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PROMPT-FISSION-NEUTRON SPECTRA OF ^{233}U, ^{235}U, ^{239}Pu AND ^{240}Pu RELATIVE TO THAT OF ^{252}Cf

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PROMPT-FISSION-NEUTRON SPECTRA OF ^{233}U, ^{235}U, ^{239}Pu AND ^{240}Pu RELATIVE TO THAT OF ^{252}Cf*

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ABSTRACT

The prompt-neutron-induced-fission spectra of ^{233}U, ^{235}U, ^{239}Pu and ^{240}Pu are measured relative to the prompt-spontaneous-fission-neutron spectrum of ^{252}Cf. The fission of ^{233}U, ^{235}U, and ^{239}Pu is induced by $\approx$550 keV neutrons and that of ^{240}Pu by $\approx$850 keV neutrons. The emitted fission neutrons are observed over the energy range $\lesssim$ 0.5-10.0 MeV using time-of-flight techniques. Analysis of the measured values indicates that the average-fission-neutron energies are -123±30(^{233}U), -157±24(^{235}U), -76±29(^{239}Pu) and -46±29(^{240}Pu) keV relative to that of ^{252}Cf. The experimental results are compared with those of ENDF/B-V and a simple behavior of average-prompt-fission-neutron energies is suggested.

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I. INTRODUCTION

For approximately four decades prompt-fission-neutron spectra have been extensively investigated.[1-10] Despite this effort ambiguities in both absolute and relative spectra remain a matter of concern.[11] This has recently been reflected in a renewed experimental and theoretical interest in the problem area.[12] This interest stimulated the present measurements which had the limited objective of determining the average prompt-fission-neutron energies of ^{233}U, ^{235}U, ^{239}Pu, ^{240}Pu relative to that of ^{252}Cf to improved accuracies. The measurements were made promising by the concurrent availability of these five trans-actinide samples in high purity and good abundance.[13] The measurements were carried out over a short period of time using the same apparatus thereby avoiding complex and uncertain cross-normalization of measurements made at widely different times. Throughout, ^{252}Cf spectra were used as the basic reference, although the relative results for the other isotopes are not particularly sensitive to the exact average energy accepted for the ^{252}Cf spectrum.

II. MEASUREMENT METHOD

The fast-neutron time-of-flight technique was the basic experimental method. Inherent in the use of this method is the assumption that the prompt-fission neutrons are emitted within times of $\lesssim$ 1 ns of the primary fission event. In studies of neutron-induced fission the time sequence was determined by a pulsed-accelerator beam. In measurements of neutron emission following the spontaneous fission of ^{252}Cf the time sequence was determined by a ^{252}Cf fission-fragment detector.

An essentially weightless ^{252}Cf source was deposited upon a platinum planchete ≈0.8 cm in diameter and ≈0.12 mm thick. The source intensity was ≈5×10^7 fissions/min. The source was placed at the center of a 1 mm thick 8 cm diameter steel sphere. The interior of the sphere was painted with a white-reflecting coating and was viewed with a photomultiplier tube through a side port. The source planchete was arranged so that the active deposit faced away from the photomultiplier tube cathode. A gas mixture consisting of 10% nitrogen and 90% argon flowed through the sphere at normal atmospheric pressure. Scintillations in the gas induced by the energetic fission fragments were observed. The response time of the scintillation detection was $\lesssim$ 2 ns as determined by observation of prompt-fission gamma-rays. The pulse-height selection of signals from the scintillation detector accepted $\gtrsim$ 90% of the fission events with very little alpha-particle feed through.

The uranium and plutonium samples were solid-metal right-circular cylinders. Each cylinder was ≈2 cm long. The ^{235}U and ^{239}Pu samples were ≈2 cm in diameter and the ^{233}U and ^{240}Pu samples ≈1.5 cm in diameter. The isotopic purity of all samples was $\gtrsim$ 94% and minor isotopic contamination was neglected. Chemical impurities were negligible except for the ≈1% aluminum content of the plutonium samples. The ^{233}U and plutonium samples were contained in ≈0.1 mm thick steel cans. The ^{235}U sample was bare. During the measurements, the plutonium and uranium samples were placed within a steel shell identical to that used for the above ^{252}Cf scintillation counter so that any shell-induced spectral perturbations were identical for the californium, uranium and plutonium measurements.

The neutron source for the uranium and plutonium measurements was the $^7Li(p,n)^7Be$ reaction.[14] The source was pulsed for durations of ≈1.5 ns at a repetition rate of 1 MHz. The average incident-neutron energy for the ^{233}U, ^{235}U and ^{239}Pu measurements was ≈530 keV. For the ^{240}Pu measurements the incident energy was ≈825 keV. The incident-energy spread was ≈60 keV in all pulsed-beam cases.

The uranium or plutonium samples were placed ≈13 cm from the neutron source at the focus of the Argonne 10-angle time-of-flight system.[15] These samples were interchanged with the ^{252}Cf scintillation-counter source. Jigs and sample holders assured accurate alignments.

