neutrons having a Maxwellian energy distribution. The value of $\bar{R}_p(E = 0.0253)$ given in ENDF/B-V[7] is also 0.662. In the neutron-energy region from a few keV to about 2 MeV, the present results for $\bar{R}_p(E)$ are larger by 1 or 2 percent than other existing data. For example, the present data are about 1.7% larger than the values given in ENDF/B-V[7] in the neutron energy region around 300 keV. Since fast breeder reactors have a neutron spectrum which covers the 300-keV energy region the larger values of $\bar{\nu}_p(E)$ for ^{233}U obtained in this present work may have some significance for reactor design.

The present values of $\bar{R}_p(E)$ indicate a linear dependence on neutron energy up to about 2 MeV which is contrary to the suggestion of Boldeman, et al.[8] Boldeman et al.[8] presented measurements of the energy dependence of both $\bar{\nu}_p(E)$ and the average kinetic energy of the fission fragments for

^{233}U and concluded that these measurements were mutually consistent and that a minimum in $\bar{\nu}_p(E)$ was expected at about 150 keV. (See Fig. 3.)

II. EXPERIMENTAL METHOD

The present experiments on $\bar{\nu}_p(E)$ utilized a liquid scintillator containing 0.22% by weight natural gadolinium to detect fission neutrons (with an efficiency of 79%) and a fission chamber (efficiency ~95%) to define fission events. R. R. Spencer et al.[9] used the same large liquid scintillator in their measurement of the absolute value of $\bar{\nu}_p$ for ^{252}Cf. A description of the basic experimental techniques and some corrections made on the data are given by Hopkins and Diven[10] and by Mather et al.[11] 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 84.2 m. The liquid scintillator had a volume of about 0.910 m^3 and the diameter of the through tube was 8.9 cm.

The fission chamber was located at the center of the liquid scintillator in a beam tube which traverses the neutron detector. Neutrons from a pulsed source (19 - 30 ns in duration) produced by the Oak Ridge Electron Linear Accelerator, ORELA, were collimated to a 5.1-cm diameter beam to impinge 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 Cd liner 0.8-mm thick was inserted into the through tube and the fission chamber was placed inside this Cd liner. The purpose of the Cd liner was to reduce the number of low-energy neutrons passing from the scintillator into the through tube; the gamma-ray cascade resulting from neutron capture in Cd is ~9 MeV, which compares with 8 MeV for neutron capture in Gd.

A bias equivalent to a pulse height of about 0.9-MeV gamma-ray energy was applied to the neutron detector. Pulses above this bias were used to define coincidence with pulses from the fission chamber and to register neutron events in the interval following fission. A graph of the pulse-height spectrum resulting from neutron absorption in the large liquid scintillator is given in Fig. 2 of Ref. 2.

Both the ^{233}U sample and the ^{252}Cf source were contained in the same fission chamber. A total of 40 mg of ^{233}U was deposited on one side of 18 aluminum foils to a diameter of 5.1 cm and had an average coating thickness of 1.1 g/m^2 with the variation in thickness from center to edge being 5%. Two coated plates separated by an uncoated plate, the anode, made one section of the chamber, with the plate spacing being 0.32 cm. The chamber was filled to a pressure of 76-cm Hg with 90% argon and 10% methane. An applied potential of 60 V on two plates gave the smallest spread in the collection time and was used for all sections. The U sample was 99.97% ^{233}U, 0.02% ^{235}U, 0.001% ^{236}U, and 0.009% ^{238}U. Ten amplifier and discriminator systems were used, one for the ^{252}Cf section and 9 for the ^{233}U sections of the chamber. The division of the fission chamber into separately instrumented sections was made to reduce the pile-up of pulses caused by alpha particle emission of the ^{233}U source material. The pulse-height response of the ^{252}Cf section was monitored continuously during the experiments. In addition the pulse-height spectrum from one of the ^{233}U sections was also recorded. During the experiment the pulse-height spectrum of each section was measured at least one time.

Figure 1 shows fission fragment pulse-height distributions for the ^{252}Cf section and a ^{233}U section (No. 6) of the fission chamber. Rush[12] current amplifiers were used for all 10 sections.

