

Alpha-Particle-Induced Fission of Th^{230} , Th^{232} , and U^{233} †

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Measurements have been made of the kinetic energies of coincident-fission fragment pairs emitted in the 22.1-, 25.7-, and 29.5-MeV He^4 -induced fission of Th^{230} , Th^{232} , and U^{233} to determine the mass and total kinetic-energy distributions of the fragments. The results are found to be consistent with the hypothesis of two distinct fission modes, corresponding to predominantly symmetric and predominantly asymmetric mass divisions. From a comparison of the mass distributions obtained from double-energy measurements with the mass distributions obtained from double-velocity time-of-flight measurements, it is possible to deduce the average number of prompt neutrons emitted as a function of the fragment mass. These results are also found to be qualitatively consistent with the two-mode hypothesis.

1. INTRODUCTION

IN a previous study¹ it was found that the mass and kinetic-energy distributions from the charged particle-induced fission of Ra^{226} , Th^{230} , and U^{233} could be interpreted in terms of two distinct fission modes which produced predominantly symmetric and predominantly asymmetric mass divisions, respectively. Assuming that the total kinetic-energy release is due to the Coulomb repulsion of the two fragments, the results indicated that for each mode the average distance between the two charge centers at scission was approximately independent of the mass ratio of the fragments. However, this "breaking distance" was found to be greater for the symmetric mode than for the asymmetric mode. This larger breaking distance corresponded to a smaller total kinetic-energy release for symmetric mode fission. Furthermore, for each mode it was found that the widths of the total kinetic-energy distributions (or distributions of breaking distances) were also approximately independent of the fragment mass ratio. In addition, it has been shown in other studies that the hypothesis of two fission modes was consistent with the variation in the radiochemical mass distributions as a function of the excitation energy of the compound nucleus^{2,3} and also with the data on charge distributions from high-energy proton-induced fission.⁴

It appears that the empirical hypothesis of two fission modes gives a reasonable characterization of the fission process at moderate excitation energies if the two modes are assigned the following general characteristics: (1) For each mode the breaking distance and the distribution of breaking distances is approximately independent of the fragment mass ratio; (2) the average breaking distance is greater for symmetric mode than for asym-

metric mode fission; (3) the two modes yield predominantly symmetric and predominantly asymmetric mass distributions, respectively, with the shapes of the mass distributions for each mode being approximately independent of the excitation energy for a given fissioning nucleus; and (4) the probability of fission from the symmetric mode increases relative to asymmetric mode fission as the excitation energy of the fissioning system is increased.

It has also been suggested⁵⁻⁷ that the general features of these results may be due to the rigidity of closed-shell neutron or proton configurations in the emitted fragments. A theory (hereafter referred to as the "fragment-shell theory"), based on the special properties of these closed-shell configurations, has been shown⁵⁻⁷ capable of qualitatively reproducing some of the general features of the data without resorting to the empirical hypothesis of two distinct fission modes.

The purpose of the experiment reported here was to investigate in more detail the fission of heavy nuclei at moderate excitation energies to obtain more information on the validity of the two-mode hypothesis. In this experiment, measurements were made of the kinetic energies of the two coincident fragments produced by He^4 -induced fission of Th^{230} , Th^{232} , and U^{233} at laboratory bombarding energies of 22.1, 25.7, and 29.5 MeV. The results are consistent with the two-mode interpretation and yield additional information concerning the properties required by the two modes.

In addition, these fission reactions have been investigated using time-of-flight techniques to measure the velocities of the two emitted fragments.⁸ Using the methods described by Terrell,⁷ a comparison between the mass distributions obtained by double-energy measurements and those that are obtained by double-velocity experiments gives the average number of neutrons emitted as a function of the fragment mass.

† Work performed under the auspices of the U. S. Atomic Energy Commission.

¹ H. C. Britt, H. E. Wegner, and Judith C. Gursky, *Phys. Rev.* **129**, 2239 (1963).

² M. Turkevich and J. B. Niday, *Phys. Rev.* **84**, 52 (1951).

³ H. B. Levy, H. G. Hicks, W. E. Nervi, P. C. Stevenson, J. B. Niday, and J. C. Armstrong, Jr., *Phys. Rev.* **124**, 544 (1961); H. G. Hicks, H. B. Levy, W. E. Nervi, P. C. Stevenson, J. B. Niday, and J. C. Armstrong, Jr., *ibid.* **128**, 700 (1962).

⁴ A. C. Pappas and J. Alstad, *J. Inorg. Nucl. Chem.* **17**, 195 (1961); **15**, 222 (1960); G. Rudstam and A. C. Pappas, *Nucl. Phys.* **22**, 468 (1961).

⁵ P. Fong, *Bull. Am. Phys. Soc.* **8**, 385 (1963) and private communication.

⁶ R. Vandenbosch, *Nucl. Phys.* **46**, 129 (1963).

⁷ J. Terrell, *Phys. Rev.* **127**, 880 (1962).

⁸ S. L. Whetstone, Jr., following paper, *Phys. Rev.* **133**, B613 (1964).

The neutron distributions obtained provide an additional test of the two-mode hypothesis.

2. EXPERIMENTAL PROCEDURE

In this experiment the mass and total kinetic-energy distributions of the fragments are obtained from semiconductor-detector measurements of the kinetic energies of the two coincident fragments. The two semiconductor detectors were mounted in a back-to-back arrangement at an angle of 90° relative to the beam direction. The details of this chamber and the experimental procedure have been described previously.¹

The energy calibration of the semiconductor detectors was obtained by observing fragments from the spontaneous fission of Cf^{252} and comparing the results to the double-velocity measurements of Whetstone.⁹ The observed kinetic energies correspond to the kinetic energies of the fragments after the emission of prompt neutrons. For each mass division the measured average final total kinetic-energy release was corrected to give the average initial total kinetic-energy release before neutron emission, using for the average number of prompt neutrons emitted values obtained from energy-balance considerations. These corrections and the data analysis procedures have been described in detail previously.^{1,10}

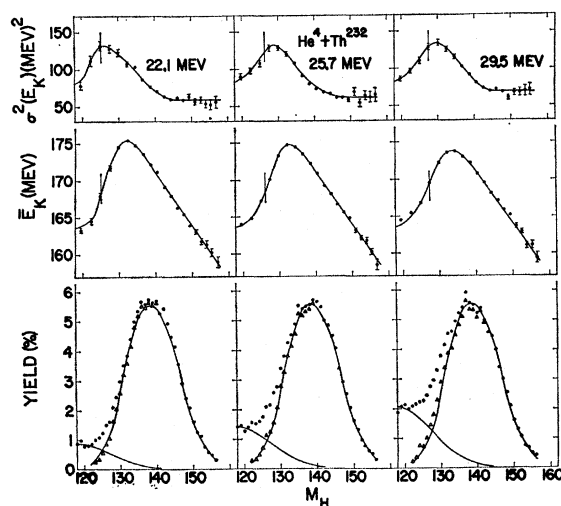


FIG. 1. Yield, average initial total kinetic energy, and variance of the total kinetic energy as a function of the mass of the heavy fragment for the $\text{He}^4 + \text{Th}^{232}$ reactions. Closed circles in the yield distributions represent the gross measured yields. Open triangles in the yield distribution represent the values obtained for the yield from the asymmetric mode as described in the text. The distribution shown by the solid line for the symmetric mode has the assumed shape described in the text, and the solid-line asymmetric distribution represents the best fit by eye to the data taken at the three energies and is the same for all three cases. Yield distributions have each been normalized to give an area of 200% for the asymmetric component. The vertical markers on the \bar{E}_K and $\sigma^2(E_K)$ distributions indicate the masses at which an equal yield was obtained for symmetric and asymmetric mode fission.