Ten neutron-flight paths were concurrently employed in the measurements distributed over an arc of ≈140°. The flight paths varied from ≈500 to 510 cm and were measured to within <0.3 cm. The uranium and plutonium samples were so arranged as to be cylindrically symmetric with respect to the flight paths. The ^{252}Cf scintillation-counter source was arranged so that

it could be rotated to average-out any small angular anisotropies. Ancillary measurements did not show any significant angular-dependent effects.

The neutron detectors were 12.7 cm diameter organic scintillators ≈2 cm thick. Their sensitivites varied with low-energy cut-off points ranging between 250 and 600 keV. Pulse-shape-sensitive circuitry was used to suppress the gamma-ray response of the scintillators. Detector stability was important but absolute sensitivity was not relevant as the measurements were entirely ratio determinations.

The time calibration of the measurement system was based upon the use of calibrated delay lines. The measured results were least-square fitted with a linear time relation. The results of the fitting procedure were consistent with the measured values to within $\lesssim$ 2 ns (i.e. $\lesssim$ one time channel). The calibration procedure was repeated subsequent to each ^{252}Cf measurement period with results that were consistent to $\lesssim$ 1%. The time scale was further verified by observation of ≈525 keV neutrons elastically scattered from the measurement samples. The scattered-neutron energy determined from the time measurements agreed with that determined from the setting of the pulsed accelerator to within ≈5 keV. It was concluded that uncertainties in the time scale were not governing factors in the overall experimental uncertainties particularly in view of the fact that ratios were the experimental objective.

Many aspects of the above apparatus and measurement method were very similar to those routinely employed in this Laboratory in neutron-scattering measurements as described in Refs. 15, 16 and 17.

III. EXPERIMENTAL RESULTS

The experiments were carried out in three independent sets (A, B and C), each employing ten independent flight paths and detectors. Data obtained with each detector were treated independently. All measurements included a determination of the ^{252}Cf spectrum. All neutron-induced fission samples were used in at least two sets of measurements and some in all three. Thus 20-30 independent determinations of the fission-neutron spectrum relative to that of ^{252}Cf were made for each of the neutron-induced fission samples. This high degree of redundancy of essentially independent measurements gave a good measure of true uncertainties.

Careful attention was given to fission-spectrum background determinations. The character of the background contribution to the ^{252}Cf spontaneous-fission measurements was different from that encountered in the pulsed-beam uranium and plutonium measurements. In the ^{252}Cf measurements the timing sequence was terminated by a random and high-rate fission signal. As a consequence there was a small distortion of the ^{252}Cf spectra due to accidental termination of events. The effect was not large as the background component was $\leq$ 5% of the observed fission-neutron maximums. In the pulsed-beam measurements the timing sequence was terminated by a signal, derived from the pulsed accelerator, at a fixed 1 MHz rate. However, the ^{233}U and ^{240}Pu samples were small and radioactive and in all the pulsed-beam measurements the primary neutron source contributed to the backgrounds. Thus the backgrounds in the pulsed-beam measurements were larger than those encountered in the ^{252}Cf measurements (e.g., in the worst cases 20-50% of the observed neutron maximums) but relatively constant in time. Instrument tests indicated that uncertainties in the differential or integral linearity of the measurement system did not significantly effect the background determinations or the measurements generally.

The background treatment in both of the above types of measurements was identical. Two background time intervals were selected for each spectrum. The first was a very constant region just prior to the observation of the fission (or target) gamma-ray. The second was a constant region delayed in time corresponding to neutron energies lower than the detector cut-off point. The average background in both intervals was determined and a linear interpolation made between the two corresponding average-background values. This interpolation extended in time from the fission gamma-ray through the region of observed fission neutrons. For the pulsed beam measurements the interpolated background values were essentially constant. For the ^{252}Cf measurements the interpolation sloped with time due to the above-noted accidental rate. However, the backgrounds were small in the ^{252}Cf measurements and the above linear approximation was a good representation of the true accidental distribution.

The time zero was referenced to the prompt-fission-gamma-ray peak. In doing so care was taken to assure that the identification of the gamma-ray was not skewed in time and that the prompt-target gamma-ray of the pulsed-beam measurements was not confused with gamma-rays from the actual fission. The gamma-ray peak position was sensitive to the energy response of the particular measurement system and this, in turn, was partly governed by the biases employed in the pulse-shape sensitive systems discriminating against gamma-ray emission. These various effects could influence the time-zero position

by a few ns. Such shifts are not important when dealing with modest energy neutrons but can be important in the measurement of high-energy neutrons (e.g., 10 MeV and above). The gamma-ray time resolutions of the present systems were approximately 2 ns. These values were smaller than frequently encountered in such measurements and thus small distortions of the gamma-ray position may not have been observed in some past measurements to the detriment of, particularly, the determination of the high-energy portion of the fission-neutron spectrum. In the present measurements the gamma-ray position was carefully investigated with and without active pulse-shape-sensitive systems. In addition, the target gamma-ray position was determined in the pulsed-beam measurements using a bismuth scattering sample. With these checks it was felt that the gamma-ray position was generally determined to within 2-3 ns in the present measurements. This estimate was supported by the internal consistency of essentially independent measurements carried out with both the ^{252}Cf chamber and the pulsed beam.