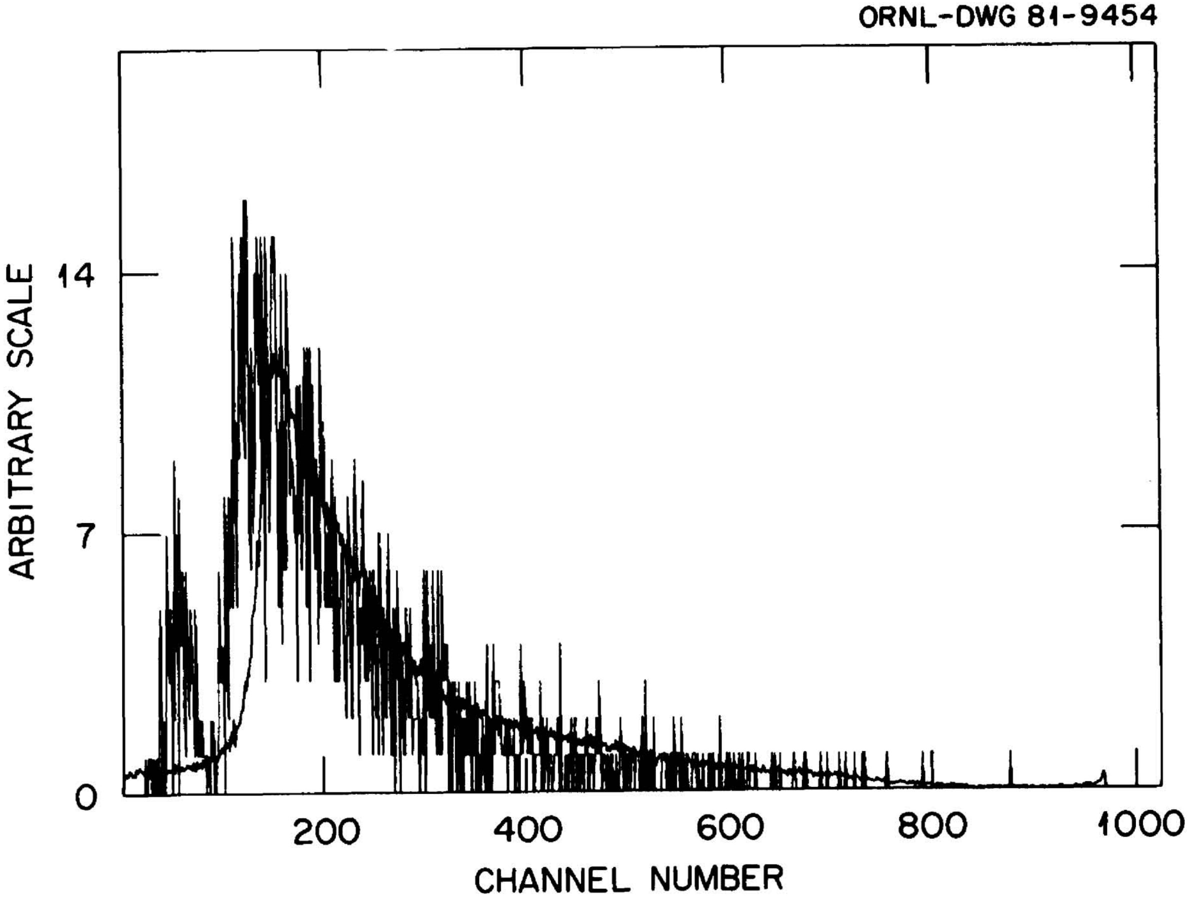

Fig. 1. Pulse-height distribution of fission fragments from ^{233}U and ^{252}Cf fission. The data with the parallel peak 35 are for ^{233}U. The bias applied to the ^{233}U fission section responsible for the small peak in the ^{233}U data.

Gated integrators were used on the fast output pulse of these amplifiers to produce a pulse suitable for pulse-height analysis. A coincidence between a fission chamber pulse and a pulse produced by prompt fission gamma rays from the large liquid scintillator was required for the pulse-height measurement. The bias on the ^{252}Cf section was set at one-half the pulse height of the peak of the fast current pulse distribution. Because of the alpha particle activity of the ^{233}U the electronics bias was set higher than for ^{252}Cf as shown in Fig. 1. The alpha particle count rate increases as the bias is decreased and the small peak seen in Fig. 1 for the ^{233}U pulse-height distribution results from a coincidence of alpha particle pulses with pulses from the large liquid scintillator.

Fission neutrons are moderated in the scintillator, diffuse, and are absorbed primarily in the Gd with the neutron absorption rate in the scintillator increasing after fission, reaching a peak at about 8 μs, and then decreasing exponentially such that 90% of the neutrons are absorbed in the 40 μs counting gate used in the experiments. Two gate starting times were used, 0.75 and 2.4 μs after fission, and were selected on the basis of Boldeman's[4] analysis of the delayed gamma rays following fission, the measured time dependence of pulses following fission and measurements of $\overline{R}_p(E)$ for different starting times of the neutron counting gate (see Ref. 1). Data were recorded simultaneously for the two gates used in the present experiments. These two gates will be referred to as delayed gates of 0.75 and 2.4 μs. A fixed non-extending dead time of 0.12 μs on the neutron detector system sets the minimum possible delay between fission and the starting of the neutron counting gate.

Data obtained for gates during which a fission pulse occurred were stored separately from the other data. In addition an extending dead time of 100 μs was applied to the gate generator when a fission pulse occurred.

Measurements of the time distributions after fission of pulses from the neutron detector showed a decrease in the count rate for times less than about 2 μs. This observation is consistent with Boldeman's[4] suggestion that delayed gamma rays from fission are detected in large liquid scintillator measurements of $\bar{\nu}_p$. In Boldeman's 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. No difference was observed in the time distribution of pulses following fission by ^{233}U or ^{252}Cf except in the time interval below ~2 μs.

Two methods were used to estimate the background recorded from all sources except that due to delayed gamma rays from fission in the foreground counting gates. In one technique a random pulse generated by a radioactive source, isolated from the neutron detector, started the counting gate to measure the background to be used for the data obtained for ^{252}Cf. In the second method a neutron detector positioned in the neutron beam was used to generate a counting gate to use for the data obtained for ^{233}U. Neutron bursts produced by the ORELA may be expected to vary from pulse-to-pulse. Part of the background in the experiments is due to 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 in a burst, a neutron detector was used to generate a gate for background analysis of the ^{233}U data. A neutron detector consisting of a pulse ionization chamber filled with argon (80%) and BF_3 (20%) was placed 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 0.5 counts per 40 μs. In the neutron-energy region above 1 MeV, where the background was 0.5 counts per 40 μs, the corresponding number of neutrons detected from ^{233}U fission was in excess of 2.5 counts in the 40 μs counting gate.

III. DATA ANALYSIS AND UNCERTAINTIES

The data were recorded using 800 time-of-flight channels with 20 channels allocated to recording the number of events occurring in the counting gate initiated by a fission event or a random source. In the analysis the data were reduced to 20 contiguous energy intervals where the average value of $\overline{R}_p(E)$ in an interval was obtained by weighting each $\overline{R}_p(E)$ for a given channel with the energy width of that time channel and dividing by the width of the energy interval.