⁹ S. L. Whetstone, Jr., Phys. Rev. **131**, 1232 (1963).

¹⁰ H. C. Britt, H. E. Wegner, and S. L. Whetstone, Jr., Nucl. Instr. Methods **24**, 13 (1963).

The targets used had areal densities of $10\text{--}30 \mu\text{g}/\text{cm}^2$ and were prepared by evaporation onto nickel backings of $50\text{--}100 \mu\text{g}/\text{cm}^2$. These same targets were used in the double-velocity experiment.⁸

For these experiments the cyclotron conditions were made as similar as possible to those used for the double-velocity measurements.⁸ The average beam energies were within 0.2 MeV of the average energies used in the velocity measurements and during the measurements the beam energy was held to within ± 0.2 MeV of its average value. In the energy measurements the solid angle subtended by the detectors was approximately 9×10^{-2} sr as compared to 4×10^{-3} sr for the velocity measurements.

3. EXPERIMENTAL RESULTS

The mass distributions, the average initial total kinetic-energy release \bar{E}_K , and the variance of the average initial total kinetic-energy release $\sigma^2(E_K)$, as functions of the fragment mass, are shown in Figs. 1–3. The gross characteristics of these distributions are shown in Table I. In each case the results are based on a total of $3\text{--}4 \times 10^4$ recorded events.

The results of the present experiment are very similar to those obtained earlier.¹ In contrast to the Ra^{226} reactions investigated earlier,¹ the symmetric yield is small compared to the asymmetric yield, and the asymmetric peak is closer to the symmetric mass division. Therefore,

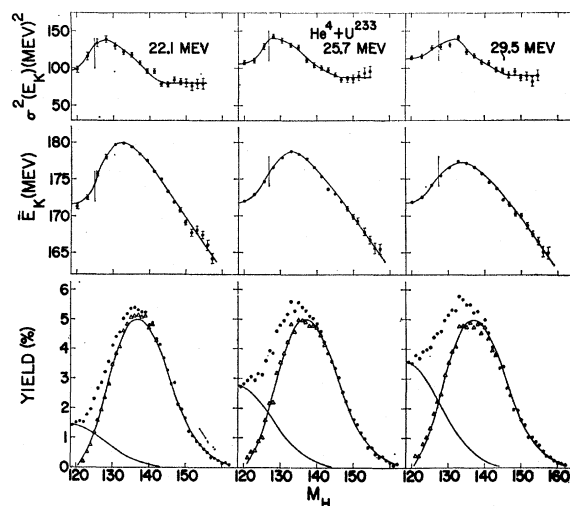


FIG. 2. Yield, average initial total kinetic energy, and variance of the total kinetic energy as a function of the mass of the heavy fragment for the $\text{He}^4 + \text{U}^{233}$ reactions. Closed circles in the yield distributions represent the gross measured yields. Open triangles in the yield distribution represent the values obtained for the yield from the asymmetric mode as described in the text. The distribution shown by the solid line for the symmetric mode has the assumed shape described in the text, and the solid-line asymmetric distribution represents the best fit by eye to the data taken at the three energies and is the same for all three cases. Yield distributions have each been normalized to give an area of 200% for the asymmetric component. The vertical markers on the \bar{E}_K and $\sigma^2(E_K)$ distributions indicate the masses at which an equal yield was obtained for symmetric and asymmetric mode fission.

results from a detailed two-mode analysis of the type performed for the Ra^{226} reactions become uncertain because of the difficulty in determining the distributions of total kinetic-energy release (or breaking distances) for the symmetric mode fissions; consequently, a detailed two-mode analysis of this type was not performed.

However, an attempt was made to apply a two-mode interpretation to the observed mass distributions shown in Figs. 1-3. It was assumed that the symmetric mass component had the same shape as the component observed earlier for the Ra^{226} reactions.¹ For each reaction this component was normalized at symmetry and then subtracted from the observed mass distribution. (This procedure involves the assumption that there is no yield from the asymmetric mode at symmetry.) The resulting mass distributions for the asymmetric mode were normalized to an area of 200% and are shown in Figs. 1-3. The solid-line fit to the asymmetric mass distributions has the same shape at all bombarding energies for a given reaction.

TABLE I. Characteristics of the total kinetic-energy distributions for the reactions studied.^a

Target	E_{lab} (MeV)	$\langle E_{K'} \rangle$ (MeV)	$\langle E_K \rangle$ (MeV)	$\sigma^2(E_K)$ (MeV) ²	Double velocity ^b $\langle E_K \rangle$
Th^{230}	25.7	166.0±2	169.6	108±6	167.5
	29.5	164.9±2	169.0	110±6	166.0
Th^{232}	22.1	167.5±2	171.4	101±6	169.1
	25.7	166.8±2	171.1	103±6	168.2
U^{233}	29.5	166.0±2	170.7	107±6	167.0
	22.1	172.4±2	175.5	119±6	176.3
	25.7	171.1±2	174.6	123±6	174.9
	29.5	169.9±2	173.9	123±6	174.2

^a E_{lab} is the laboratory bombarding energy for the He^4 particles. The quantities $\langle E_{K'} \rangle$ and $\langle E_K \rangle$ are the average final total kinetic-energy release and the average initial total kinetic-energy release, respectively. $\sigma^2(E_K)$ is the variance of the average total kinetic-energy distribution.

^b The double-velocity results are reported in more detail in Ref. 8. In some cases, there is a slight difference between the bombarding energies, E_{lab} , for the double-energy and the double-velocity measurements.

Thus, it can be seen that although the relative competition between the two modes varies quite rapidly with the excitation energy, the mass distributions can be resolved into two components whose shapes for a given reaction are approximately independent¹¹ of the excitation energy. Also, the heavy mass peaks for the asymmetric mode show similar shapes for the three reactions, as is seen in Fig. 4. This constancy in shape and position of the heavy mass peak resembles trends that have been previously noted for asymmetric fission of heavy elements at low-excitation energies.¹²

Also shown in Figs. 1-3 are the values obtained for

¹¹ For a given reaction the distribution obtained for the asymmetric component does show a slight broadening as the excitation energy of the compound nucleus increases. This is consistent with a slight broadening of the symmetric-component shape as the excitation energy is increased, as has been observed previously for symmetric fission reactions [E. F. Neuzil and A. W. Fairhall, Phys. Rev. **129**, 2705 (1963)].

¹² I. Halpern, Ann. Rev. Nucl. Sci. **9**, 245 (1959).

