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EMISSION PROBABILITIES OF PROMPT NEUTRONS FROM SPONTANEOUS AND NEUTRON-INDUCED FISSION

by

R. B. Leachman





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ABSTRACT

A method is developed for using the relatively easily measured fission parameters, together with the mass equation of fission and the evaporation model of the nucleus, to determine the emission probabilities of the fission neutrons. The distribution of the kinetic energies of the fragment pairs enters into these calculations in a sensitive manner. Neutron emission probabilities are computed for the fission of the compound nuclei U^{234} , U^{236} , and Pu^{240} , for which cases reasonably adequate data on the fragment pair energies are available. Although the corresponding data for the fission of the compound nuclei Th^{233} , U^{238} , and U^{239} are considerably poorer, neutron emission probabilities are also computed.

The calculated results from this method are in good agreement with direct measurements of fission neutrons.

ACKNOWLEDGMENTS

The author wishes to acknowledge the suggestions made by C. D. Coryell on the mass surface, V. F. Weisskopf on the neutron temperatures, H. H. Barschall and R. G. Thomas on the evaporation model, and J. A. Wheeler on the solution of the integral equations. Also, the considerable work done by M. T. Goldstein and I. J. Cherry on the calculations is appreciated.



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INTRODUCTION

The emission probabilities of fission neutrons are of interest in both reactor and weapons work. Experimental determinations of even the average number of neutrons $\overline{\nu}$ emitted per fission have been handicapped by the variation in the detection efficiencies of neutron detectors with neutron energy and the difficulty of determining these efficiencies. Also, because of the low detection efficiencies, it has been particularly difficult to make absolute measurements of the variation in the number of fission neutrons emitted in each fission event. Until the recent development of the large volume scintillation tanks containing cadmium, the neutron detectors used for fission neutrons had low efficiencies, usually of only a few percent. Such low detection efficiency made multiplicity measurements of fission neutron emission particularly difficult.

As a result of these experimental difficulties there have existed large uncertainties in $\overline{\nu}$, especially for fission induced by neutrons in the vicinity of 1 to 14 Mev, in which region the detector must be shielded from the neutrons inducing fission. The present calculations are an effort to use the more easily measured parameters of fission to calculate neutron emission probabilities for several fissile nuclides at several excitation energies inducing fission. These more easily measured parameters are the masses of the fission products as obtained from mass spectrographic measurements and the energy surface from energy systematics, together with the measured kinetic energy of the fission products. These quantities are used in the mass equation of fission to determine the excitation energy of the fission products. This excitation energy is used with the evaporation model of the nucleus to compute neutron emission probabilities of fission.

In the following analysis these quantities will all be used in their probability distributions and all distributions will be normalized to unity. In the initial analyses that were made, the calculations were by graphic means and by hand calculations. However, the bulk of the data that have been compiled in this report is the result of rather extensive calculations on the IBM 701.

BASIC ASSUMPTIONS

Three basic assumptions are used in the present method. These assumptions are: (a) The basic determination of the kinetic energy of the fission products is the Coulomb

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energy of repulsion of the nuclear charges of the two fission products from binary fission.

(b) A semi-empirical mass surface, including the effects of shell edges, can be used to obtain masses of atoms for which mass spectrographic measurements have not been made and also can be used to obtain neutron binding energies for nuclei for which the binding energies have not been measured.

(c) A simple form of the evaporation model of the nucleus can be used to calculate the emission probabilities of neutrons for the primary fission products as a function of the excitation energy of these nuclides.

Although complete investigations have not been made of these three assumptions, particularly of (b) and (c), the available data do indicate that all three are reasonable assumptions.

In particular, the Coulomb energy assumption in (a) has been confirmed by the kinetic energy measurements of fission products for rather wide variations of excitation inducing fission. Both Wahl¹ and Friedland² have measured the kinetic energy of the fission products of 14-Mev neutron induced fission of U²³⁵ and have found essentially the same average kinetic as for thermal-neutron induced fission. In addition, Wahl has found the same condition existing for high- and low-energy neutron induced fission of both Pu²³⁹ and U²³⁸. Similarly, Segre and Weigand³ have found the same kinetic energy of the fission products for spontaneous fission of Pu²⁴⁰ and for thermal-neutron induced fission of Pu²³⁹. An independent confirmation of the Coulomb origin of the kinetic energy of the fission products is provided by the analysis of this energy as a function of the mass ratio of the fission products by Fong.⁴ Fong has found that, except for an anomalous effect in the region of symmetrical fission, the average kinetic energy as a function of mass ratio is of Coulomb origin.