The neutron time scale was determined from the measured neutron flight paths and flight times in the conventional manner using a relativistic calculation. There was also a qualitative consistency between small structural artifacts in some of the measured neutron spectra and the known resonance structure due to the air contained in the 5 m flight paths.[17]

Measurement periods were approximately 48 hours for each sample of each set. The peak intensities of the observed fission-neutron spectra varied from a minimum of 1500-2000 events in the worst cases (e.g. shorter running times with ^{240}Pu) to 10,000 events in the best cases (e.g. long running times with ^{252}Cf). Illustrative ^{252}Cf and pulsed-beam induced (^{239}Pu) spectra are shown in Figs. 1 and 2. These figures illustrate: the stability of the background, the clean separation between prompt gamma-rays and the onset of the neutron events obtained with the relatively long flight paths, and the neutron-energy-sensitivity range of the detectors. The distributions have a very slight trace of structure that can be qualitatively correlated with resonance phenomena in either the steel shell surrounding the samples/source or air in the long flight paths. In these ratio measurements this structure cancels over any reasonable energy average.

Each of the time spectra was converted to an energy spectrum and the energy-dependent ratio of each sample spectrum constructed relative to that of the respective ^{252}Cf spectrum. Statistical errors were the dominant source of uncertainty and they were carried throughout the conversion-to-ratio process. The next most significant source of error had to do with the determination of time zero. Here, and for other and smaller sources of uncertainty, error was difficult to quantify thus consistency of a number of independent measurements was accepted as the best measure of true uncertainty. It was assumed that all of the measured fission spectra could be described by a simple maxwellian of the form

$$N(E) \approx \sqrt{E}\ e^{-aE}\ , \quad a \equiv 3/(2 \cdot \text{average energy}) \qquad (1)$$

An alternate and often used form is the "Watt" spectrum.[8] Its application to the present experimental interpretation would not substantively change the results but would introduce some additional complexity. Therefore, the Watt