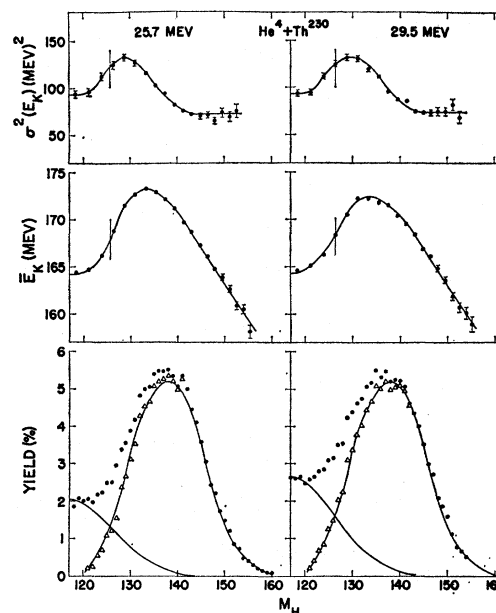


FIG. 3. Yield, average initial total kinetic energy, and variance of the total kinetic energy as a function of the mass of the heavy fragment for the $\text{He}^4+\text{Th}^{230}$ reactions. Closed circles in the yield distributions represent the gross measured yields. Open triangles in the yield distribution represent the values obtained for yield from the asymmetric mode as described in the text. The solid-line distribution shown by the solid line for the symmetric mode is the assumed shape described in the text, and the solid-line asymmetric distribution represents the best fit by eye to the data taken at the two energies and is the same for both cases. Yield distributions have each been normalized to give an area of 200% for the asymmetric component. The vertical markers on the $\sigma^2(E_K)$ and E_K distributions indicate the masses at which an equal yield was obtained for symmetric and asymmetric mode fissions.

\bar{E}_K and $\sigma^2(E_K)$ as a function of the mass of the heavy fragment. A composite of all of these distributions is shown in Fig. 5. Both the \bar{E}_K and the $\sigma^2(E_K)$ distributions show variations (Fig. 5) with the excitation energy

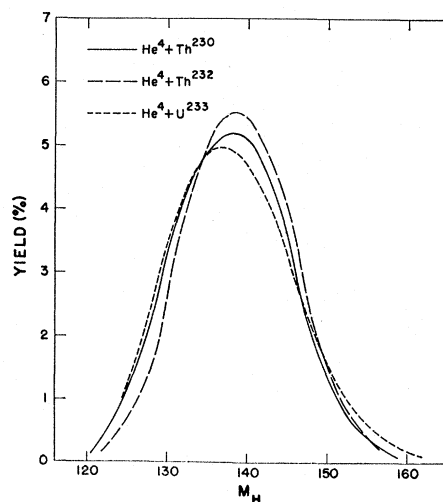


FIG. 4. Composite of the mass distributions for the asymmetric mode obtained in the three reactions.

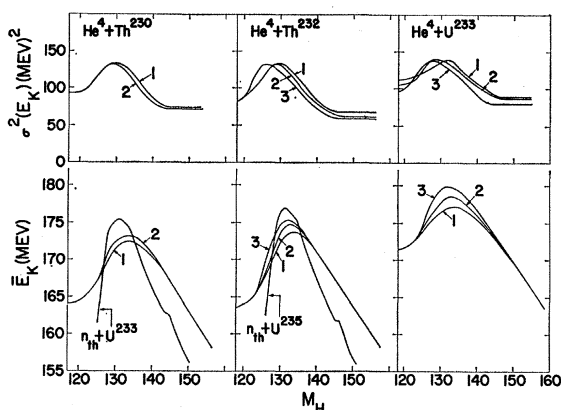


Fig. 5. Composite of the distributions obtained for the average initial total kinetic energy and the variance of the average initial total kinetic energy as functions of the mass of the heavy fragment. Also shown are the \bar{E}_K distributions obtained by Milton and Fraser (see Ref. 13) for thermal neutron-induced fission of U^{233} and U^{235} . The curves labeled 1, 2, and 3 refer to bombarding energies of 29.5, 25.7, and 22.1 MeV, respectively.

(or He^4 bombarding energy) that are qualitatively consistent with the predictions of the two-mode hypothesis. As the excitation energy is increased, the peak in the $\sigma^2(E_K)$ distribution shifts to a larger value of m_H and the \bar{E}_K distributions begin to increase rapidly at a larger value of m_H and reach a smaller maximum \bar{E}_K . These characteristics are related to the strong dependence on excitation energy for the symmetric yield relative to the yield from the asymmetric mode. Figures 1–3 show that as the excitation energy is increased there is a corresponding increase in the value of m_H at which the two modes give equal contributions. In the two-mode interpretation the peak in the $\sigma^2(E_K)$ distribution is a result of a combination of the two total kinetic-energy distributions which are centered at two different average total kinetic energies. The position of this peak, therefore, is correlated with the value of m_H for which there are equal contributions from the two modes. This correlation is clearly seen in Fig. 5. Similarly, \bar{E}_K at any value of m_H is a weighted average of the average total kinetic energies for the two modes. The results shown in Fig. 5 are consistent with the average total kinetic energy for each mode as a function of m_H being independent of the excitation energy. Then the variation of \bar{E}_K with excitation energy in the transition region ($122 < m_H < 144$) simply reflects the variation with excitation energy of the symmetric yield relative to the asymmetric yield (see Figs. 1–3).

In the simple model that was discussed in Ref. 1, it was assumed that the intrinsic value for $\sigma^2(E_K)$ for each mode was the same, so that the peak in the measured $\sigma^2(E_K)$ curve should occur at the mass number corresponding to equal contributions from both modes. However, in the current investigation, and in the previous one¹ for cases where the asymmetric mode was dominant, it was found that the peak in the $\sigma^2(E_K)$ distribution occurred at a value of m_H which is slightly greater

than the value corresponding to equal contributions from the two modes. Therefore, if the two-mode interpretation is correct, the intrinsic value for $\sigma^2(E_K)$ in the transition region must be different for the two modes. The $\sigma^2(E_K)$ distributions from this experiment and the previous one¹ are consistent with the intrinsic values of $\sigma^2(E_K)$ for the symmetric mode being approximately independent of m_H as is found¹ for He^3 -induced fission of Bi and Au and the intrinsic value of $\sigma^2(E_K)$ for the asymmetric mode increasing with decreasing m_H in the region $m_H < 140$ as is found¹³ for thermal neutron-induced fission.

Figure 5 also shows the \bar{E}_K distributions obtained by Milton and Fraser¹³ for thermal neutron-induced fission of U^{233} and U^{235} . These reactions form the same compound nuclei as the He^4 -induced fission of Th^{230} and Th^{232} but at a lower excitation energy. It can be seen in Fig. 5 that at large values of m_H the kinetic energy decreases much more rapidly for the thermal neutron reactions than for the He^4 -induced reactions, but that for the range of energies studied in the He^4 reactions, the kinetic-energy release at large values of m_H is independent of the excitation energy. For the He^4+Th^{232} reactions the excitation energies for first-chance fission range from 10.5 to 17.8 MeV above the $n_{th}+U^{235}$ excitation energies. The presence of large amounts of second- and third-chance fission gives a lower excitation energy range (12.6 to 7.7 MeV) for the average fissioning nucleus (see Appendix). Figure 5 indicates for the He^4 -induced fission reactions that over an excitation energy range of ~ 5 MeV the \bar{E}_K distribution for the asymmetric mode is independent of the excitation energy. Thus, it appears that the \bar{E}_K distribution from thermal neutron-induced fission is not the same as the \bar{E}_K distribution for the asymmetric mode in higher excitation energy-fission reactions. This difference may be due to the limited energy available near the fission threshold or, alternatively, to the different angular momenta of the fissioning nuclei in the two cases. A similar effect is seen in the mass distributions shown in Fig. 6. Here, the mass distribution for the asymmetric component from the He^4

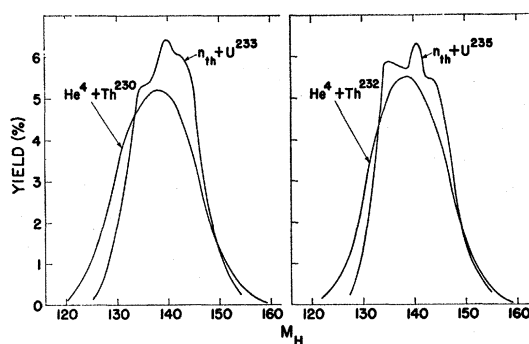


Fig. 6. Mass distributions obtained for the asymmetric component in the He^4+Th^{230} and He^4+Th^{232} reactions compared to the thermal-neutron results of Milton and Fraser (see Ref. 13).