It should be pointed out that the Coulomb origin of the fission energy can explain the variation in the kinetic energy of the fission products for a particular mass ratio only if some assumptions of variable breaking distances for fissioning nuclei are made. However, such assumptions do not invalidate the analysis being made.

The semi-empirical mass surface has been discussed by many authors and has received a thorough analysis by Coryell.⁵ Coryell has shown the effect of closed nuclear shells on

- ¹J. S. Wahl, Phys. Rev. 95, 126 (1954).
- ²S. S. Friedland, Phys. Rev. 84, 75 (1951).
- ³E. Segrè and C. Weigand, Phys. Rev. 94, 157 (1954).
- ⁴ p. Fong, A Theory of Fission (unpublished).
- ⁵C. D. Coryell, Ann. Rev. Nuclear Sci. 2, 305 (1953).

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various parameters in the expression for this mass surface and has demonstrated that the parabolic shape applies for given values of mass number A over the range of nuclear charge Z that has been measured. Unfortunately, the shape of the mass surface has not been measured as far from the stable nuclear charge as the neutron-rich primary fission products are found.

The validity of the semi-empirical mass surface to compute neutron binding energies has been demonstrated⁶ by comparing these computed energies with measured binding energies. Although a mass surface that did not include the effects of closed nuclear shells was used, the results of these analyses showed that satisfactory agreement between the measured and calculated binding energies would be obtained if the effects of nuclear shells were included. However, once again, these comparisons were for nuclides considerably nearer stable Z than are the primary fission products.

The evaporation model of the nucleus as described by Blatt and Weisskopf⁷ has been tested by comparing results from it with experimental results of particle emission in several types of experiments. Although this model provides a convenient picture of particle emission, Cohen,⁸ in a summary of the comparisons with experimental results, has shown that the model has limited accuracy. Cohen has shown the need for a nuclear temperature dependence on the amount of the excitation and the method of formation of the compound nucleus. Again, the experimental tests of this model have been made only with nuclides much nearer stable Z than are the primary fission products.

MASS EQUATION OF FISSION

The basic mass equation of fission for U^{236} is

$$M(U^{236}) + E_b = M(A^L, \delta^L, Z^L) + M(A^H, \delta^H, Z^H) + E_C + E_X.$$
 (1)

This equation is written in units of Mev where the conversion between the masses M and energy is by the factor 931.15 Mev/mass unit. Since the equation is for the primary fission products before neutron emission, $A^H + A^L = 236$ and $Z^L + Z^H = 92$ are required for this case of U²³⁶ fission to conserve nucleon number and charge number. Here and throughout

⁸B. L. Cohen, Phys. Rev. <u>92</u>, 1245 (1953).

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⁶J. A. Harvey, Phys. Rev. <u>81</u>, 353 (1951) and Sher, Halpern, and Mann, Phys. Rev. <u>84</u>, 387 1951).

⁷J. M. Blatt and V. F. Weisskopf, <u>Theoretical Nuclear Physics</u>, John Wiley and Sons, New York, 1952.



this report, the superscripts L and H refer to the light and heavy fragments, respectively. In Eq. 1, δ is the even-odd parameter discussed by Coryell.⁵ The masses of the fission products are the ground state masses of these unstable fission products. Furthermore, E_C is the Coulomb energy of the fission products; E_X is the sum of the excitation energies of these primary fission products; and E_b is the bias energy inducing fission with reference to spontaneous fission, where $E_b = E_n + B_n$.⁹ The binding energy B_n is that of a neutron to the nucleus undergoing fission and E_n is the energy of the neutron inducing fission.

The masses in this mass equation of fission are based on masses from mass spectrographic data. These mass spectrographic measurements are usually of about 1-Mev uncertainty and the masses in the fission product region are accurate relative to the masses of the fissioning nuclei because of the method of comparison with the lighter elements used in the mass measurements of the heavy, fissionable nuclei. The masses of the fissionable nuclei are from the compilation of Huizenga and Magnusson.¹⁰ The masses of the stable atoms in the region of the fission products were obtained from the mass measurements of Duckworth et al.¹¹ and of the Minnesota group.¹² These mass measurements are shown in Fig. 1, after having been converted to masses of odd A and to the non-integer stable nuclear charge for that mass number. The conversion to odd A is by the δ values of Fermi.¹³ The conversion to the stable nuclear charge is by the stable charge values and parabolic surface constants of Coryell.⁵ The conversion from these masses for the valley of the mass surface to the masses of the primary fission products was made by the same conversion parameters.