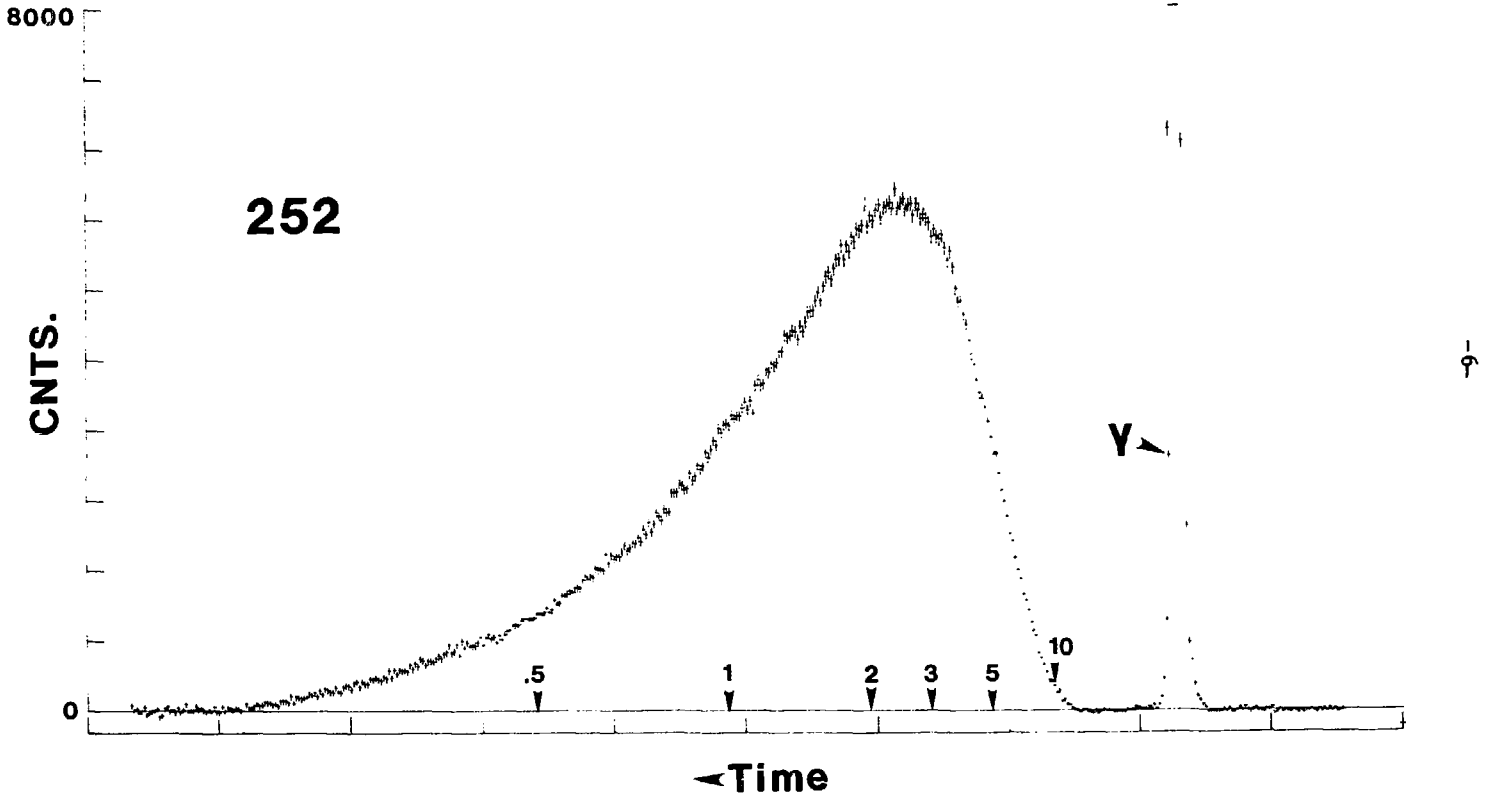

Fig. 1. Illustrative ^{252}Cf measured prompt-fission-neutron time-of-flight spectrum. Small backgrounds have been subtracted as described in the text. Prompt gamma-ray response and neutron energy scale in MeV are indicated.

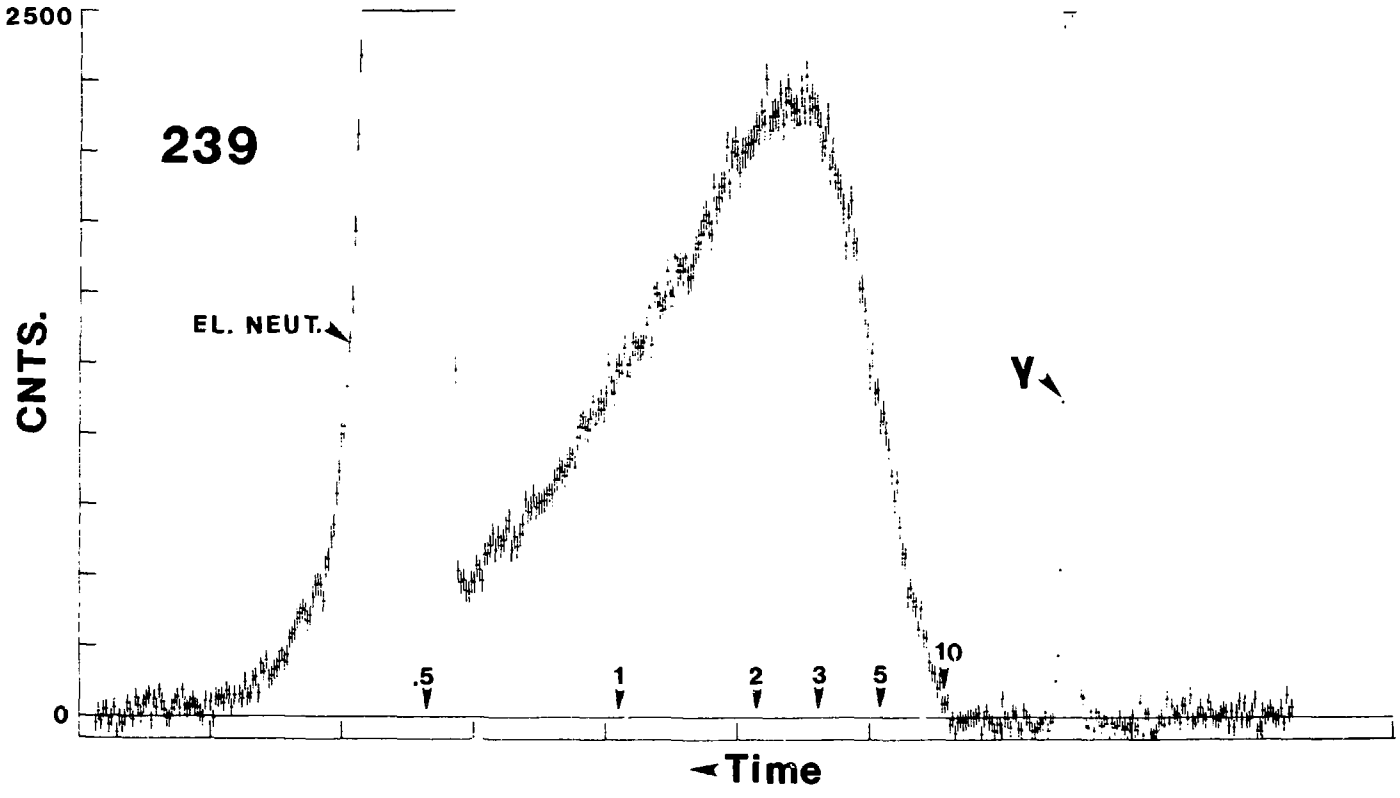

Fig. 2. Illustrative pulsed-beam (^{239}Pu) measured prompt-fission-neutron time-of-flight spectrum. The prompt gamma-ray and scattered-neutron response are noted together with an indication of the neutron energy scale in MeV.