Figure 2 shows the time distribution of events in the large liquid scintillator following fission. These time distributions were then corrected for pile-up as described later. For the ^{233}U a random coincidence of alpha particle pulses with pulses from the large liquid scintillator produces "false fissions." The background due to the false fissions was removed as described later. In the present work the ^{233}U events were contaminated with ^{252}Cf fissions which resulted in 0.002 fissions/s, which were accounted for in the analysis of the data.

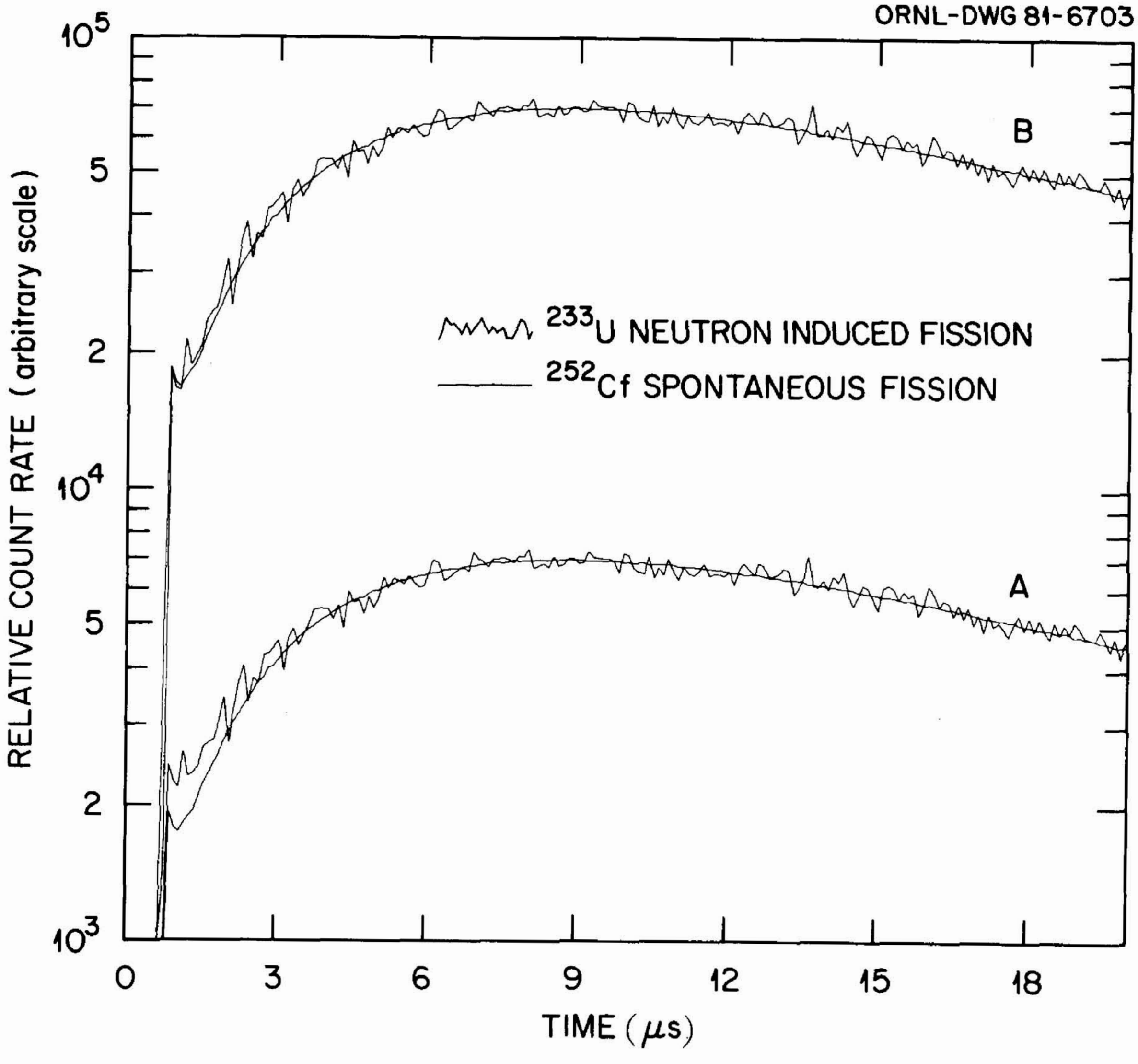

Fig. 2. Distribution of pulses from the large liquid scintillator following fission of ^{233}U and ^{252}Cf. In curves A correction was made for events resulting from delayed gamma rays using the data on delayed gamma rays compiled by Boldeman.[4] In curves B the yield of the parent isotope with the 0.62 μs half-life was increased by a factor of about 8 for ^{233}U fission. The Y axis normalization is arbitrary.

Uncertainty in Using a ^{252}Cf Standard Sample

A ^{252}Cf source distributed over a sector of a circle to simulate the radial distribution of the fissile isotope on a circular aluminum plate formed one section in the fission chamber. The observed number of counts in the large liquid scintillator following a detected fission from this source was the same within 0.03% as that detected from the ^{252}Cf source used by R. R. Spencer et al.[9] in an absolute measurement of $\bar{\nu}_p$ for ^{252}Cf. Only one ^{252}Cf source out of 5 used in the present experimental program[1,2] has shown a significant difference in the number of counts detected following fission and that source was the one chosen as the primary standard in Refs. 1 and 2. That primary standard showed about 0.25% fewer counts (neutrons) per fission than the other sources for the experimental condition of Ref. 1.