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⁹J. L. Fowler (Oak Ridge National Laboratory Report ORNL-1374, December 1952) first pointed out that the Coulomb energy of fission is constant, and thus the excitation energy of the fragments increases as the energy of the neutron inducing fission.

¹⁰J. R. Huizenga and L. B. Magnusson, Argonne National Laboratory Report ANL-5158, November 1953.

¹¹E. M. Pennington and H. E. Duckworth, Can. J. Phys. <u>32</u>, 808 (1954); B. J. Hogg and H. E. Duckworth, Can. J. Phys. <u>32</u>, 65 (1954) and <u>31</u>, 942 (1953); H. E. Duckworth and R. S. Preston, Phys. Rev. <u>82</u>, 468 (1951) and <u>79</u>, 402 (1950); Duckworth, Woodcock, and Preston, Phys. Rev. <u>78</u>, 479 (1950); Duckworth, Preston, and Woodcock, Phys. Rev. <u>79</u>, 188 (1950); Duckworth, Kegley, Olson and Stanford, Phys. Rev. <u>83</u>, 1114 (1951); C. L. Kegley and H. E. Duckworth, Phys. Rev. <u>83</u>, 229 (1951); and B. J. Hogg and H. E. Duckworth, Can. J. Phys. <u>30</u>, 637 (1952).

¹²Collins, Nier, and W. H. Johnson, Jr., Phys. Rev. <u>86</u>, 408 (1952); Collins, Johnson, Jr., and Nier, Phys. Rev. <u>94</u>, 398 (1954); and R. E. Halsted, Phys. Rev. <u>88</u>, 666 (1952).

¹³E. Fermi, <u>Nuclear Physics</u>, notes compiled by J. Orear, A. H. Rosenfeld, and R. A. Schluter, University of Chicago Press, Chicago, 1949.



Fig. 1. Masses of the valley of the semi-empirical mass surface. Data are mass spectrographic data converted to odd A and to the non-integer charge Z that, for each value of A, corresponds to the valley of the mass surface.



DISTRIBUTION IN FISSION FRAGMENT MASS

It has been shown¹⁴ that for fission induced by excitations of less than about 40 Mev, definite peaks exist in the mass distribution of the fission yield of the various fissioning nuclides that are all characterized by a low probability of fission for symmetrical fission. As is readily seen in Eq. 1, the sum of the fission energies will be dependent upon the masses of the fission products. This is emphasized by rewriting Eq. 1 as

$$\mathbf{E}_{\mathbf{C}} + \mathbf{E}_{\mathbf{X}} = \phi(\delta, \mathbf{R}_{\mathbf{m}}, \mathbf{R}_{\mathbf{Z}}).$$
(2)

Further, the yield for symmetrical fission is found to increase as the energy inducing fission E_b increases.¹⁵ However, in all cases being investigated, the yield in the symmetrical fission region is sufficiently low that this change of yield may be neglected.

In the analysis for each nucleus undergoing fission, the mass yield is divided into three groups of mass ratio R_m . In each case the division of the mass yield is chosen such that one division is at the 82-neutron shell. The spike in the mass yield curve at this shell as found by mass spectrographic means has not been included, but instead the smooth mass yield curve is used. The neutron emission characteristics of each group of fission masses for each fissioning nuclei are investigated separately and then combined to determine the neutron emission characteristic of that fissioning nucleus.

DISTRIBUTION IN NUCLEAR CHARGE

The nuclear charge is seen by Eqs. 1 and 2 to be a parameter in the determination of the energies of fission. Fortunately for simplicity of calculation, the distribution in the nuclear charge for each mass ratio of fission is sharp, and little error is induced by using only the most probable nuclear charge.

Glendenin¹⁶ has investigated the nuclear charge of the fission products after neutron emission as a function of the stable nuclear charge of that mass of fission product. More recently Pappas¹⁷ has included the effect of nuclear shells on the line of nuclear charge

¹⁶Reference 14, Paper 52.

