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VARIATION WITH NEUTRON ENERGY OF THE YIELD
OF HIGH KINETIC ENERGY FRAGMENTS
FROM ^{233}U , ^{235}U and ^{239}Pu

L. G. Miller and M. S. Moore

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FROM ^{233}U , ^{235}U and ^{239}Pu

by

L. G. Miller and M. S. Moore

IDAHO NUCLEAR CORPORATION

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L. G. Miller and M. S. Moore

Abstract

A fission chamber has been devised which detects with high efficiency those fission events in which the kinetic energy release of the heavier binary fragment is maximal. This detector has been used, in conjunction with a conventional fission detector (whose efficiency is independent of fragment energy), in a study of the relative yield of high kinetic energy fission of ^{233}U , ^{235}U and ^{239}Pu , induced by low energy neutrons. The variations in the yield of high kinetic energy fragments indicate a qualitative correlation with results of measurements of the relative symmetric fission yield as a function of neutron energy. This correlation, however, suggests that the increased relative symmetric fission is accompanied by an increased relative yield of high kinetic energy fragments, contrary to what has previously been assumed on the basis of current models of the fission process.

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Variation with Neutron Energy of the Yield
of High Kinetic Energy Fragments
from ^{233}U , ^{235}U and ^{239}Pu

L. G. Miller and M. S. Moore

I. Introduction

It was first noted by Melkonian and Mehta⁽¹⁻³⁾ that the relative yield of fragments of ^{235}U fission induced by neutrons below 100 eV appears to vary in a way which depends on the thickness of the target of fissile material. From resonance to resonance, the relative yield of fragments from thick targets (targets which are so thick that fragments from only those events near the surface can escape) differs from the yield from thin targets, from which all the fragments can escape. This behavior was interpreted by Melkonian and Mehta as evidence that the kinetic energy of ^{235}U fission fragments varies from resonance to resonance.

In a previous experiment^(4,5), the present authors observed a variation of the yield of long-range heavy fragments for the fission of ^{235}U by neutrons of low energy (< 1 eV). The analysis of these early results appeared to show that there are channel effects in the total kinetic energy of fragments of fission induced by low energy neutrons.

A variation with neutron energy of the ^{235}U fission product mass distribution has also been noted. Cowan et al.^(6,7) observed a variation in the relative yield of symmetric and asymmetric ^{235}U fission products, over the neutron energy region of 8 to 60 eV, by radiochemical techniques. Faler and Tromp⁽⁸⁾ observed similar variations below approximately 1 eV. The radiochemical determinations of the

relative symmetric fission yield of ^{233}U , ^{239}Pu and ^{241}Pu as a function of neutron energy, were measured several years ago by Régier et al. (9,10).

II. Experimental Arrangement

The Materials Testing Reactor (MTR) crystal spectrometer was used as a source of monochromatic neutrons whose energy was varied from 0.02 to 10 eV. Primary emphasis was given to the energy region below 2 eV, to permit a comparison with the mass distribution data for the three isotopes under consideration (8-10).

The experiment consisted of the simultaneous measurement of the following quantities as a function of incident neutron energy: (1) the fission fragment yield with a detector whose efficiency depends on the fragment total kinetic energy, which will be designated as the TKE detector, (2) the fission fragment yield as measured with a conventional detector, i.e., one whose efficiency is independent of the fragment kinetic energy, and (3) the neutron beam flux with a detector whose efficiency varies inversely as the neutron velocity. The fission fragment detectors were argon gas scintillation counters, in the initial stages of the work (for ^{235}U and ^{239}Pu) using RCA 6342A photomultiplier tubes with p-quaterphenyl wavelength shifter, and in the later stages (for ^{235}U and ^{233}U) 56 UVP tubes without a wavelength shifter. The neutron beam monitor consisted of a single one-inch diameter proportional tube, containing BF_3 gas, at 67 cm pressure, depleted to 11% ^{10}B .

A pictorial view of the fission chamber is shown in Fig. 1. The detector whose efficiency is dependent upon fragment energy (TKE detector) consisted of a layer of fissile material approximately

600 $\mu\text{g}/\text{cm}^2$ thick, sandwiched between two copper or nickel foils of thickness 0.0001 inch. Each of the two absorber foils was thick enough to stop about 80% of the heavier of the two fragments emitted isotropically from the layer of fissile material. Coincidence was required between the two fission fragments, and the number of coincidences per unit time was recorded as a function of neutron energy. The detector whose efficiency was independent of fragment energy (conventional detector) consisted of a layer of fissile material approximately 200 $\mu\text{g}/\text{cm}^2$ in thickness, deposited on a thick backing, and viewed in the conventional way by a single photomultiplier tube.