formulation was avoided. With the assumption of Eq. 1 the spectral ratios are simply expressed as

$$\frac{N_1(E)}{N_2(E)} \equiv R(E) \propto e^{E(a_2 - a_1)} \qquad (2)$$

Of course, detector efficiencies, small fluctuation artifacts common to both factors of the ratio, etc. cancel in the formulation of Eq. 2. Clearly, the logarithms of the ratio quantities is a linear function of the energy and no significant deviation from this simple behavior could be observed in the present experiments. This suggests that the assumption of a maxwellian distribution for all of measured spectra was reasonably valid.

The logarithms of the experimental ratios were least-square fitted with the simple linear function of Eq. 2. Several such distributions and their corresponding linear fits are illustrated in Fig. 3. There were 20-30 such distributions for each ^{252}Cf/sample ratio. The slopes obtained from the fitting procedures were averaged by set (ten per set). The uncertainty in the average slope per set was defined as the RMS deviation of the individual values from the weighted mean. This was believed to be a conservative measure of uncertainty as one anomalous value can lead to a large RMS error. Indeed, the consistency of the derived ratio values was generally better than indicated by the RMS error. The energy range accepted for the evaluation of the energy-dependent slope of the ratios varied from detector to detector and from run to run but generally fell in the range 0.55-0.85 to 8.0-10.0 MeV.

The above procedures lead to (a_2-a_1) of Eq. 2 by set. Assuming a reference ^{252}Cf average-fission-neutron energy, the average-fission-neutron energy of the spectra of the other samples is defined. The Grundl and Eisenhauer[18] average ^{252}Cf fission-neutron energy of 2.13 MeV was accepted as the reference value. It is based upon the evaluation of a number of experimental results and supported by the more recent measurements of Boldeman et al.[19] With this reference energy the incremental average-fission-neutron energies for each of the samples relative to ^{252}Cf follow directly. The results are given by set and with corresponding uncertainty in Table I. These incremental values are not particularly sensitive to the exact choice of the ^{252}Cf reference energy. The incremental values from the three independent sets were averaged to obtain the weighted average incremental values shown in Table I. Concurrently, the ratio $^{239}Pu/^{235}U$ was constructed with results that were consistent with the ratios of the respective parts to ^{252}Cf.

Care was taken to use similar measurement environments for both the ^{252}Cf and the pulsed-beam measurements. However, the latter inherently involved finite-sized samples that led to multiple events not characteristic of the ^{252}Cf measurements. These multiple events did degrade the energies of the spectra observed in the pulsed-beam measurements. The magnitude of the degradation was calculated using a monte-carlo simulation of the experimental measurements and the data base of ENDF/B-IV.[2] The resulting correction factors were applied and the final results are given in Table I. The corrections were generally relatively small as the samples themselves were