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¹⁴C. D. Coryell and N. Sugarman, eds., <u>Radiochemical Studies: The Fission Products</u>, Appendix B, Div. IV, Vol. 9, National Nuclear Energy Series, McGraw-Hill Book Company, Inc., New York, 1951.

¹⁵R. A. Schmitt and N. Sugarman, Phys. Rev. <u>95</u>, 1260 (1954).

¹⁷A. C. Pappas, Massachusetts Institute of Technology, Laboratory for Nuclear Science Technical Report No. 63, 1953.



stability in such an analysis of the charge of the fission products. It is seen from such analyses that the distribution in the nuclear charge is about two charge units wide at half maximum.

In Fig. 2 is shown the variation of the fission energies as a function of the nuclear charge for fission of U^{236} into the most probable fission mass ratio. It is seen from this figure that the roughly 4-Mev width of the excitation energy distribution caused by the charge distribution is small compared to the roughly 15-Mev widths of the excitation energy distribution sto be used. Consequently, only one nuclear charge, the most probable nuclear charge, is used in the calculations for each mass ratio of fission.

DISTRIBUTION IN KINETIC ENERGY

The distributions $I(E_I)$ in the measured total kinetic energy of fission, E_I , are obtained from the double ionization chamber measurements of the kinetic energy made by Brunton and Hanna¹⁸ and Brunton and Thompson.¹⁹ Through the use of the momentum equation which applies for binary fission, these measurements of the kinetic energy of the fission pairs by the ionization are expressed in terms of the probability as a function of the mass ratio R_m of fission. Thus, it is possible to determine the distribution in the total kinetic energy of the fragment pairs for each mass ratio being considered.

A comparison²⁰ of the mass distribution obtained in a similar manner with the mass distribution obtained by radiochemical means¹⁴ has demonstrated the existence of a dispersion in the measurement of energy by ionization of approximately 8-Mev full width at half maximum for each fragment and thus of approximately 11.4-Mev full width at half maximum for the total kinetic energy of the fragment pairs. Similarly, a comparison²¹ of the directly measured velocities of fission fragments with the velocities inferred from the ionization chamber measurements demonstrated a dispersion of approximately 9 Mev for each fragment. Further, this comparison of velocities showed that the ionization chamber measurements of the kinetic energies I_{I}^{L} and E_{I}^{H} are low by 5.7 and 6.7 Mev, respectively. The energy that is not observed as ionization has been explained²² in terms of the ionization defects of fission fragments.

²²J. K. Knipp and R. C. Ling, Phys. Rev. <u>82</u>, 30 (1951).



¹⁸D. C. Brunton and G. C. Hanna, Can. J. Research <u>A28</u>, 190 (1950).

¹⁹D. C. Brunton and W. B. Thompson, Can. J. Research <u>A28</u>, 498 (1950).

²⁰R. B. Leachman, Phys. Rev. <u>83</u>, 17 (1951).

²¹R. B. Leachman, Phys. Rev. <u>87</u>, 444 (1952).



Fig. 2. Fission energy dependence upon nuclear charge ratio for the most probable mass ratio of U^{236} fission. The Coulomb energy is normalized to the most probable measured kinetic energy (corrected for ionization defect) at the most probable charge ratio. The variation with nuclear charge is calculated on the basis of a Coulomb origin. The sum $E_C + E_X$ is calculated from Eq. 1. The nuclear charge distribution is from Glendenin. 16



In Fig. 3 is shown a typical distribution $I(E_I, R_m)$ as obtained by Brunton and Hanna¹⁸ for fission of the compound nucleus U^{236} at the most probable mass ratio.

This distribution contains the distribution in the ratio of the nuclear charge R_Z . For a rigorous treatment of the emission probabilities a distribution of energies as a function also of the ratio of nuclear charge R_Z is needed. However, calculations from the data in Fig. 2 show the distribution in the kinetic energy is changed only by a very small amount by the effects of the distribution in the nuclear charge.

To convert the measured distributions $I(E_1, R_m)$ to the distribution in the true kinetic energy of fission $C(E_C, R_m)$, it is necessary to unfold the effects of the dispersion and transpose by the amount of the ionization defects in the ionization chamber measurements of energy to obtain this true, or Coulomb, energy of fission. Unfortunately, the comparisons^{20,21} of fission parameters are not sufficiently sensitive to determine both the breadth and the shape of the dispersion functions that lead to the experimental data. However, these analyses do indicate that a Gaussian shaped dispersion does satisfactorily fit the data. For ease of calculations, Gaussian dispersions are used in the present analysis.