¹³ J. C. D. Milton and J. S. Fraser, Can. J. Phys. **40**, 1626 (1962).

reactions was approximately independent of the excitation energy, but this component has a slightly different shape from the mass distribution observed in thermal neutron-induced fission.

4. NEUTRON EMISSION FROM THE FRAGMENTS

a. Method

It has been pointed out by Terrell⁷ that the fragment mass distributions obtained from measurements of the kinetic energies of the two fragments can be significantly distorted by prompt neutron emission. This is because the kinetic energies that are measured are the final energies of the fragments after the emission of prompt neutrons. Since the final fragment velocities are on the average nearly equal to the initial fragment velocities, the ratio of the final fragment energies can be written as

$$\frac{E_L}{E_H} = \frac{m_L - \nu_L}{m_H - \nu_H} \left(\frac{V_L}{V_H} \right)^2 = \frac{m_L - \nu_L}{m_H - \nu_H} \left(\frac{m_H}{m_L} \right)^2, \quad (1)$$

where ν_L and ν_H are the number of prompt neutrons emitted from the fragments with initial masses m_L and m_H . The common assumption used in obtaining the mass distributions from measurements of the fragment kinetic energies is that

$$E_L/E_H = m_H^*/m_L^*, \quad (2)$$

where m_L^* and m_H^* are the "pseudomasses" obtained by double-energy measurements. Terrell⁷ has shown from Eqs. (1) and (2) that the error introduced into the double-energy mass distributions by transforming the data using Eq. (2) is given by

$$m_L^* - m_L \cong \Delta m \cong \frac{m_H \nu_L - m_L \nu_H}{A} = \nu_L - \frac{m_L \nu_T}{A}. \quad (3)$$

This neutron distortion effect has been illustrated in a previous paper.¹⁰

Because of the neutron-distortion effect, the mass distribution obtained from double-energy measurements is the same as the initial mass distribution (i.e., $\Delta m = 0$) only for the special case where $\nu_L/\nu_H = m_L/m_H$. It can be seen from Eq. (3) that ν_L (and ν_H) can be determined from a comparison of the mass distributions obtained in double-energy and double-velocity experiments if $\nu_T = \nu_L + \nu_H$ is known as a function of the mass division. At present, there are no direct measurements of ν_T as a function of the mass division for any of the reactions studied here. However, it is possible to calculate ν_T for these cases using energy-balance considerations. These calculations are described in more detail in the Appendix.

To obtain values for Δm , the mass distributions from the two experiments must first be corrected for the dispersion due to the emission of the prompt neutrons. For both measurements these neutron dispersions were estimated using the relations given by Terrell.⁷ For the

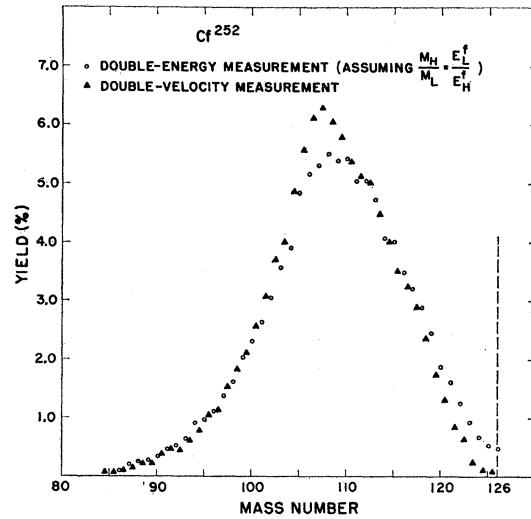


FIG. 7. Mass distributions obtained for fragments from the spontaneous fission of Cf^{252} by double-energy and double-velocity techniques. The mass distribution has been obtained from the double-energy measurement by assuming that the ratio of the initial fragment masses is equal to the ratio of their measured final energies.

double-energy measurements the additional experimental dispersion was found to be negligible in comparison to the neutron dispersion,¹⁰ and for the double-velocity measurements an estimated experimental dispersion⁸ was added to the calculated dispersion due to neutron emission. These dispersions were then unfolded from the observed mass distributions using both the method of negative variances described by Terrell⁷ and a fitting procedure which is described by Whetstone.⁸ The data treated by these two methods yielded essentially the same values for Δm . After the dispersion was removed from the mass distributions, values were obtained for Δm by comparing the cumulative mass distributions in a manner similar to that used by Terrell.⁷

Since the energy measurements by semiconductor detectors are not absolute, but must be referred to an energy calibration based on time-of-flight results and since the response of the semiconductor detectors is still not completely understood,¹⁴ it is very difficult to estimate the systematic errors that may be present in the double-energy mass distributions due to errors in the energy calibration of the detectors. For this reason it was thought valuable to compare the results obtained for Cf^{252} by the above method with determinations of ν which have been made by Bowman *et al.*¹⁵ and Terrell.⁷ Figure 7 shows the mass distributions obtained by the two techniques. It is seen that the mass distribution obtained from the double-energy measurements is significantly distorted compared to the initial mass distribution obtained in the double-velocity measurements. The values obtained for ν from these two mass distribu-

¹⁴ H. C. Britt and H. E. Wegner, *Rev. Sci. Instr.* **34**, 274 (1963).

¹⁵ H. R. Bowman, J. C. D. Milton, S. G. Thompson, and W. J. Swiatecki, *Phys. Rev.* **129**, 2133 (1963).

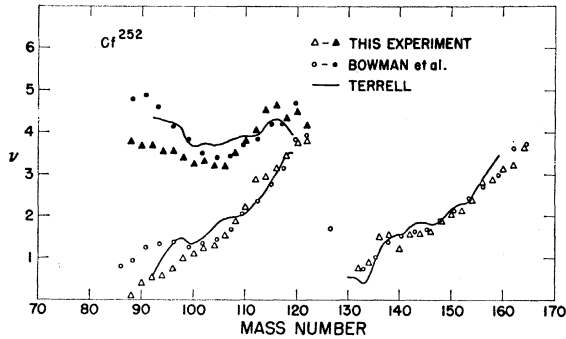


FIG. 8. Results obtained for the average number of prompt neutrons as a function of the fragment mass for the spontaneous fission of Cf^{252} . The solid triangles represent the calculated values for the average total number of neutrons emitted from both fragments (see Appendix). Also shown are the results of Bowman *et al.* (Ref. 15) and Terrell (Ref. 7).

tions and the values calculated for ν_T as described in the Appendix are shown in Fig. 8 compared to the direct measurements of Bowman *et al.*¹⁵ and the results obtained by Terrell⁷ from a comparison of double-velocity and radiochemical data. The results of this experiment agree quite well with the results obtained by other methods except that at very asymmetric mass divisions the calculated values for ν_T appear to be somewhat low. In this region, however, the estimated uncertainties are large for all of the results.

b. Results

For the He^4 results detailed comparisons were made for 29.5- and 25.7-MeV He^4 -induced fission of Th^{230}