The observed value of $\bar{\nu}_p$ for a given ^{252}Cf source material may depend upon the manner in which the source is used. Variations in the observed $\bar{\nu}_p$ may be expected if the sources are not in the same position in the through tube of the neutron detector. Some variation in $\bar{\nu}_p$ may also be expected if the ^{252}Cf deposits on the fission foils differ from one chamber to the next.

An uncertainty of 0.07% has been assigned to the present work for the uncertainty in the use of a ^{252}Cf standard neutron source. This uncertainty applies to all the energy intervals. It is emphasized that the ^{252}Cf source uncertainty of 0.25% used in Refs. 1 and 2 should be retained pending final publication of the results of those works. Although it has not been definitely established, the experimental evidence suggests some fault in the primary ^{252}Cf source of Refs. 1 and 2.

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

A correction of -0.05% was made to the measured value of $\overline{R}_p(E)$ to account for the distribution of the fission samples about the center of the detector. The variation in the observed counts following fission of ^{252}Cf was measured as a function of the position of the sample about the center of the through tube and was found to be 0.015% per cm and this information was used to correct the measured $\overline{R}_p(E)$. A ±0.05% uncertainty has been assigned to this correction and this uncertainty applies to all energy intervals.

Corrections for False Fissions

A false fission is defined as the random coincidence of a pulse from the fission chamber with a pulse from the large liquid scintillator. Since a low-electronic bias was desired for operation of the fission chamber in order not to eliminate many fission fragment pulses, some pulses generated by alpha decay of the source material were detected. In order to estimate the number of false fissions as a function of time after the neutron burst, random pulses, "mock alpha," were introduced into the electronic system in the same manner as pulses from the fission chamber. The time dependence relative to the neutron burst of the coincidence between these "mock alpha" pulses and pulses from the neutron detector was recorded enabling a direct measurement of the fraction of the events recorded as fissions which were false fissions. The fraction of the events which are false fissions varies inversely with the neutron flux. For one experiment the false fissions comprised 0.6% of the recorded fission events near 500 eV and about 0.1% at 1 MeV and the uncertainty in associating the ^{233}U alpha particle rate with the "mock alpha" pulses was 0.02%. In principle the correction for false fissions does not introduce correlated uncertainties.

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 and a fixed nonextending deadtime of 0.12 μs was imposed upon the neutron detector system to insure recovery of the system. Correction of the data for pulse pile up (deadtime) was made in the manner given by Ribrag et al.[13] The correction factor obtained for the ratio $\overline{R}_p(E)$ for ^{233}U was about 0.35% in the low-energy region ($E \leq 100$ keV) and about 0.18% at 3.5 MeV of the present work.

Ribrag et al. assume that the background distribution measured is also present in foreground measurements, that is, counting gates generated by fission and their first step is to calculate the probability that an event produced by a neutron will pile up with a background event. The results of the first step of their analysis then yields the neutron distribution, which would be observed in the absence of background. Finally, the neutron distribution is corrected for pulse pile up. Spencer et al.[9] give a more complete discussion of the correction of data for pulse pile up.

Uncertainties in the method of making the pulse pile-up corrections lead to systematic errors which are energy dependent through the variation of $\overline{\nu}_p(E)$. It has been assumed that the relative uncertainty will be zero at the neutron energy, about 10.5 MeV, where $\overline{\nu}_p(E)$ for ^{233}U equals $\overline{\nu}_p$ for

^{252}Cf. This assumption may not be true because the distribution P(N), the fraction of the fission events which emit N prompt fission neutrons, may not be the same even though $\overline{R}_p(E) = 1$. An uncertainty of 20% of the correction has been assumed for the pulse pile-up correction and this uncertainty is shown in Table I.

Effect of the Anisotropy of Fission Fragment Emission on $\overline{\nu}_p(E)$

As the incident neutron energy increases the fission fragment anisotropy increases and more of the fragments have flight paths correlated with the direction of the neutron beam. Since most of the neutrons are emitted from the fragments after fission, the neutrons will also have an anisotropic distribution. For neutron induced fission of ^{233}U, Blumberg and Leachman[14] report values of the anisotropy factor greater than unity for neutron energies above 1 MeV.

Using a fission chamber which detects all fission events, the effect of anisotropy, $A > 1$, is to decrease the observed value of $\overline{\nu}_p(E)$ since more of the neutrons will have directions which result in absorbtion in regions where the large liquid scintillator has lower detection efficiency (see Fig. 3, Ref. 2).

In addition other physical changes in the fissioning nucleus such as a change in the neutron fission spectrum may take place with the increase in anisotropy which should also be considered in the analysis of data on $\overline{\nu}_p(E)$ obtained using the large liquid scintillator. However, as will be presented later, the present detector system has little sensitivity to variations in the fission neutron spectrum.

Table I. Experimental Values of $\bar{\nu}_p(E)$ for ^{233}U Relative to $\bar{\nu}_p$ for ^{252}Cf.