In the initial work on ^{235}U and ^{239}Pu , a slow coincidence system (2τ of approximately 250 nsec) was used, with the RCA 6342A phototubes as the coincident fission fragment detectors. Later, a fast coincidence system ($2\tau \simeq 10$ nsec) was used with the faster 56 UVP quartz-windowed phototubes, to minimize backgrounds due to alpha and gamma pileup. This was especially necessary in the ^{233}U measurements.

The bias conditions which were established by requiring a coincidence between both fragments are somewhat complex, and a supplementary set of experiments was undertaken in order to understand them. These experiments, described in detail in a paper by M. S. Moore and L. G. Miller⁽¹¹⁾, consisted of a high resolution measurement of range-energy relations of coincident fission fragments slowed in noble gases.

The results of the range energy studies were used in the present work as follows: It was assumed that the empirical "universal" curve of energy loss as a function of velocity, shown to be valid for electronic stopping of median light and heavy fragments by argon, krypton, and xenon gas and by nickel foil, can be used to describe the response of the detector

shown in Figure 1 to all fission fragments being slowed by copper or nickel foil. With the use of the Lindhard dimensionless range-energy parameters⁽¹²⁻¹⁶⁾ as modified empirically in the Moore-Miller paper⁽¹¹⁾, calculations were made to determine which fragment masses and kinetic energies are selectively viewed in the present experiment. From an initial pulse height distribution of unsloved fragments with known fragment masses and kinetic energies, calculations were made of the pulse height distributions of fragments emerging from the nickel foil at various angles corresponding to equal increments of solid angle. From these residual pulse height distributions the response of the TKE detector could be determined. The calculation involved only one free parameter, the energy below which a fission event was not registered. It was found that the relative number of coincidences to singles agreed very well with the observed rates if it was assumed that fragments emerging from the absorber foil into the scintillation chamber with less than 10 MeV of energy were not detected.

The results of the calculation are shown in Fig. 2-4. These results may be summarized very simply: The observed coincidences are due almost completely to those events in which the heavier of the two fragments has a high kinetic energy. It is important to note this point: The TKE detector has almost no efficiency for detecting changes in the symmetric fission yield. Symmetric fission occurs with low kinetic energy release, and the TKE fission chamber in the present experiment responds to such events with about 15% of the efficiency it has for the highest kinetic energy events. The events for which the detector has the highest response are those in which the most energetic of the heavier of the two fragments

is emitted. It should be noted that these are the fission events in which fine structure has been observed in both the unsloped energy distributions of ^{233}U , ^{235}U and ^{239}Pu fission fragments⁽¹⁷⁻²⁰⁾, and in the slowed velocity distributions⁽²¹⁾ of ^{235}U fragments.

III. Experimental Results

1. ^{235}U

The work done in ^{235}U is the most extensive, both in determination of the relative long-range fragment yield and of the relative symmetric fission yield. In Fig. 5 is plotted the ratio of the counting rate from the "TKE" detector to that from the "conventional" detector, over the whole range of energies studied. These data are in substantial agreement with the earlier work^(4,5), except at the lowest energies (< 0.04 eV). The sharp rise in this ratio near 0.02 eV, previously observed, is not found in the present data.

Plotted also in Fig. 5 are the results of the determination of the relative symmetric yield, as determined by Faler and Tromp⁽⁸⁾. The quantity plotted is the ratio of symmetric to asymmetric fission, or the valley-to-peak ratio. (Since the relative asymmetric fission yield is not found to vary with neutron energy, the above quantity gives the relative variation in the yield of symmetric fission.) There appears to be a qualitative correlation in the results of the two experiments, although opposite in sign to that expected.

2. ^{239}Pu

Shown in Fig. 6 is a comparison of the relative yield of coincident long-range fission fragments and of the relative yield of symmetric fission, as measured by Regier et al.⁽¹⁰⁾. Qualitatively, the correlation is seen to exist as for ^{235}U . Quantitatively, it should be noted that

the changes in the relative symmetric fission yield for ^{239}Pu are much larger than for ^{235}U whereas the changes in the long-range fragment yield are of the same order of magnitude or smaller.