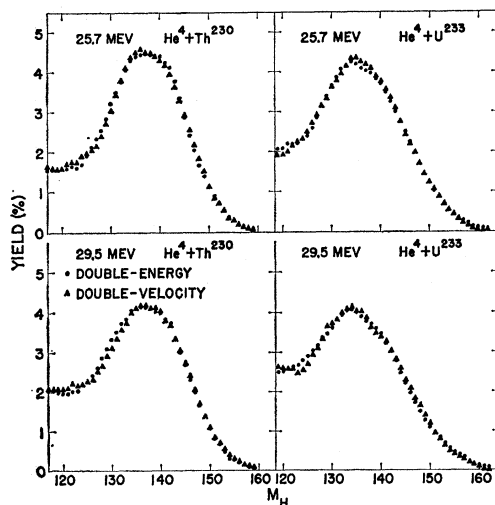


FIG. 9. Mass distributions obtained for fragments from the He^4 induced fission of U^{238} and Th^{230} by double-energy and double-velocity techniques. The mass distribution has been obtained from the double-energy measurement by assuming that the ratio of the initial fragment masses is equal to the ratio of their measured final energies. Masses for which only one point is visible indicate that the yield from the double-energy and double-velocity measurements is the same.

and U^{238} . These cases represent the cases for which the double-velocity data are believed to be most reliable.⁸ The mass distributions obtained in the double-energy measurements, shown in Fig. 9, are almost identical to the initial mass distributions from the double-velocity experiments. This indicates that the neutrons are distributed approximately according to the relationship $\nu_L/\nu_H = m_L/m_H$. The values obtained for ν and the calculated values of ν_T are shown in Fig. 10. Note that the calculated values of ν_T vary considerably over the mass range of interest, so that the distributions obtained for ν reflect both the variations in the relative excitation energies of the fragments and the variations in ν_T . Figure 11 shows ν/ν_T as a function of the fragment mass. In this form the results give an indication of the relative excitation energy for the fragments and, furthermore, the values obtained for ν/ν_T are not as sensitive to uncertainties in the ν_T calculations as are the values of ν .

Uncertainties in the values for ν result from uncertainties in ν_T and in the values of Δm which are obtained from a comparison of the two sets of data. The uncertainties in ν_T , which are discussed in the Appendix, do not have a gross effect on the ν/ν_T distribution, at least to first order. Estimates have been made of (1) the uncertainties in the values of Δm that are due to the statistical uncertainties in the mass distributions, (2) the uncertainties in the estimated mass dispersion and the method of correcting for dispersion,

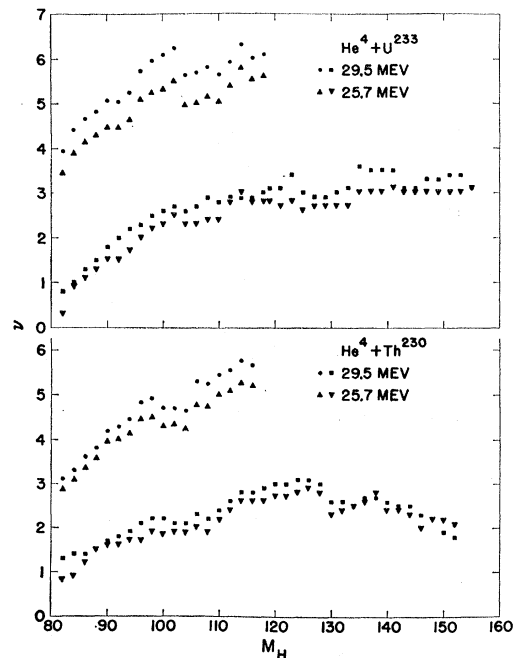


FIG. 10. The results obtained for the number of neutrons emitted as a function of the fragment mass for the He^4 -induced fission of U^{238} and Th^{230} . The circles and triangles represent the calculated values for the average total number of neutrons emitted from both fragments.

and (3) the uncertainties in the absolute calibration of the detectors. The largest uncertainties were associated with energy calibration of the semiconductor detectors, and they have been estimated to be less than ± 0.3 mass unit over the mass region shown in Figs. 10 and 11. The uncertainties mentioned above are also present in the Cf^{252} results shown in Fig. 8, which agree with previous measurements.^{7,15} In addition to the uncertainties mentioned above, there is a possibility of systematic errors in the results which are due to the uncertainties in response of semiconductor detectors to fission fragments.¹⁶

In addition to double-energy, double-velocity comparisons, distributions of ν as a function of fragment mass have been obtained by two other indirect methods.^{7,17} A method used by McHugh¹⁷ involves the analysis of mass-spectrometric results obtained for the most probable charge as a function of the final mass of the fragment. From the charge distribution it was shown that it is possible to obtain information on ν_T as a function of the fragment mass ratio. In addition, if the most probable charge as a function of the initial fragment mass is known, it is possible to obtain ν as a function of the fragment mass. Using this approach, the charge distributions obtained for He^4 -induced fission of Th^{232} and U^{235} were analyzed to obtain distributions of ν and ν_T as a function of the fragment mass. The values of ν were obtained by assuming that the most probable fragment charge as a function of the initial fragment mass is given by the minimum potential energy (MPE) theory which proposes that the distribution of nucleons in the initial fragments is determined by minimizing the sum of the nuclear potential energy and the Coulombic repulsion energy. The results which were obtained¹⁷ for ν_T and ν in the mass region $126 \leq m \leq 150$ give values for ν which are approximately independent of the fragment mass. For light fragments in the region $80 \leq m \leq 86$ the values obtained for ν were found to increase with increasing mass. These trends¹⁷ are es-

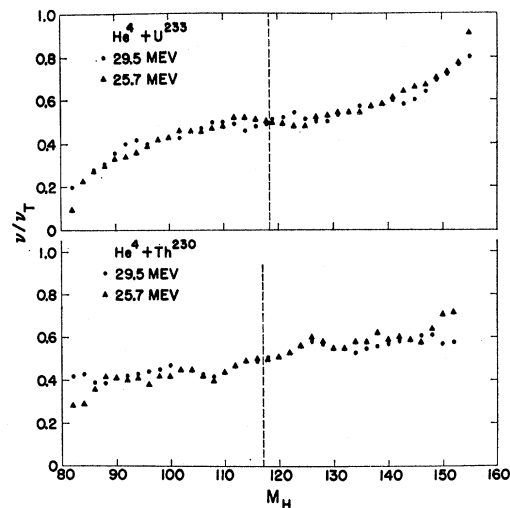


FIG. 11. The ratio of the average number of neutrons from each fragment to the total number of neutrons emitted from both fragments as a function of the fragment mass. Masses for which only one point is visible indicate that the value for ν/ν_T is the same for both bombarding energies.

entially the same as are observed in the present results (Fig. 10).