The expression for the dispersion in the data obtained from ionization chamber measurements is

$$D_1(E_I, E_C) \propto \exp\left[-\left(\frac{E_I + 12.4 - E_C}{u_1}\right)^2\right].$$
 (3)

The 12.4-Mev constant in the exponential is the ionization defect term and u_1 is the dispersion width. It is seen from Fig. 2 that the dispersion introduced by the distribution in the nuclear charge is of approximately Gaussian shape and thus this dispersion similarly is expressed as a Gaussian in

$$D_2(E_1, E_C) \propto \exp\left[-\left(\frac{E_1 + 12.4 - E_C}{u_2}\right)^2\right].$$
 (4)

The term u_2 in the exponential is the width of this dispersion. It can readily be shown that the combined effect of these two dispersions is itself a Gaussian dispersion. Equation 5,

$$D(E_{I}, E_{C}) \propto \exp\left[-\left(\frac{E_{I} + 12.4 - E_{C}}{u}\right)^{2}\right], \qquad (5)$$

is the expression for this combined dispersion, where u is the combined width. In the bulk of the calculations, 7.2 Mev is taken as the value for u. This is obtained by weighting the

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Fig. 3. Ionization energy distribution from Brunton and Hanna.¹⁸ The experimental data are corrected for only the instrumental errors listed by the authors.



dispersion obtained from the comparison of mass distributions somewhat more than the dispersion obtained from the comparison of the velocities.

DISTRIBUTION IN COULOMB ENERGY

In keeping with the assumption of a Coulomb origin of the kinetic energy of fission, the energy distribution $C(E_C, R_m)$ is considered independent of the even-odd term δ . In this way the even-odd term has an effect in the excitation energy of the fission products E_X .

The distribution of the energy of fission from ionization chambers is converted into the kinetic energy distribution by the convolution

$$I(E_{I}, R_{m}) = \int_{-\infty}^{\infty} dE_{C} C(E_{C}, R_{m}) D(E_{I}, E_{C}) .$$
(6)

Such an integral equation with the empirical data of $I(E_I, R_m)$ as the solution is difficult to solve exactly. For this reason, an approximate method of solution contained in the Appendix is used throughout this paper. This approximate method involves fitting the empirical $I(E_I, R_m)$ distribution with a series of Gaussian functions. In this manner, all the expressions used in the analysis are Gaussian and thus the solutions of the integral equation 6 and in the integral equation to follow are facilitated.

DISTRIBUTION IN EXCITATION ENERGY

The distribution $X(E_b, E_X, \delta, R_m)$ in the total excitation energy of fission is obtained by the use of Eq. 1 or 2. This total excitation energy is the excitation energy shared by the fragment pairs of binary fission. However, it is necessary to obtain the excitation energy distributions of the individual fragments X^L and X^H to determine the neutron emission probabilities of fission that are needed.

There exists little experimental evidence on which to decide how the excitation energy E_X is divided into the excitation energies E_X^L and E_X^H of the individual fission fragments. Consequently, a simple division of excitation energy is used, such that the excitation energies of the individual fragments X^L and X^H are identical distributions of parameters. These parameters are constrained to cause the sum of the excitation energies of the individual fragment ζ is used, $X^L(\zeta) = X^H(\zeta)$. Under such conditions, the combination of these individual excitation energies into the total excitation energy distribution is given by







$$X(E_{b}, E_{X}, \delta, R_{m}) = \int_{-\infty}^{\infty} dE_{X} \int_{-\infty}^{\infty} dE_{X}^{H} X^{L}(E_{b}, E_{X}^{L}, \delta^{L}, R_{m}) X^{H}(E_{b}, E_{X}^{H}, \delta^{H}, R_{m}).$$
(7)

The solution of Eq. 7 involves the use of the constraining equation $E_X = E_X^L + E_X^H$ to yield

$$X(E_{b}, E_{X}, \delta, R_{m}) = \int_{-\infty}^{\infty} dE_{X}^{L} X^{L}(E_{b}, E_{X}^{L}, \delta^{L}, R_{m}) X^{H}(E_{b}, E_{X} - E_{X}^{L}, \delta^{H}, R_{m}), \qquad (8)$$

which is solved by the method in the Appendix.