3. ^{233}U

Shown in Fig. 7 is the observed relative variation of long-range fragments for ^{233}U . Plotted also in Fig. 7 are a few data points, taken from Regier et al.⁽⁹⁾, on the relative symmetric fission yield. Again, qualitatively, a correlation appears to exist, though here there are large changes in the relative long-range fragment yield, but only small changes in the symmetric fission yield.

IV. Discussion

In an experiment of this nature, where the experimentalist is trying to measure effects which are very small (approximately a few per cent), it is important that all possible sources of error be considered and eliminated. In the present experiment the following tests and corrections, where necessary, have been made:

1. Background

On the MTR crystal spectrometer, the background, taken for each data point, is determined by turning the crystal 3° clockwise for a predetermined length of time. The adequacy of this background procedure for subtracting the effects of all except Bragg beam neutrons was checked, by interposing a 1/4-inch thick ^{10}B filter in the Bragg beam. With this filter, which had a transmission of 1% for neutrons of 6 eV, the "Bragg-beam" response for all Bragg beam energies < 2 eV is found to be identical to the potential scattering.

2. Second-Order Contamination

Wherever possible, the presence of higher-order neutrons in the Bragg beam was tested with appropriate filters. In particular, ^{235}U data between 0.26 and 0.40 eV were taken with a filter consisting of 0.010-inch thick foils of Rh, Hf and In to completely eliminate neutrons which could form the 1.14 eV resonance in ^{235}U . ^{233}U data between 0.3 and 0.8 eV were obtained with a ^{233}U filter which had a transmission of $\lesssim 0.5\%$ for neutrons of 1.8 and 2.3 eV. The data obtained for neutron energies < 0.04 eV have, in general, not been corrected for the large amount of order contamination which is known to exist because the neutron spectrum peaks at 0.06 - 0.07 eV. It is in the region below 0.04 eV that the present data on ^{235}U are in disagreement with previous data⁽⁵⁾.

3. Variation of Absorber Foil Thickness and of Bias on the Detector

It is assumed that the observed variation in yield of long range fragments with neutron energy is attributed to a combination of a change in the kinetic energy of certain fragments and the geometrical solid angle restrictions imposed by the presence of absorber foils; i.e., the change in yield due to a change in fragment range is $d\Omega/\Omega = \cos \theta / (1 - \cos \theta) dr/r$ where dr/r is the relative change in fragment range, $d\Omega/\Omega$ is the relative change in yield, and θ is the angle the fragment makes with respect to the normal to the absorber foil. The data were generally taken with absorber foils of thickness 0.10×10^{-3} inches. In order to test for the expected increase in the variation of relative yield of long range fragments, the data were also taken with absorber foils of thickness 0.150×10^{-3} inches. The variation in relative long-range heavy fragment yield was expected to be enhanced by perhaps an order of

magnitude. The data obtained, shown in Fig. 8, show an enhancement of perhaps a factor of 4, although the statistical accuracy of the data leaves much to be desired.

One might expect that a similar enhancement of the effect could be obtained by raising the level at which the coincidence detectors are biased. However, detailed calculations show that the variation is not strongly dependent on the bias level. In order to check this, bias levels affecting the two sides of the thick foil in coincidence were set extremely high for one series of runs and extremely low for a second series (giving a factor of 10 difference in count-rate). The effect as seen over the 0.3 eV resonance in ^{235}U showed no appreciable change from the normal bias conditions, within the statistical accuracy obtained. The results of these tests are shown in Fig. 9.

4. Response of the Detector Systems to Prompt Fission Neutrons, Scattered Neutrons, and Gamma Radiation

It has been found that phototubes used in gas scintillation fission detectors are sensitive to some extent to gamma radiation. The present system was tested by replacing the thick (0.6 mg/cm^2) foil of fissile material with a much thicker (0.001-inch) foil of Cd, thus enhancing the effects of gamma sensitivity. Corrections to the data were then calculated, under the assumption that the response of the phototubes to the prompt gammas emitted by fissile materials upon neutron absorption is the same as for Cd capture gammas. These corrections were found to be $\approx 0.01\%$, and were neglected.

The correction for a possible response to prompt fission neutrons was neglected: the assumption is made that $\bar{\nu}$, the average number of prompt neutrons emitted per fission, does not vary with incident neutron

energy^(22,23). Thus a response of the detectors to prompt neutrons should follow the response to all fission events, so that any response of this nature could only decrease the size of the observed effect.

As a measure of the response of the system to scattered neutrons, the chamber was run in the spectrometer Bragg beam with 0.001 inch blank Ni foils (no fissile material). Corrections for this effect were also negligible ($< 0.2\%$).