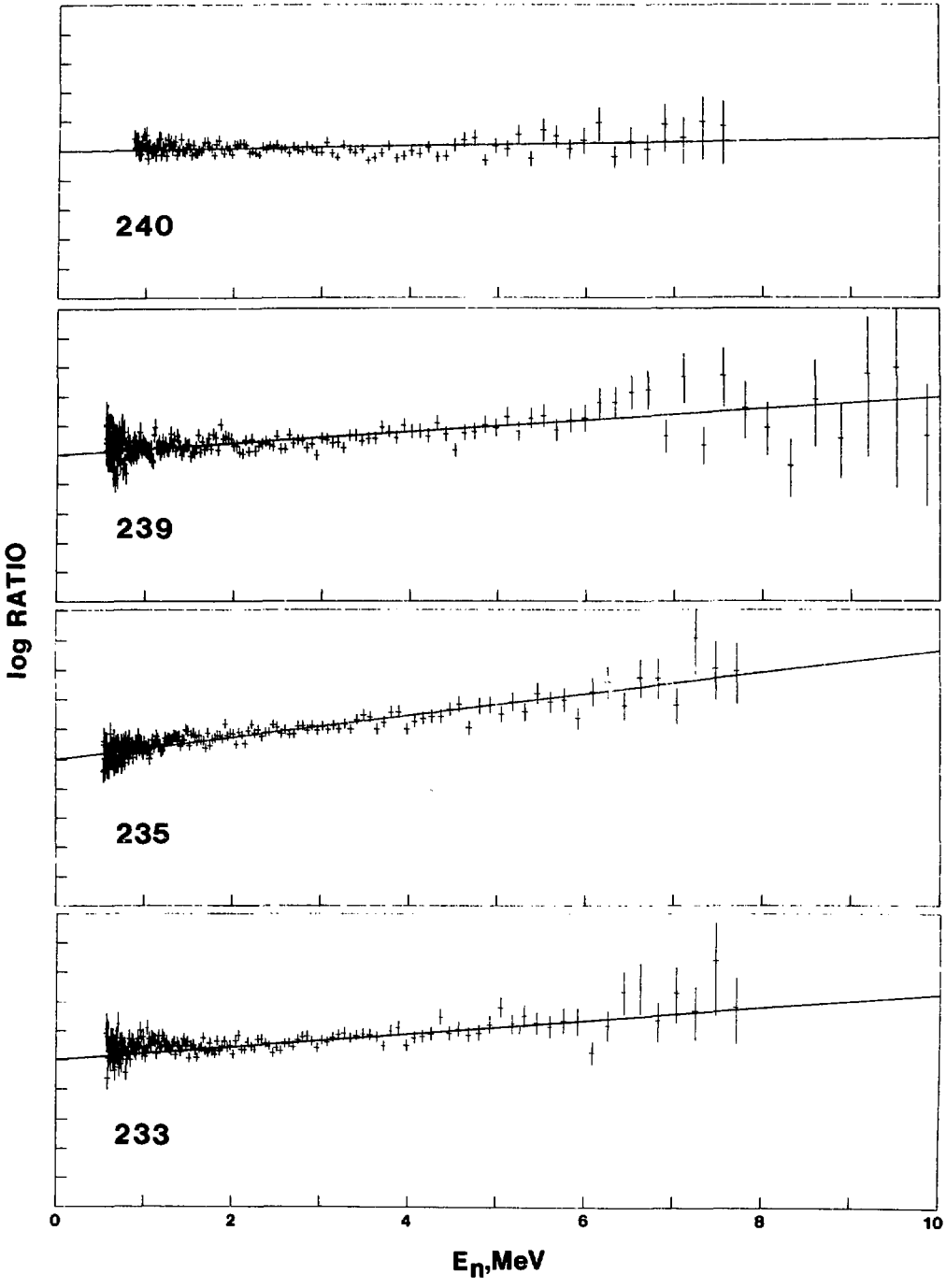

Fig. 3. Energy-dependent fission-neutron-spectra ratios relative to ^{252}Cf. Measured values are indicated by crossed data points. The result of a linear fit to the measured values is indicated by a solid curve. The respective target masses are given numerically.

TABLE I. Relative Average-prompt-fission-neutron Energies in keV

Measurement Set	Measured Quantity			
	$^{233}U/Cf$[a]	$^{235}U/Cf$[a]	$^{239}Pu/Cf$[a]	$^{240}Pu/Cf$[a]
Set A[b]	-180 ± 53	--	--	-82 ± 59
Set B[b]	-119 ± 40	-179 ± 31	-86 ± 36	-30 ± 30
Set C[b]	-119 ± 60	-175 ± 37	-111 ± 48	-101 ± 60
Weighted Average[c]	-139 ± 31	-177 ± 24	-101 ± 29	-61 ± 29
Corrected Average	-123 ± 30	-157 ± 24	-76 ± 29	-46 ± 29
ENDF/B-V[a]	-56.9	-99.2	-18.0	-56.9
ENDF/B-IV[d]	-114.0	-135.0	-33.0	-93.0

[a]Assuming $\bar{E}$ of ^{252}Cf = 2.13 MeV.

[b]Weighted averages of 10 measurements, uncertainties are the RMS deviation from weighted mean of 10 measurements.

[c]Uncertainty defined as average deviation of n sets divided by $\sqrt{n}$.

[d]At E incident of present measurements, assuming $\bar{E}$ of ^{252}Cf = 2.13 MeV.

small but the correction factors did systematically change the measured values. Perturbations due to (α,n) reactions were estimated and believed to be insignificant.

The present results for ^{235}U and ^{252}Cf are in remarkably good agreement with the corresponding evaluated quantities deduced by Grundl and Eisenhauer as a result of a review of the data base available to 1975.[18] The present $^{239}Pu/^{235}U$ values are in good agreement with results previously reported from this Laboratory[21] (and with a number of other results as given in the review of Ref. 3). The present results are also consistent with those average-fission-neutron-spectrum energies recently reported by Andreichuk, et al.[22] The relative incremental values between average fission-neutron energies of ^{233}U, ^{235}U and ^{239}Pu obtained from the present experiments are reasonably consistent with the comparable quantities given in ENDF/B-V. That is not so for the case of ^{240}Pu. ENDF/B-V employs a "Watt" representation for ^{233}U, ^{235}U and ^{239}Pu and a maxwellian representation for ^{240}Pu but the different formalisms have a very small influence on the present comparisons. If one accepts the ^{252}Cf average-fission-neutron energy of 2.13 MeV as a reference value, the present results imply average-fission-neutron energies for ^{233}U, ^{235}U and ^{239}Pu that are generally 60-70 keV lower than given by ENDF/B-V and values that are only marginally, if at all, consistent with the recent ^{235}U measurements of Bertin et al.[10] However, the agreement with the recent experimental results of Johansson et al.[1] is good.