It is also possible to obtain distributions of ν as a function of the fragment mass from a comparison of the initial mass distributions from double-velocity experiments with the final mass distributions obtained by radiochemical means. This technique has been used successfully by Terrell⁷ for spontaneous and thermal neutron-induced fission reactions. A comparison of this type was made between radiochemical and time-of-flight results for He^4 -induced fission of U^{233} and between radiochemical data for 14-MeV neutron-induced fission of U^{235} and time-of-flight data for 25.7-MeV He^4 -induced fission of Th^{232} . In the latter case these two different reactions form the same compound nucleus at approximately the same excitation energy. The results obtained from this comparison for ν as a function of the fragment mass are shown in the following paper.⁸ For both of these reactions the ν distributions which are obtained show a peak in the region of mass 105 and a corresponding minimum near mass 130. This structure is not evident in the results obtained from the comparison of double-energy and double-velocity results (Fig. 10) or in the mass-spectrometric results.¹⁷

The interpretation of the ν distribution depends very sensitively on whether or not the structure observed in the results from the comparison of the radiochemical and double-velocity data is real. For the case of 25.7-MeV He^4 -induced fission of U^{233} the radiochemical data consists of a small number of points with poorly known uncertainties. Since this case was also investigated by a comparison of double-energy and double-velocity data, it is possible to make a different type of comparison. From the result shown in Fig. 10 for ν as a function of the fragment mass and the prompt mass distribution,

¹⁶ One additional possible source of error in these measurements, which cannot be estimated at the present time, is associated with the method used to calibrate the semiconductor detectors. The calibration, which has been described previously (see Ref. 10), involves a comparison of the semiconductor detector data with time-of-flight results for Cf^{252} . At present, the response of semiconductor detectors as a function of the mass and energy of the fragments is not completely understood; it may be that the calibration appropriate for Cf^{252} fragments is not appropriate for the calibration for fragments from a different fissioning nucleus with a different distribution of fragment mass and kinetic energy. However, at present, it seems most reasonable to assume that the response of semiconductor detectors to various mass fragments is sufficiently similar so that the pulse-height-to-energy calibration can be represented by a linear function which is the same for all fissioning nuclei and which can be determined by calibration with Cf^{252} fragments. The strongest evidence of this is that, using a linear calibration based on Cf^{252} results, the \bar{E}_K versus mass distributions for the He^4 -induced fission reactions being investigated agree reasonably well with the \bar{E}_K versus mass distributions obtained from double-velocity experiments (see Table I and Ref. 10).

¹⁷ J. A. McHugh, Jr., University of California Radiation Laboratory Report, UCRL-10673 (unpublished).

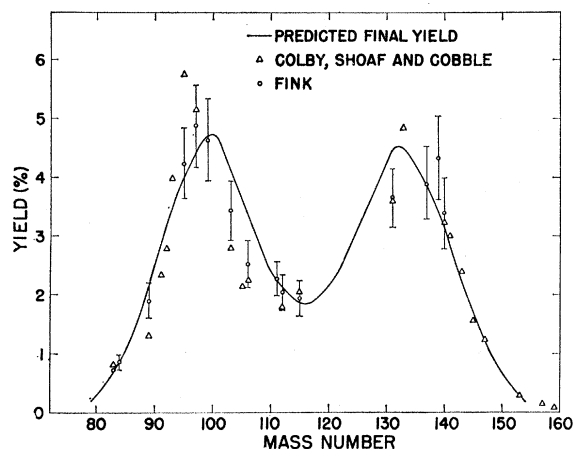


FIG. 12. The results obtained for the final-mass yield distribution compared to the radiochemical measurements of Colby, Shoaf, and Cobble (Ref. 18) and Fink (Ref. 19) for 25.7-MeV He^4 -induced fission of U^{238} . The error bars represent $\pm 15\%$ uncertainties which were suggested by the experimenter (Ref. 19) as the approximate accuracy for this type of measurement.

it is possible to generate a final-mass yield distribution and then compare this distribution to the measured radiochemical points. This has been done for 25.7-MeV He^4 -induced fission of U^{238} , and the results are shown in Fig. 12. It is seen that the calculated final mass distribution is in fair agreement with the results of Fink¹⁸ especially since he has indicated that the points at mass numbers 103 and 105 may be somewhat low. Agreement with the results of Colby *et al.*¹⁹ is also satisfactory if the radiochemical points in the region 103–112 are low and the point at 95 is high. This comparison and the large discrepancies between the radiochemical measurements in the two experiments^{18,19} indicate that the reliability of the structure obtained in the comparison of radiochemical and double-velocity results is in considerable doubt for the case of He^4 -induced fission of U^{238} .

For the case of He^4 -induced fission of Th^{232} the radiochemical mass distribution used in the comparison is more complete. In this case the ν distribution obtained from the comparison of radiochemical and double-velocity data is similar to the results in Fig. 10 except for a moderately well-defined peak near mass 105 and a corresponding minimum near mass 130. In this case the uncertainties in the ν distribution due to the statistical uncertainties in the time-of-flight data are greater, but these uncertainties do not seem to be great enough to negate the structure which is observed. Again, this structure could be generated by systematically low values for the radiochemical measurements in the region of mass 105.

It is possible that the structure observed in the comparison of radiochemical and double-velocity data is

real and somehow this structure has been washed out in the results from both the comparison of double-energy and double-velocity results and the mass-spectrometric measurements.¹⁷ At present, this hypothesis seems more difficult to justify. The comparison of the double-energy and double-velocity results for Cf^{252} (Fig. 8) yielded a ν distribution which reproduced the structure observed in other measurements, and at present there is no reason to believe that the data taken for the U^{238} and Th^{230} reactions should be of a poorer quality than the Cf^{252} data. The mass dispersion²⁰ in the double-energy measurements does not seem large enough to destroy the structure seen in the double-velocity to radiochemical comparison, and the calibration uncertainties¹⁶ for the double-energy measurements should not be of a type that would create or destroy structure. For the mass-spectrometric results,¹⁷ a dip in the ν distribution near mass 130 would imply a charge distribution for the primary fragments which is grossly different from the predictions of the minimum potential energy theory.

Thus, at present it seems that the results for the double-energy to double-velocity comparison are probably most reliable. However, because it is difficult to estimate the reliability of the radiochemical data used in the double-velocity to radiochemical comparison, the possibility that the structure observed in this comparison is real cannot be ruled out. Because similar structure is observed for the two reactions investigated by this method, it seems that if this structure is not real the most reasonable hypothesis is that it is due to systematically low values for the radiochemical measurements in the region of mass 105. The probability of these yields being systematically low is unknown at present.

c. Discussion

The current "fragment-shell" theories⁵⁻⁷ and the two-mode hypothesis both appear to be qualitatively consistent with the general characteristics of the mass and total kinetic-energy distributions that have been observed. However, these two approaches may imply dependences for ν as a function of the fragment mass which are qualitatively different so that from further, more detailed measurements of the properties of the neutron distributions it may be possible to differentiate between these two models.

The fragment-shell theories postulate that the characteristics of the mass and energy distributions of the emitted fragments can be explained as due to an extraordinary rigidity of fragments near the closed-shell configuration $Z=50$ without resorting to the empirical hypothesis of two distinct fission modes. Therefore, whenever one fragment is near the closed shell of 50 protons, fragment yields should be small, the kinetic-energy release should reach a maximum, and the light

¹⁸ R. D. Fink, Ph.D. thesis, Massachusetts Institute of Technology, 1962 (unpublished), and private communications.

¹⁹ L. J. Colby, Jr., M. L. Shoaf, and J. W. Cobble, *Phys. Rev.* **121**, 1415 (1961).

²⁰ The mass resolutions in these experiments can be represented by a Gaussian dispersion function with a full width at half-maximum of ~ 3 mass units for the velocity measurements and ~ 4 mass units for the energy measurements (see Ref. 10).

fragment should get most of the deformation energy. It would therefore be expected that ν would show a minimum near this closed-shell configuration and a maximum for the complementary light fragments. It is expected at higher excitation energies that this structure in the ν distributions will begin to disappear, but for the excitation energies obtained in the present experiment the mass distributions still show predominantly asymmetric fission, and there is still a significant dip in the \bar{E}_K distribution. Therefore, for these moderate excitation energies it should probably be expected that there will still be prominent structure in the ν distribution in the region near the closed-shell $Z=50$ fragment. Thus, structure of the type observed in the double-velocity to radiochemical comparisons seems to be consistent with the expectations of the current fragment-shell theories.