			$\Delta\bar{R}_p(E)$				
E1	E2	$\bar{R}_p(E)$	a	b	c	d	e
0.5200	1.052	0.6579	0.0033	0.0031	0.0005	0.001	0.0037
1.052	5.093	0.6593	0.0019	0.0026	0.0005	0.001	0.0025
5.093	10.14	0.6652	0.0027	0.0032	0.0005	0.001	0.0032
10.14	51.12	0.6629	0.0017	0.0024	0.0005	0.001	0.0024
51.12	102.8	0.6657	0.0023	0.0023	0.0005	0.001	0.0028
102.8	207.8	0.6682	0.0019	0.0024	0.0005	0.001	0.0025
207.8	303.4	0.6700	0.0021	0.0025	0.0005	0.001	0.0027
303.4	420.6	0.6765	0.0022	0.0026	0.0005	0.001	0.0028
420.6	529.9	0.6715	0.0024	0.0030	0.0005	0.001	0.0029
529.9	621.4	0.6781	0.0032	0.0035	0.0005	0.001	0.0036
621.4	738.7	0.6849	0.0034	0.0037	0.0005	0.001	0.0038
738.7	854.4	0.6970	0.0047	0.0084	0.0005	0.001	0.0051
854.4	968.2	0.6978	0.0066	0.0084	0.0005	0.001	0.0068
968.2	1054.	0.6929	0.0091	0.0120	0.0005	0.001	0.0093
1054.	2164.	0.7067	0.0030	0.0029	0.0005	0.001	0.0035
2164.	3262.	0.7477	0.0050	0.0061	0.0004	0.0009	0.0053
3262.	4536.	0.8044	0.0067	0.0090	0.0003	0.0008	0.0069
4536.	6732.	0.8847	0.0059	0.0110	0.0001	0.0006	0.0061
6732.	9625.	1.0032	0.0064	0.0084	0.0001	0.0003	0.0067
9625.	12731.	1.14	0.01	0.012	0.0003	0.0003	0.01

a. Statistical uncertainty.
b. Standard deviation from the mean (29 values).
c. Uncertainty in the pile up correction.
d. Uncertainty due to correcting for delayed gamma rays.
e. Total estimated uncertainty.

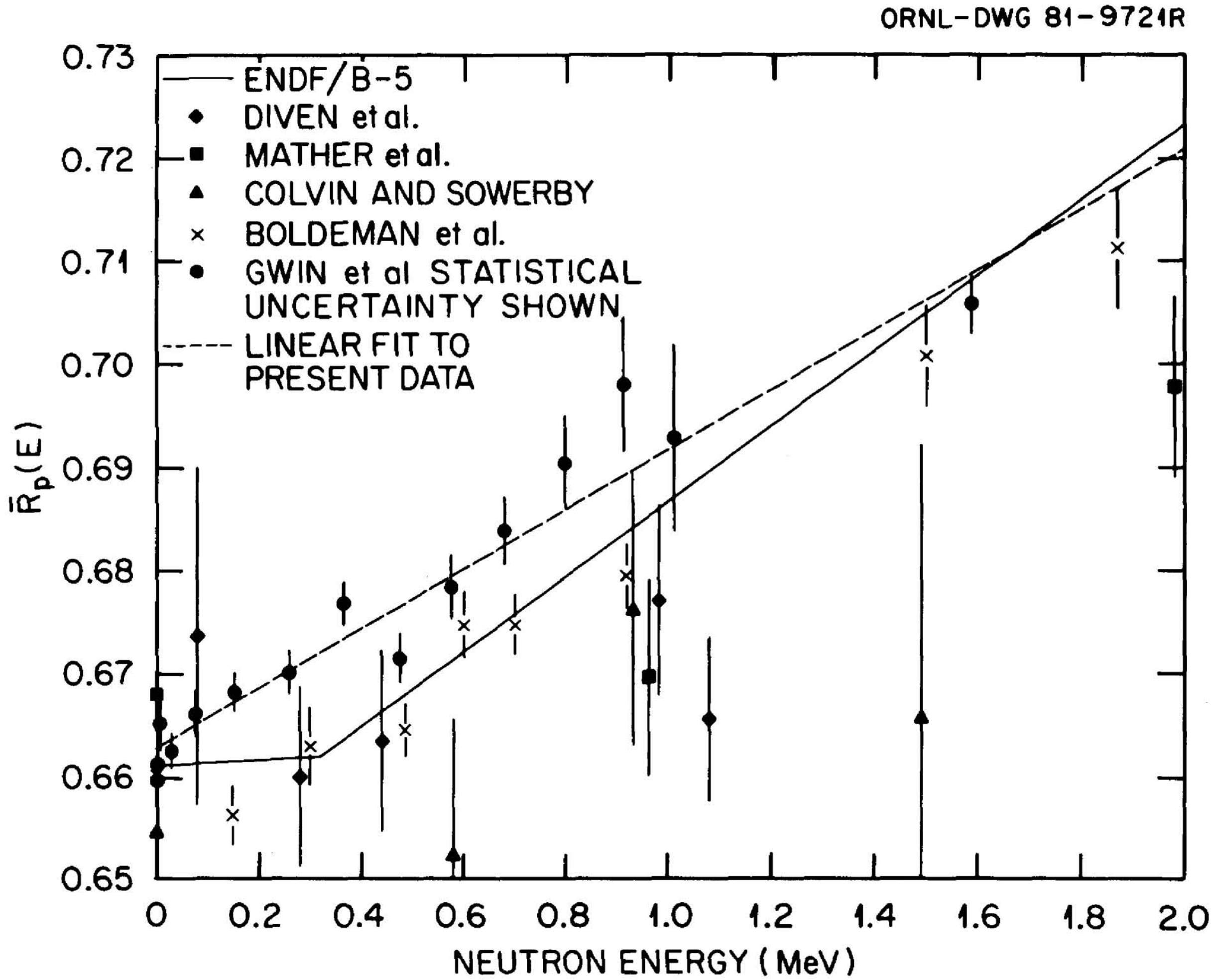

Fig. 3. $\bar{\nu}_p(E)$ of ^{233}U relative to $\bar{\nu}_p$ for ^{252}Cf over the neutron energy range 0.025 to 200 eV. $\bar{\nu}_p(E)$ values from Refs. 3, 7, 11, 16, and 17.