The present results suggest a simple linear dependence of average-fission-neutron energy relative to that of ^{252}Cf on Z^2/A as illustrated in Fig. 4. Here, as elsewhere throughout this paper, the conclusions are strictly applicable to the measured energy range of $\approx$0.55-10 MeV. They are also obviously sensitive, on an absolute scale to the choice of the reference ^{252}Cf average energy accepted for normalizing the measured ratio quantities.

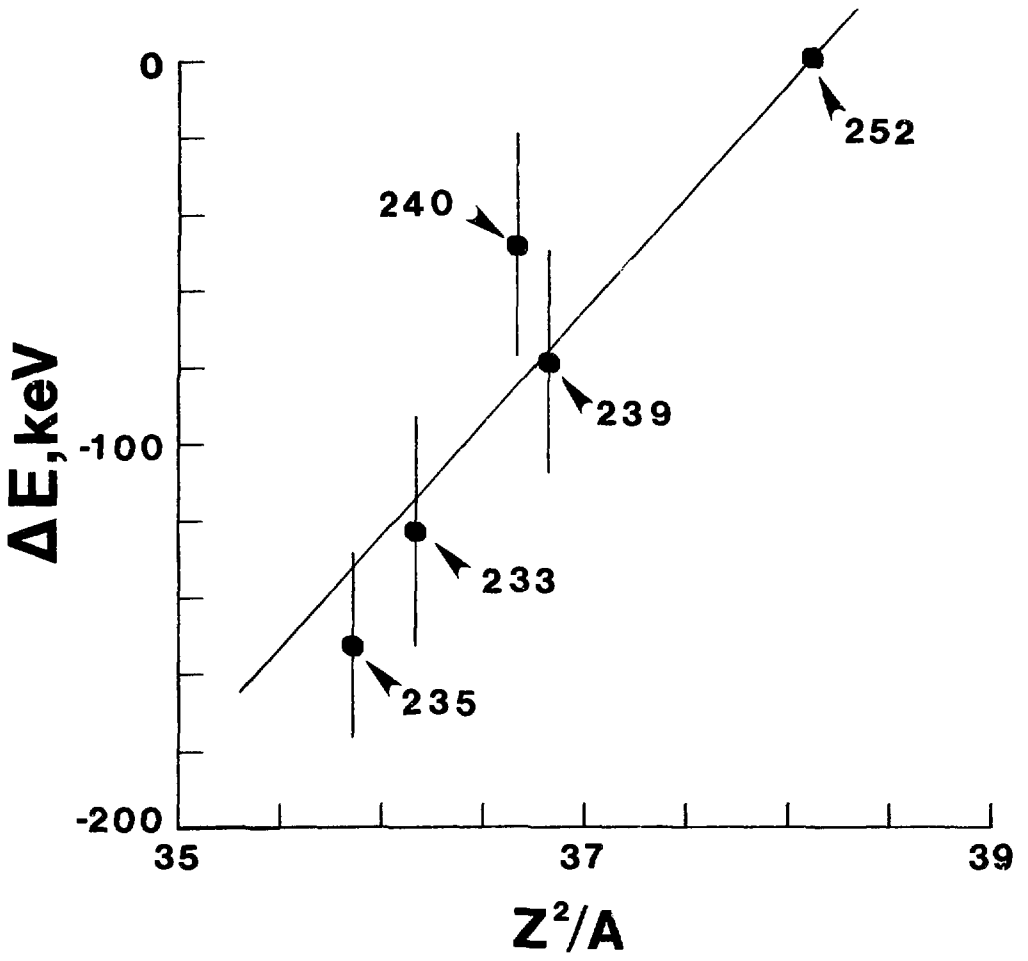

Fig. 4. Average-fission-neutron energies relative to that of ^{252}Cf as a function of Z^2/A of the target nuclide.