In Fig. 4 is a typical excitation energy distribution of the individual fragments. It is to be noted that the approximate method of solution leads to impossible negative probabilities of $X(E_X, R_m, E_b, \delta)$ and to impossible negative excitation energies. However, these negative probabilities and energies are preserved throughout the analysis since they do, in a mathematical sense, have meaning.

EVAPORATION MODEL OF THE NUCLEUS

The evaporation model of the nucleus as formulated by Blatt and Weisskopf⁷ involves the use of the square root of the nuclear excitation energy in the exponent in the expression for the neutron emission as a function of the excitation energy. This relation is derived from thermodynamic considerations. However, Blatt and Weisskopf also give the simpler form

$$n(\epsilon)d\epsilon \propto \epsilon \exp\left(-\epsilon/T\right)d\epsilon, \qquad (9)$$

which is shown by Cohen⁸ to fit the experimental data satisfactorily. Here ϵ is the neutron energy in Mev and $n(\epsilon)$ is the probability of emission of a neutron with energy ϵ . This simpler form in Eq. 9 is easier to use in the computations of the emission probabilities N^L and N^H of neutrons. These expressions for the emission probabilities, where μ is a summation parameter, are

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Fig. 4. Excitation energy and neutron emission probabilities calculated for U^{236} fission into the most probable mass ratio. The bias energy E_b is zero in the $X^{L, H}$ case plotted.



$$-\left[\exp\left(-\frac{\mathbf{E}_{\mathbf{X}}^{\mathbf{L}}-\mathbf{B}_{\nu}^{\mathbf{L}}}{\mathbf{T}}\right)\right]\left[\begin{array}{c}2\nu-3\\1+\sum\limits_{\mu=1}\left(\frac{\mathbf{E}_{\mathbf{X}}^{\mathbf{L}}-\mathbf{B}_{\nu}^{\mathbf{L}}}{\frac{\mathbf{T}}{\mu!}}\right)^{\mu}\end{array}\right]\right\}$$
for $\mathbf{E}_{\mathbf{X}}^{\mathbf{L}} \ge \mathbf{B}_{\nu+1}^{\mathbf{L}}$ and $\nu > 1$.

Similar expressions with H superscripts apply for the heavy fragments. Equations 10 are based on Eq. 9 and the assumption that a neutron is always emitted when it is energetically possible.

As has been shown by Cohen, the temperature T appropriate for Eq. 9 is dependent upon the method of excitation of the compound nucleus. It has been suggested by Weisskopf²³ that $\overline{^{23}}$ V. F. Weisskopf, private communication, 1952.

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the temperatures derived by fitting the (n, 2n) cross section data²⁴ with Eq. 10 for nuclides in this mass region are the most appropriate for the use of Eq. 10 in the present calculations. This use of temperatures from (n, 2n) data does not involve the uncertainties of the Coulomb barrier in the formation of the compound nucleus. Furthermore, these are temperatures that are derived for excitations of approximately the same magnitude as the most probable excitation of the fission products after fission. The temperatures derived by fitting these (n, 2n)data by Eq. 10 are given in Fig. 5. As mentioned before, a question exists about the variation of the nuclear temperature with the neutron-proton ratio for nuclides of a particular mass. However, since the neutron-proton ratio varies only slightly from stable nuclides to those of the neutron-rich fission products. Accordingly, T = 1.4 Mev is used for both the heavy and light fragments for the calculations in this paper.

The binding energies B^L and B^H required in Eq. 10 are computed from the semiempirical mass surface discussed on page 8. As an illustration of the resulting binding energies, Table I contains those for the most probable mass ratio of the fission yield for fission of U^{236} . Although the binding energies of the light fragments are slightly larger than those of the heavy fragments in Table I, the opposite is true for the other two mass ratios used. Also, as discussed above, these binding energies are computed from the part of the semi-empirical mass surface that is three or four charge units from the region of stability and in a region in which few, if any, measurements have been made to check its validity.

TABLE I. NEUTRON BINDING ENERGIES CALCULATED FROM THE SEMI-EMPIRICAL MASS SURFACE

Calculations are for the fission products of U^{236} with a mass ratio $R_m = 141/95$ and for the most probable (non-integer) nuclear charge. The even-odd term δ_2 represents a primary fission product with an even number of neutrons; δ_1 represents an odd number of neutrons.