The two-mode hypothesis may lead to a different prediction for the dependence of ν on the mass of the fragment. If there are really two distinct fission modes, then the existence of the asymmetric fission mode is perhaps due to a preference for forming closed-shell configurations as cores for the emitted fragments. On the other hand, the symmetric mode may represent the case where these closed-shell cores are not formed in the descent from the saddle to scission so that fission takes place from symmetric scission-point shapes. One might, therefore, expect that fission from the asymmetric mode would show the general characteristics predicted by the "fragment-shell" theories while symmetric mode fission would more nearly correspond to expectations for fission from a homogeneous charged-liquid drop. If this interpretation of the two modes is correct, the neutron emission from the asymmetric mode would probably show a dependence similar to the sawtooth behavior observed in low-energy fission, and the symmetric mode might give a dependence in which the average number of neutrons was approximately constant or slightly increasing with the fragment mass. The measured neutron distribution would then be a composite of contributions from the two modes. If the ν distributions for the two modes are folded together, it is possible to obtain a composite distribution which is similar to the results shown in Figs. 10 and 11 of the present paper. The depressed yield near mass 80 is interpreted in terms of the rigidity of the $N=50$ closed-shell core. The lack of a depressed yield near the $Z=50$ closed shell ($m_H \sim 130$) can be due to the fact that both modes make significant contributions in this region. In this region the neutron yield from fragments from the asymmetric mode would be depressed, but the neutron yield from symmetric mode would still be high. When the neutron yields from the two modes are then folded together, there is a tendency to wash out any structure in the $m_H \sim 130$ region. Thus, it appears that the results shown in Figs. 10 and 11 may be at least qualitatively consistent with the hypothesis of two distinct fission modes.

It might also be noted at this point that the earlier neck model of fission²¹ may have its justification in terms of the "fragment-shell" concepts. The two-mode version of the neck model would include a symmetric liquid-drop shape for one mode and the conventional asymmetric shape for the other.^{22,23} The "fragment-shell" theories may very well turn out to be discussing the mode which predominates at low excitations of the fissioning nucleus where shell effects should be strongest.

5. SUMMARY

The observed mass and total kinetic-energy distributions appear to be consistent with the hypothesis of two distinct fission modes. The mass distributions can be represented as the sum of a symmetric and an asymmetric component, with the shapes of the mass distributions for each component being approximately independent of the excitation energy of the compound nucleus, and with the relative contribution from the symmetric component increasing rapidly with increasing excitation energy. Furthermore, it was found that both the average total kinetic-energy release and the variance in the average total kinetic-energy release as functions of the fragment mass changed slightly as the excitation energy of the compound nucleus was changed in a manner that was consistent with the two-mode interpretation.

Because of the conflicting nature of the neutron distributions obtained by the two different comparisons, it is not possible to conclude from these results whether or not it is necessary to invoke the empirical hypothesis of two distinct fission modes in order to explain the characteristics of fission at moderate excitation energies. It does seem that at present more weight should be given to the results from the double-energy to double-velocity comparison, and these results appear to require the hypothesis of two fission modes.

In any case the two-mode hypothesis and the current fragment-shell theory, which does not assume two fission modes, appear to have different implications concerning the basic nature of the fission process, but the predictions of these two models for the characteristics of the mass and the total kinetic-energy distributions are qualitatively similar. The area, presently accessible to measurement, where these two models may give qualitatively different predictions is in the variation of the average number of neutrons as a function of the fragment mass. Therefore, more detailed measurements of the properties of prompt neutrons from fission at moderate excitation energies may be of considerable value in the understanding of the basic nature of the fission process.

ACKNOWLEDGMENTS

We gratefully acknowledge many helpful discussions with J. Terrell and R. B. Leachman on the analysis and

²¹ S. L. Whetstone, Jr., Phys. Rev. **114**, 581 (1959).

²² R. B. Leachman, Trans. Am. Nucl. Soc. **5**, 19 (1962).

²³ P. Armbruster and H. Meister, Z. Physik **170**, 274 (1962).

interpretation of these results. We are grateful to W. S. Hall for the preparation of the computer code used in the analysis of these results and to Mrs. Judith C. Gursky for preparing the targets and helping with the data analysis. We would like to thank R. D. Fink for allowing us to use his data before publication.

APPENDIX: CALCULATIONS OF ν_T

It was shown in the text that in order to obtain values for the average number of prompt neutrons ν as a function of fragment mass, the total number of neutrons emitted from both fragments ν_T must be known as a function of mass division. For the cases being studied, there is no detailed experimental information available on ν_T ; therefore, values of ν_T had to be estimated. The values for ν_T were estimated using energy balance considerations in a manner similar to that described previously,^{1,10} except that an attempt was made to take into account the effects of multiple-chance fission.

The calculations proceed in the following manner. First, a total average excitation energy E_T^* was defined as the difference between the total energy release E_R which has been estimated by Milton,²⁴ and the average initial total kinetic-energy release \bar{E}_K which is measured in the present experiment. E_T^* is then composed of three components:

$$E_T^* = E_n^* + E_\gamma + E_{\text{evap}},$$

where E_n^* and E_γ are the average energies available for the emission of prompt neutrons and prompt gamma rays from the fragments; E_γ is assumed to be the same in all cases and is taken equal to 7.5 MeV^{25,26}; and E_{evap} is the average energy dissipated in the emission of evaporation neutrons before fission. The quantity E_{evap} depends on the average number of neutrons ν_{evap} which is a function of fragment mass division because the fragment mass distribution is a function of the excitation energy of the fissioning nucleus.

The quantity E_{evap} was calculated as a function of fragment mass, using the assumption that the mass distribution could be represented as the sum of two components whose shapes were independent of the excitation energy of the fissioning nucleus. Using this assumption, there are now only two quantities to be determined, ν_{evap}^a and ν_{evap}^s , which represent the average number of neutrons evaporated before fission for the asymmetric mode and for the symmetric mode, respectively. Since fission for the compound nuclei discussed here is predominantly asymmetric, ν_{evap}^a can be determined from the values which have been obtained for Γ_n/Γ_γ .^{27,28} The

²⁴ J. C. D. Milton, University of California Radiation Laboratory Report, UCRL-9883 Rev., 1962 (unpublished), and private communication.

²⁵ J. C. D. Milton and J. S. Fraser, Phys. Rev. Letters **7**, 67 (1961).

²⁶ J. C. D. Milton and J. S. Fraser, Phys. Rev. **111**, 877 (1958).

²⁷ R. Vandenbosch and J. R. Huizenga, Proc. U. N. Intern. Conf. Peaceful Uses At. Energy, 2nd Geneva, 1958 **15**, 284 (1958).

²⁸ R. Vandenbosch, H. Warhanek, and J. R. Huizenga, Phys. Rev. **124**, 846 (1961).