Assuming an anisotropy of 1.1 for the fission fragments, it has been estimated from geometric considerations that the observed $\bar{\nu}_p(E)$ would decrease by about 0.25%. At neutron energies above 6 MeV, the fragment anisotropy for ^{233}U reaches 1.3 (Ref. 17) and a detailed calculation is probably needed to make the correction to the observed $\bar{\nu}_p(E)$, but the approximate calculational method yields a decrease in the observed value of 1%. Mather et al.[11] estimated that for neutron (7.5 MeV) induced fission of ^{235}U the correction for anistropy was 0.2% at 7.5 MeV where the anisotropy of the fission fragments is about 1.3. No uncertainty has been included for the effect of anisotropy on the measured $\bar{R}_p(E)$.

Effect of Delayed Gamma Rays on $\bar{R}_p(E)$

Delayed gamma rays from fission produce pulses in the neutron detector. Boldeman [4] compiled a set of delayed gamma-ray parameters for ^{252}Cf, ^{235}U, and ^{239}Pu, which included the half life, energy of the gamma cascade, and the yield per fission of the parent isotope. In correcting values of $\bar{R}_p(E)$ for ^{233}U Boldeman[4] used the delayed gamma-ray data for both ^{233}U and ^{235}U.

In the present experiment two counting gates were used to record scintillator pulses. Gates were opened 0.75 μs and 2.4 μs after the initiating event. Data were obtained simultaneously in both gates during their overlapping period up to 40 μs. The observed values of $\bar{R}_p(E)$ using data obtained with the 2.4-μs delayed gate was about 0.25% less than that obtained using a gate delayed 0.75 μs. Using the delayed gamma-ray data suggested by Boldeman,[4] a net 0.1% reduction in the observed value of

$\overline{R}_p(E)$ would be expected for data obtained using a 2.4-μs delayed gate relative to data taken with a gate delayed 0.75 μs.

Figure 2 shows the time distribution of pulses from the neutron detector following fission of ^{233}U and ^{252}Cf. The lower curves in Fig. 1 represent the time-to-detection data corrected for delayed gamma rays using the parameters suggested by Boldeman for ^{235}U and ^{252}Cf. In the upper curves of Fig. 1, the ^{233}U data were further modified by assuming that the yield of the isotope having a 0.62-μs half-life was about eight times larger than for ^{235}U. In the detection of the gamma rays, the yield and efficiency of the detector are multiplicative factors and the factor of five given above is influenced by the uncertainty (~25%) in the detector efficiency for a particular gamma-ray cascade. This uncertainty derives from both the assumed description of the gamma-ray cascade as well as the estimation of the detector efficiency for a known gamma-ray cascade. Yields of the parent isotopes emitting the delayed gamma rays may be expected to vary with the energy of the neutron inducing fission. The data shown in Fig. 2 for ^{233}U were obtained for neutron energies extending from 500 eV to 10 MeV. Using the data shown in the lower curves of Fig. 2 and assuming that the difference in the shape for ^{233}U and the ^{252}Cf data is due to delayed gamma rays, the observed value of $\overline{R}_p(E)$ would be ~0.3% smaller for the 2.4-μs delayed gate than for the 0.75-μs delayed gate, which is in reasonable agreement with the observations. The data presented in Fig. 2 are not good enough for establishing data for delayed gamma rays, however, for ^{233}U the data do indicate a larger yield per fission of the parent isotope having the 0.62-μs half life than Boldeman and Frehaut report for ^{235}U.

Spencer et al.,[9] measured the absolute value of $\bar{\nu}_p$ for ^{252}Cf using a low, about 0.9 MeV, equivalent pulse height and a high, 3 MeV, bias on the neutron detector and obtained agreement for the two results within 0.1% after correcting the low-bias data for delayed gamma rays using the delayed gamma-ray data suggested by Boldeman and Frehaut.

In the present work, the data obtained using the gate delayed 2.4 μs after the initiating event were used to extract $\bar{R}_p(E)$ for ^{233}U. The data of Boldeman for the delayed gamma rays from ^{235}U and ^{252}Cf were used to estimate the contribution of delayed gamma rays to $\bar{R}_p(E)$ for the interval 2.4 to 40 μs following fission. A correction of -0.1% was calculated and an uncertainty of 0.15% has been assigned for the effect of delayed gamma rays on $\bar{R}_p(E)$ for ^{233}U, with no neutron energy dependence of delayed gamma rays considered.

Correction for Fission Neutron Spectrum Differences

A fission spectrum for ^{233}U neutrons was calculated using the Watt formula and the parameters given in ENDF/B-V for neutron energies from thermal to 10 MeV. These spectra were then used in a Monte Carlo code, used by Spencer et al.[9] for the ^{252}Cf $\bar{\nu}_p$ measurements, to obtain the efficiency of detecting ^{233}U fission neutrons relative to ^{252}Cf fission neutrons. Spencer et al. used a Maxwellian distribution having an average energy of 2.09 MeV for the ^{252}Cf neutrons. The correction obtained was (0.076 ± 0.004)% (statistical uncertainty) except at 5 MeV where the factor was .028%. A systematic uncertainty of 0.03% has been assigned to the correction factor for fission spectrum differences, which were applied to the data in all energy intervals.