5. Counting Losses, and the Possibility of a Beam Intensity Dependence

Dead time corrections are expected to be $< 0.3\%$ at the peak of the Maxwellian neutron spectrum. In order to examine a possible neutron beam intensity dependence (other than counting losses), the crystal spectrometer was set at an energy for maximum count rate and 1-inch thick plates of lead were inserted in the Bragg beam to reduce the counting rates in the chamber, in order to obtain the relative yield as a function of beam intensity over a range of a factor of 10, at the same energy and beam composition. The ratio of TKE detector count rate to conventional detector count rate was measured and found to be independent of count rate, to within a statistical error of 2%.

6. Orientation of Chamber

Even though the attenuation of the neutron beam by the fission foils is only expected to be approximately 0.04%, a test was made of possible beam inhomogeneities and differences in response due to changes in position. The chamber (see Fig. 1) was rotated so as to reverse the order in which the neutrons passed through the thin and thick fission foils. Such a detector rotation had no effect on the count rate ratio, indicating that the neutron beam inhomogeneities and attenuation through either foil could be neglected.

7. Gain Shifts

The data were taken automatically on the crystal spectrometer in such a manner that the entire region between 0.8 and 10 eV, or between 0.1 and 2 eV, for example, was obtained in a run lasting for perhaps 12 hours, and then all data points were repeated at 12-hour intervals, in sequence, until the desired statistical accuracy was obtained. Very slight long range gain shifts could be readily detected, by comparing data taken at the same angle in successive runs. It was found that the long range drifts of all systems introduced corrections to the data of $< 0.4\%$.

8. Pulse Height Distributions

In the earlier stages of the work⁽⁵⁾, the pulse height distributions were found to be remarkably good. In the later stages, where the RCA 6342A photomultipliers were replaced with much faster Phillips 56 UVP tubes, the pulse height distributions were found to be somewhat poorer. In Figs. 10 and 11 are shown pulse height distributions of the conventional fission spectrum, and of the TKE detector fission spectra obtained with and without requiring coincidences.

An additional test of possible systematic errors involved a calculation of the relative fission cross section of the various fissile samples as a function of neutron energy, from the conventional detector yield and the beam monitor. The results of the determination of the fission cross section for ^{235}U are shown in Fig. 12. Aside from resolution broadening, the data are in agreement with the accepted ^{235}U fission cross section data of Shore and Sailor⁽²⁴⁾ and Michaudon⁽²⁵⁾.

V. Conclusions

It appears to be fairly well substantiated that there is a correlation between the variation in the yield of symmetric fission, as determined by radiochemical studies⁽⁸⁻¹⁰⁾, and in the yield of long-range heavy fragments, as reported in the present paper. The data taken on the range-energy relations and slowing-down properties of fission fragments in absorbing gases and metal foils seem to show that the longest range heavy fragments correspond to those events which give the highest kinetic-energy release.

The expected correlation of the fission product mass distribution studies as a function of neutron energy with the fragment kinetic energy variation is based on a consideration of the nuclear structure of the fragments at the scission point as suggested by Vandebosch⁽²⁶⁾. Brunner and Paul⁽²⁷⁻³⁰⁾ have discussed the importance of the shell structure of the nascent fragments in determining the fission product mass distribution, and Terrell^(31,32) has noted the effects of fragment shell structure on prompt neutron emission probabilities. It has been reasonably well established⁽³³⁾ that in symmetric fission, the kinetic energy release is anomalously low. This is illustrated, for example, by data taken by Safford⁽³⁴⁾ as shown in Fig. 13. Symmetric fission is found to occur only for relatively low total kinetic energy release. The energy which does not appear as kinetic energy in symmetric fission can be accounted for by a larger number of prompt neutrons emitted by the symmetric fragments.

According to Vandebosch⁽²⁶⁾, the variation in the kinetic energy release with fragment mass number can be attributed to the fragment deformation probability. If both fragments are relatively soft to

deformation, scission would be expected to occur for larger effective separations of the fragment charge centers, resulting in a lower kinetic energy (the case at symmetric fission). If one of the fragments has a nearly closed-shell configuration, then it will be more nearly spherical; the effective separation distance at scission will be relatively lower; the total kinetic energy release will be high; and the number of prompt neutrons emitted, as the fragment reaches its final equilibrium shape, will be low.