IV. CONCLUSIONS

The results of the present experiments suggest several conclusions (or composites thereof) as follows; 1) The incremental average-fission-neutron-energy differences as given in ENDF/B-V are generally consistent with the present experimental results. 2) The average energies of the spectra for the body of the isotopes (i.e. ^{233}U, ^{235}U and ^{239}Pu) as given in ENDF/B-V are not consistent with the present experimental results and the ^{252}Cf fission-neutron spectrum as given in Ref. 18 and as reported from recent measurements[19] (the status of the ^{240}Pu is less certain). 3) The above suggests that the ^{252}Cf spectrum as given in Ref. 18 and as recently measured[19] is too soft by approximately 65 keV or that the body of ENDF/B-V fission spectra are too hard by about the same amount. The above conclusions are not sensitive to the alternate choice of Watt[8] or maxwellian spectral representations and are expected to be as valid at thermal as at the low bombarding energies of the present experiments. They are explicitly applicable only to the measured spectrum-energy range of 0.05-10.0 MeV. However, spectrum intensities at higher energies are very weak and therefore the higher-energy region is of minor importance in many applications. The shape of the fission spectrum at very low energies (e.g., below 500 keV) remains of some controversy and strong deviations from either the Watt of maxwellian form could distort the above conclusions. More generally, isotope-dependent variations in spectral shapes could distort the present results. No such differences have been reported. None are given in ENDF/B-V and the present experimental results are consistent with a common spectral shape for all isotopes (of course, accounting for differences in the respective average energies).

The above comments are a matter of concern in some applications. This is illustrated by the sensitivity of FBR integral parameters to average-fission-neutron-spectrum energies. Calculations were carried out using one-dimensional spherical models of the fast critical assemblies ZPR-6, 6A (^{235}U-fueled) and 7 (^{239}Pu-fueled). For each assembly two sets of homogeneous cross sections with 27 broad energy groups ($\Delta u \approx 0.5$) were generated using the SDX code. The two sets for each assembly were generated with different fission-spectrum temperatures. For assembly 7 (^{239}Pu) the reference and modified fission-spectrum temperatures were 1.40 and 1.44 MeV, respectively. The fission spectrum was taken to be a maxwellian in each case. The corresponding average-fission-neutron energies were 2.1 and 2.16 MeV, respectively. For assembly 6A (^{235}U) the respective temperatures and average energies were 1.33 and 1.37 MeV (temperatures) and 1.995 and 2.055 keV (average energies). For both assemblies the average-fission-neutron energies differed by $\approx$ 60 keV. This energy increment approximately corresponds to the increase in average-fission-neutron energies of ENDF/B-V relative to version IV.

The results of the above changes in average-fission-neutron energies upon calculated eigenvalue and central reaction-rate ratios for ZPR-6 assemblies 6A and 7 are summarized in Table II. The increases in average-fission-neutron energy increase k_{eff} by 0.35% (7) and 0.24% (6A). This effect motivates a hardening of fission spectra to improve agreement between calculated and measured k_{eff} values. Increases in fission-neutron energies tend to decrease reaction-rate ratios only slightly (0.1-0.3%) excepting those involving a threshold reaction. The ratios f^{28}/f^{49} and f^{28}/f^{25} increase by $\approx$3% with the hardening of fission-neutron spectra. Again, there

is a motivation to harden the fission spectra to improve the agreement between macroscopic measurement and calculation. Whatever the exact character, the average-fission-neutron energy is a sensitive parameter in the neutronic performance of FBR systems.

^{252}Cf is the basic nu-bar standard usually determined by prompt (e.g. scintillation) or delayed (e.g. bath) detection techniques. The measurement methods can be sensitive to the average-fission-neutron energy in one way or another depending upon the method employed. Thus changes in the ^{252}Cf fission-neutron-spectrum energy could influence the general nu-bar values, and these are among the most sensitive of all fission-reactor parameters.

The present measurements raise some serious questions that should be addressed.

TABLE II. Sensitivities of ZPR-6 Assemblies to Variations In Average-fission-neutron Energies.[a]

	ZPR-6 Assembly -7 (^{239}Pu-fueled)		
	Reference Chi T = 1.40 MeV $\bar{E}$ = 2.10 MeV	Modified Chi T = 1.44 MeV $\bar{E}$ = 2.16 MeV	% Diff. (Rel. to Ref.)
Eigenvalue	0.96801	0.97140	+0.3505
Central Reaction Rate Ratios			
f^{25}/f^{49}	1.08976	1.08857	-0.1089
c^{25}/f^{49}	0.33128	0.33042	-0.2596
f^{28}/f^{49}	0.02315	0.02378	+2.7294
c^{28}/f^{49}	0.15647	0.15609	-0.2411
c^{49}/f^{49}	0.29801	0.29717	-0.2811
f^{40}/f^{49}	0.20103	0.20273	+0.8444

	ZPR-6 Assembly -6A (^{235}U-fueled)		
	Reference Chi T = 1.33 MeV $\bar{E}$ = 1.995 MeV	Modified Chi T = 1.37 MeV $\bar{E}$ = 2.055 MeV	% Diff. (Rel. to Ref.)
Eigenvalue	0.97276	0.97510	+0.2401
Central Reaction Rate Ratios			
c^{25}/f^{25}	0.29043	0.28988	-0.1898
f^{28}/f^{25}	0.02278	0.02349	+3.1351
c^{28}/f^{25}	0.14378	0.14354	-0.1660

[a]Values calculated with ENDF/B-IV nuclear data using a homogeneous spherical model as outlined in the text.

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