TABLE II. Results of neutron calculations.^a

Reaction	E_{He^4}	E_n	E^*	$\bar{\nu}_T$	ν_{evap}^a	ν_{total}	Measured ^b ν_{total}
$\text{He}^4 + \text{Th}^{230}$	29.5	17.2	24.1	4.70	0.65	5.35	5.14
	25.7	13.5	20.4	4.28	0.38	4.66	4.55
$\text{He}^4 + \text{Th}^{232}$	29.5	17.8	24.3	4.37	0.79	5.16	5.15
	25.7	14.1	20.6	3.62	0.79	4.41	4.56 ± 0.08
	22.1	10.5	17.0	3.42	0.43	3.85	4.03
$\text{He}^4 + \text{U}^{238}$	29.5	17.5	23.4	5.70	0.15	5.85	
	25.7	13.8	19.7	5.05	0.15	5.20	
	22.1	11.2	16.1	4.36	0.15	4.51	
Cf^{252}				3.78		3.78	3.77

^a E_{He^4} , E_n , and E^* are the He^4 laboratory bombarding energy, the equivalent neutron bombarding energy for the reaction producing the same compound nucleus, and the compound nucleus excitation energy, respectively. $\bar{\nu}_T$ is the average number of prompt neutrons emitted from the fragments per fission. ν_{evap}^a is the average number of neutrons evaporated from the compound nucleus before fission for fission from the asymmetric mode. ν_{total} is the average total number of neutrons emitted per fission.

^b From Diven and Hopkins (Ref. 30). These values are based on the measurement of ν_{total} for 14.5-MeV neutron-induced fission of U^{235} and the observed slope of 0.16 neutrons/MeV. The $\text{He}^4 + \text{Th}^{230}$ values are obtained from the $n + \text{U}^{235}$ results, with a shift in the energy scale of 0.7 MeV.

probabilities for first-, second-, and third-chance fission were estimated in the manner described by Vandenbosch, Warhanek, and Huizenga,²⁸ and these were used to determine ν_{evap}^a . The probabilities for multiple-chance fission for the symmetric mode were then determined using the excitation energy dependence for the ratio of symmetric to asymmetric yields Y_s/Y_a determined from the radiochemical data of Hicks *et al.*³ for He^4 -induced fission of Th^{232} . In the determination of Y_s/Y_a it was assumed that this ratio was equivalent to the measured ratio $\text{Cd}^{115}/\text{Mo}^{99}$; the experimental data were corrected to give the ratio $\text{Cd}^{115}/\text{Mo}^{99}$ for first-chance fission, using the method described by Vandenbosch, Warhanek, and Huizenga.²⁸ It was further assumed that for the other reactions studied the rate of change of Y_s/Y_a with excitation energy was the same as that for the $\text{He}^4 + \text{Th}^{232}$ reaction.

The values obtained for ν_{evap}^a are shown in Table II for the reactions studied. In all cases the values obtained for ν_{evap}^s were less than 0.1 (i.e., greater than 90% of the symmetric mode fission corresponded to first-chance fission). From the values obtained for ν_{evap}^s and ν_{evap}^a , values were obtained for E_{evap}^s and E_{evap}^a , using the neutron binding energies of Cameron²⁹ and assuming that the average kinetic energy of the neutrons was 1.2 MeV. Then, E_{evap} can be determined as a function of the fragment mass, using the values for the relative yield from the two modes at each mass number.

Using the values calculated for E_{evap} , E_n^* can be determined as a function of the fragment mass. Then, the total number of prompt neutrons ν_T emitted from the fragments was assumed to be given by

$$\nu_T = E_n^*/(B_n + 1.2),$$

where B_n was the average binding energy for the two

²⁹ A. G. W. Cameron, Chalk River Report CRP-690, AECL-433, 1957 (unpublished).

fragments which was determined from the tabulations of Milton.²⁴

When this method was used to evaluate ν_T for Cf^{252} fission, it was found that the value obtained for $\bar{\nu}_T$ (the weighted average of ν_T over the mass distribution) was 3.60, which is comparable to the accepted value of 3.77.³⁰ This difference corresponds to the average value of E_n^* being 1.5 MeV too low, which is within the uncertainties of the energy calibration procedure. To correct for this, all of the values for E_n^* were increased

³⁰ B. C. Diven and J. C. Hopkins, Nucl. Phys. (to be published).

by 1.5 MeV. In order to compare with experimental values, $\bar{\nu}_T$ was calculated for each case, and the results are shown in Table II. Also shown in Table II are values for $\nu_{\text{total}} = \bar{\nu}_T + \nu_{\text{evap}}^a$, which corresponds to average number of neutrons emitted per fission. Direct measurements have been made by Diven and Hopkins³⁰ of ν_{total} as a function of energy for neutron bombardment of U^{233} and U^{235} . Values for ν_{total} deduced from these measurements for the excitation energies of the present experiment are also shown in Table II. It is seen that the calculated values for ν_{total} agree very well with the measured values.

Coincident Time-of-Flight Measurements of the Velocities of Fission Fragments from Charged-Particle-Induced Fission*

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Measurements have been made of the velocities of the coincident fission fragment pairs emitted in the 25.7- and 29.5-MeV He^4 -induced fission of U^{233} and Th^{230} , the 25.7- and 29.5-MeV He^4 -induced fission of Th^{232} , and the 12.0- and 14.0-MeV H^2 -induced fission of Th^{230} . Previously unpublished data for the 21.8-MeV He^4 -induced fission of U^{233} and Th^{232} are also included. A decrease in average total fragment kinetic energy for symmetric mass division is observed in each case. The relatively large yield of symmetric mass divisions in this work permits a reliable measurement of this effect. The distributions of fragment velocities, masses, and kinetic energies are consistent with the hypothesis of two competing modes of fission. A comparison is made of the primary mass yields obtained in the present work with the yields obtained by radiochemical methods to determine the dependence of prompt neutron emission on fragment mass. The neutron emission found, although subject to poorly known and possibly large uncertainties in the radiochemical data, appears to show a rather pronounced structure of just such a nature as might be attributed to fragment-shell effects that persist to these high excitation energies. Such a structure is not observed, however, when the present primary mass data are compared with mass distributions derived from recent fragment double-energy measurements. The time-of-flight mass distributions were corrected for the influence of the pre-fission neutron emission and the detection solid angle. As a part of the correction, the dependences of the yield of symmetric mass divisions on the excitation energy of the fissioning nuclei were estimated for each of the target-projectile systems.

INTRODUCTION

WHEN the measurements reported here were begun, there was experimental evidence^{1,2} that the average amount of kinetic energy given to fission fragments was not always a maximum for the most symmetric mass divisions as is expected from the simplest theory. Apparently, there was an abrupt and large decrease in the kinetic-energy release for somewhat more symmetric mass divisions than those observed to be most probable. Since data were not available, however, for cases where the relative yield of symmetric mass divisions was appreciable, the experi-

mental results were uncertain, chiefly because of the difficulty in making reliable corrections under these conditions for the dispersion of the measurements. Preliminary time-of-flight measurements³ of charged-particle-induced fission, which give relatively large yields of symmetric mass divisions, showed that the kinetic-energy release is indeed decreased by as much as 10 MeV over a large interval of fragment-mass ratios near unity for fission at moderate excitation energy.

Recently, double-energy measurements were made with semiconductor detectors⁴ of the fission of nuclides of lower fissionability where equally probable symmetric and asymmetric mass division and even predominantly symmetric mass divisions are observed. These measurements permitted a fairly precise analysis⁴

* Work performed under the auspices of the U. S. Atomic Energy Commission.

¹ I. Halpern, Ann. Rev. Nucl. Sci 9, 245 (1959).

² E. K. Hyde, University of California Radiation Laboratory Report UCRL-9036, 1960, and UCRL-9036, revised 1962 (unpublished).

³ S. L. Whetstone and R. B. Leachman, Bull. Am. Phys. Soc. 6, 376 (1961).

⁴ H. C. Britt, H. E. Wegner, and J. C. Gursky, Phys. Rev. 129, 2239 (1963).