Correction for Missed Fissions

Because of the thickness of the fissile deposit, the bias on the fission chamber electronics, and the requirement of a coincidence between a fission chamber pulse and a pulse from the large liquid scintillator, (resulting from detecting the prompt gamma rays from fission) some fissions are missed. For a discussion of missed fissions, the reader is referred to Ref. 2. For fissile deposits of 1 g/m^2, 95% or more of the fission fragments are detected. Dickens et al.[15] measured the efficiency of a fission chamber having a coating thickness of 1.4 g/m^2 of Pu to be 98.5 ± 0.38%. Missed fissions result from fragments which do not escape the deposit and from fragments which escape the deposit but do not produce a detected pulse. Most of the fragments which do not escape the deposit have angles with respect to the through tube which are large. Since fission neutron directions are correlated with the laboratory direction of the fragments, it is to be expected that these missed fissions tend to decrease (Ref. 2) the measured value of $\overline{\nu}_p(E)$ due to geometric considerations. In addition, an electronic bias may result in the preferential selection of particular fission modes thereby changing the measured value of $\overline{\nu}_p$. Boldeman and Frehaut[5] have measured the variation in $\overline{\nu}_p$ for ^{252}Cf covered with different thicknesses of lead. For their particular scintillator they estimate that for a 1 g/m^2 deposit of ^{233}U the measured value of $\overline{\nu}_p(E)$ would be decreased by 0.03%. Since some of the experimental variation in $\overline{\nu}_p(E)$ with deposit thickness may depend upon the kinetic energy of the fragments and some upon the geometry of the detector system (see Ref. 2) a model seems needed to apply the results obtained by Boldeman and Frehaut on foil thickness effects to other experimental systems.

No correction to the present data has been made to account for missed fissions; however, an uncertainty of 0.05% at all energy intervals has been assigned for this effect.

Presentation of the Data

Table I shows the results obtained in the present work for energy intervals extending from 500 eV to 10 MeV. The values of $\bar{\nu}_p(E)$ shown in Table I were obtained using the 2.4-μs delayed gate. Also given in Table I are the statistical uncertainty, the standard deviation of the mean (29 separate runs), the uncertainty in the pulse pileup correction, the uncertainty in the correction for delayed gamma rays, and the total estimated uncertainty. The value of chi-square divided by the degrees of freedom was 1.05 for the data of Table I.

Figure 3 shows a plot of $\bar{\nu}_p(E)$ for the energy range 0 to 2 MeV. Included in Fig. 3 with the present results are the data of Diven et al.,[16] Colvin and Sowerby,[17] and Boldeman et al.[4,8] along with the values given in ENDF/B-V. The present data (Fig. 3) show an approximately linear dependence of $\bar{\nu}_p(E)$ on the neutron energy up to about 1 MeV, whereas the other data indicate a nearly constant or decreasing value up to about 0.5 MeV. The dashed line in Fig. 3 represents a weighted linear fit to the present data over the energy range 0 to 2 MeV. The relation derived was

$$\bar{\nu}_p(E) = 0.6621 \pm 0.0010 + (0.030 \pm 0.002) \times E$$

where E is in MeV. Chi-square obtained in the fit was 21 for 13 degrees of freedom. The statistical uncertainties and the energy dependent uncertainties were used in weighting for the least squares analysis. Folding the systematic uncertainty with that of the intercept of the equation above yields

$$\overline{R}_p(0) = 0.6621 \pm 0.0013 \quad .$$

The above value of $\overline{R}_p(0)$ and the indicated uncertainty are the results of the analysis of the data for $E \geq 0.5$ keV assuming a linear dependence of $\overline{R}_p(E)$ on the neutron energy and is not the result of a direct measurement.

A measurement of $\overline{R}_p$ for ^{233}U was made for the energy interval below 0.3 eV and a value

$$\overline{R}_p(E < 0.3 \text{ eV}) = 0.663 \pm 0.002$$

was derived. In the measurement of $\overline{R}_p(E < 0.3 \text{ eV})$, the neutron burst rate was 800/s and the neutron flight path was 84.2 m. Two separate experiments were performed. In one, no overlap filter was used and the neutron spectrum incident on the ^{233}U sample extended from neutron energies less than 0.01 eV to greater than 10 MeV. In the second experiment a Cd overlap filter was placed in the neutron beam and neutrons with energies less than about 0.3 eV were absorbed by the Cd. The results of the second experiment were used to provide a measure of the fissions in the first experiment which were due to neutrons having energies above 0.3 eV. About 46% of the fissions in the experiment not using the Cd overlap filter were due to neutrons having energies above the Cd cut off energy. Lemmel[6] obtained a value of 0.662 ± 0.002 for $\overline{R}_p(E = 0.0253 \text{ eV})$ in his evaluation of thermal neutron parameters. Lemmel[6] noted that $\bar{\nu}_p$ at 2200 m/s derived using 2200 m/s data (fission cross section etc) and $\bar{\nu}_p$ give a value about 1.3% less than $\bar{\nu}_p$ derived using Maxwellian data alone. The value of $\bar{R}_p(E = 0.0253 \text{ eV})$ given in ENDF/B-V[7] is 0.662.

The results for $\bar{R}_p(E \leq 0.3 \text{ eV})$ derived in the present work are in agreement with the results

$$\bar{R}_p(.025 \text{ eV}) = 0.6601 \pm 0.0015$$

of Boldeman and Dalton[3] as modified by Boldeman[4] and by Boldeman and Frehaut.[5] The difference in the present value of $\bar{R}_p(E)$ and that of Boldeman and Dalton is about 0.4% (within the added total uncertainties) for the thermal energy region. Based upon statistical uncertainties only, the present data at thermal differ from that of Boldeman and Dalton by about 1.5 standard deviations.