If it is assumed that both the symmetric fission yield and the kinetic energy release are determined by the configuration at the scission point, then arguments such as those given above lead one to expect that a lowered average total kinetic energy release should be accompanied by an increased symmetric fission yield, as well as by an increased number of neutrons emitted per fission. The present data appear to show just the opposite behavior, in all cases: An increase in the symmetric fission yield appears to be accompanied by an increased yield of high kinetic energy (long-range) fragments.

There is one possible suggested alternative explanation for the observed correlations: The total kinetic energy distribution of the fragments, which for ^{235}U thermal fission is a bell-shaped curve peaking at 166 MeV and having a width of 27 MeV, may simply undergo a broadening rather than a shift as has been previously assumed. This would imply that an increase in the yield of high kinetic energy fission (longest range fragments) could be accompanied by an increase in the yield of low kinetic energy fission (symmetric fission). In this case, $\bar{\nu}$, the average number of prompt neutrons emitted per fission, could very well remain

constant. The data of Croall and Willis⁽³⁵⁾, which showed an anti-correlation between changes in symmetric and in very asymmetric yields, suggest an even more complicated behavior.

It should be noted that the details of the channel analysis of the data on ^{235}U , previously reported⁽⁵⁾, are not consistent with the present ^{235}U data. The earlier analysis, if carried to energies between 1 and 2 eV predicted that the variation in the yield of high energy fragments should reach and remain at the value observed at 0.2 eV. Instead, as shown in Fig. 3, the yield appears to fall below the value at 0.5 eV. These results are consistent with recent measurements by Sailor et al.⁽³⁶⁾ who reported that the spins of the ^{235}U resonances at 0.3 and 1.1 eV are different. A channel analysis of the type reported previously⁽⁵⁾ is therefore not a fruitful approach. However, it is still highly likely that the variation in fragment kinetic energy is due to the contributions of different fission channels, since the energy variation in the present data follows the variation observed by Dabbs et al.⁽³⁷⁾, in the value of K , the projection of angular momentum on the nuclear symmetry axis.

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Figure Captions

Figure 1 - Pictorial view of the fission chamber in which the variation of fission fragment kinetic energy with neutron energy was detected.

Figure 2 - The energy distribution of ^{235}U fission fragments to which the TKE detector responds (solid line) and to which the conventional detector responds (dashed line). The TKE detector is seen to have a relatively enhanced response to events in which the kinetic energy of the heavy binary fragment is greatest.

Figure 3 - The total kinetic energy distribution of ^{235}U fission fragments to which the TKE detector responds (solid line) and to which the conventional detector responds (dashed line).

Figure 4 - The mass distribution of ^{235}U fission fragments to which the TKE detector responds (solid line) and to which the conventional detector responds (dashed line). The response both to symmetric events and to very asymmetric events is relatively low. (Cf. Fig. 13).

Figure 5 - Ratio of the ^{235}U fission fragment counting rate from the detector to that from the conventional detector over the whole range of energies studied. The relative symmetric yield as determined by Faler and Tromp is plotted over the same energy region, the ordinate scale being on the right hand side of the figure.

Figure 6 - Ratio of the ^{239}Pu fission fragment counting rate from the detector to that from the conventional detector over the whole range of energies studied. Relative symmetric yield as determined by Regier et al.⁽¹⁰⁾ is plotted over the same energy region, the ordinate being given on the right hand side of the figure.

Figure 7 - Ratio of the ^{233}U fission fragment counting rate from the TKE detector to that from the conventional detector over the whole range of energies studied. Relative symmetric yield as determined by Regier et al.⁽⁹⁾ is plotted over the same energy region, the ordinate being given on the right hand side of the figure.

Figure 8 - Variation in long-range heavy fragment yield for 0.0001 inch and 0.00015 inch thick nickel absorber foils.

Figure 9 - Variation in long-range heavy fragment yield for high, medium and low-bias levels.

Figure 10 - ^{235}U fission pulse height distribution obtained from the conventional detector shown in Figure 1.

Figure 11 - ^{235}U fission pulse height distribution obtained from the TKE detector shown in Figure 1 with and without requiring coincidence.

Figure 12 - ^{235}U fission cross section obtained from thin foil yield and beam monitor.

Figure 13 - Relative fission probability as a function of the kinetic energy of one of the fragments, for various values of the total kinetic energy release. These data are those of Safford⁽³⁴⁾ for ^{235}U .

FIGURE 1

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FIGURE 2

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FIGURE 3

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FIGURE 4

FIGURE 5

FIGURE 6

FIGURE 7

FIGURE 8

FIGURE 9

FIGURE 10

FIGURE 11

FIGURE 12

FIGURE 13