At 300 keV the present results for $\bar{R}_p(E)$ are 1.7% larger than those of Boldeman et al.,[8] and as can be seen in Fig. 3, the present results are consistently larger than most of the other data up to 1 MeV. Above ~1 MeV the data of Boldeman et al., and the present results for $\bar{R}_p(E)$ overlap within their total uncertainties.

The authors have no explanation for the lack of agreement of the present values of $\bar{R}_p(E)$ and the other experimental data shown in Fig. 2 between 80 keV and 1 MeV. Both the average kinetic energy $\bar{E}_k(E)$ of the fission fragments and the angular distribution of the fission fragments have been shown to vary in a nonlinear manner with neutron energy in the range from 0 eV to the MeV region. Sergachev et al.[16] and Boldeman et al.[8] observed an increase in the average kinetic energy of the ^{233}U fission fragments for the energy interval 0 to a few hundred keV. Boldeman et al.[8] point out that their measurements of the energy dependence of $\bar{E}_k(E)$ and $\bar{\nu}_p(E)$ are consistent. They state that if the mass and charge division in fission of ^{233}U + n do not change with energy, then conservation of kinetic energy requires a variation in $\bar{E}_k$ to be reflected in $\bar{\nu}_p(E)$. Boldeman et al.[8] find that both their $\bar{\nu}_p(E)$ and $\bar{E}_k(E)$ measurements indicate a minimum in $\bar{\nu}_p(E)$ at about 150 keV.

Nurpeisov et al.[15] measured the energy dependence of $\bar{\nu}_p(E)$ for ^{233}U using a multiplate ionization chamber to detect fission and ^3He counters contained in a paraffin block to detect neutrons. Although the uncertainties on $\bar{\nu}_p(E)$ reported in Ref. 15 are about 0.6% or more, the authors do report ratios $\bar{\nu}_p(E)/\bar{\nu}_p(0)$ which suggest a broad minimum in $\bar{\nu}_p(E)$ for ^{233}U between 80 and 500 keV. In the neutron energy region below about 80 keV, the present results for $\bar{R}_p(E)$ agree with the other data shown in Fig. 3 within the combined uncertainties. Between 80 keV and 1 MeV, the present results are consistently high by an amount (~1%) which is outside the <u>known</u> systematic uncertainties of the present experiment by about four standard deviations. If it is assumed that there is no significant structure in $\bar{R}_p(E)$ for ^{233}U between 1 and 2 MeV, then the present data and that of Boldeman et al.[8] agree within uncertainties for that interval.

Blumberg and Leachman,[14] Nesterov et al.,[20] Smirenkin et. al.,[21] and Bertram et al.[22] have observed an increase (greater than unity) in the anisotropy of the fission fragments as the neutron energy increases from 0 to a few hundred keV. At 300 keV, for example, the anisotropy is about

1.07 and increases to 1.1 or more at 500 keV and these changes in anisotropy can cause changes in the observed $\overline{\nu}_p(E)$ in measurements using large liquid scintillators. It is possible that a combination of energy dependent physical processes in ^{233}U fission influence the measurement of $\overline{\nu}_p(E)$ in ways which have not been considered. In Ref. 2, it was suggested that differences in $\overline{R}_p(E)$ observed for ^{235}U using a low bias (~1 MeV equivalent pulse height) and a high bias (2.3 MeV) on the neutron detection system was due to changes with neutron energy of the anisotropy of the fission fragments. For a large liquid scintillator similar to that used in the present work, the maximum calculated change in $\overline{R}_p(E)$ for a change in anisotropy from 1 to 1.1 would be 0.7% (upper limit, see Fig. 3, Ref. 2). As the anisotropy increases above unity with increasing energy the observed value of $\overline{R}_p(E)$ would be expected to decrease if laboratory neutron directions are strongly correlated with the fragment direction.

The present data $\overline{R}_p(E)$ and $\Delta\overline{R}_p(E)$ were folded with a neutron spectrum characteristic of ZPR6/7[23,24] and the neutron fission cross section to obtain an estimate of the uncertainty in the criticality constant k due to the statistical uncertainties in $\overline{R}_p(E)$. An uncertainty of 0.001 in k was obtained. Folding $\overline{R}_p(E)$ from ENDF/B-V with the same spectrum and cross section yields a value of k 0.5% lower than that obtained with the present data.

IV. CONCLUSIONS

The present measurements are in accord with the results of Boldeman et al.[3,4,5] in the thermal energy range with the difference in the two values being about 0.4%. In the neutron energy region between 100 keV and

1 MeV, the present results are 1% or greater above those of Boldeman et al.[8] and no definite explanation has been given for this difference.

In the neutron energy region up to 2 MeV, the present data are described by a linear function of the neutron energy with a value of χ^2 of 14 for 13 degrees of freedom. Although a linear relation of $\overline{R}_p(E)$ with energy may not be physically correct, the present results are described adequately by a linear function of energy up to 2 MeV.

Measurement of the time distribution of pulses from the large liquid scintillator following fission show more events per fission in the early time region ($t < 2\ \mu s$) for ^{233}U than for ^{252}Cf. The time distribution of neutron events following fission of ^{233}U can be reconciled with those following fission of ^{252}Cf if it is assumed that the delayed gammas emitted having a half-life of about 0.62 μs (Ref. 4) has a yield about a factor of 5 larger for ^{233}U than ^{252}Cf.

No explanation for the difference between the present values of $\overline{R}_p(E)$ and those of the other experimenters[8,10,11,14] has been found. In particular, the experiments of Boldeman et al.[8] are very similar to the present ones and most of the known corrections required, such as for the effect of delayed gamma rays, to derive $\overline{\nu}_p$ are of the same sign and about the same size for each experiment.

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