	$\mathbf{B}_{\nu}^{\mathbf{L}}(\delta_{1})$	$\mathbf{B}_{\nu}^{\mathbf{L}}(\boldsymbol{\delta}_{2})$	$\mathbf{B}_{\nu}^{\mathbf{H}}(\boldsymbol{\delta}_{1})$	$\mathbf{B}_{\nu}^{\mathbf{H}}(\boldsymbol{\delta}_{2})$
ν	(Mev)	(Mev)	(Mev)	(Mev)
1	4.17	5.71	4.05	5.63
2	10.26	10.26	9.93	9.93
3	15.02	16.56	14.30	15.88
4	21.60	21.60	20.63	20.63
5	26.91	28.45	25.46	27.04

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 ²⁴H. C. Martin and R. F. Taschek, Phys. Rev. <u>89</u>, 1302 (1953); H. C. Martin and B. C. Diven, Phys. Rev. <u>86</u>, 565 (1952); and Brolley, Fowler, and Schlacks, Phys. Rev. 88, 618 (1952).

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Fig. 5. Temperatures used to fit (n, 2n) data²⁴ by the evaporation model of neutron emission given in Eqs. 9 and 10.

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In Fig. 4 are the N_{ν}^{L} and N_{ν}^{H} distributions from Eq. 10, using the binding energies in Table I, and T = 1.4 Mev. It is to be noted in Table I and in Fig. 4 that both the neutron binding energies and the excitation energy of fission are dependent upon the even-odd character of the fission products.

NEUTRON EMISSION PROBABILITIES

The neutron emission probabilities as a function of the mass ratio R_m and even-odd parameter δ are calculated by the use of the integral

$$P_{\nu}^{L}(E_{b},\delta^{L},\nu^{L},R_{m}) = \int_{-\infty}^{\infty} X^{L}(E_{b},E_{X}^{L},\delta^{L},R_{m}) N_{\nu}^{L}(E_{X}^{L},\delta^{L},\nu^{L},R_{m}) dE_{X}^{L}.$$
(11)

A similar equation for H applies. In practice, Eq. 11 was solved by an IBM 701 by summation over sufficiently wide finite limits, rather than the infinite limits of Eq. 11. The emission probabilities for each value of ν^{L} and ν^{H} are then combined for all even-odd cases with proper weight for the abundance of each case of δ . Thus, the neutron emission probabilities independent of δ are obtained. These are written as $P_{\nu}^{L}(E_{b}, \nu^{L}, R_{m})$. A similar expression is used for H.

These emission probabilities are for the individual fragments but the needs are for the combined emission probabilities $P_{\nu}(E_{b}, \nu, R_{m})$ from both fragments of a fission pair. There-fore,

$$P_{\nu}(E_{b},\nu,R_{m}) = \sum_{\eta=0}^{\nu} P_{\eta}^{L}(E_{b},\nu^{L},R_{m}) P_{\nu-\eta}^{H}(E_{b},\nu^{H},R_{m})$$
(12)

is used to form these combined emission probabilities. In Eq. 12, η is a summation parameter analogous to ν . Finally, the parameter R_m is removed by combining the emission probabilities with the appropriate weight of the fission probabilities into each region of mass ratio R_m . The resulting fission neutron emission probabilities are given in Tables II and III and are discussed further in the following sections.

RESULTS

The use of the equations discussed above requires data of the fission kinetic energy E_C as a function of the mass ratio of fission. Such data are available^{18,19} from double ionization chamber measurements of fission of the compound nuclei U²³⁴, U²³⁶, and Pu²⁴⁰ and have



TABLE II. CALCULATED EMISSION PROBABILITIES OF NEUTRONS FROM THE COMPOUND NUCLEI UNDERGOING FISSION

The results are approximately normalized to the BNL-250 thermal neutron values $\vec{\nu}(U^{234}) = 2.54 \pm 0.04$, $\vec{\nu}(U^{236}) = 2.46 \pm 0.03$, and $\vec{\nu}(Pu^{240}) = 2.88 \pm 0.04$ by subtracting 5.44 Mev, 2.29 Mev, and 4.78 Mev from the calculated excitation energies of U^{234} , U^{236} , and Pu^{240} . respectively. The effects of (n, n'f) have not been included.

Compound	Energy of Neutron Introducing											
Nucleus	Fission (Mev)	$\overline{oldsymbol{ u}}$	P ₀	P ₁	Р ₂	Р ₃	P4	Р ₅	P ₆	P ₇	P.8	P ₉
994	((Spontaneous))										-	-
U ²³⁴	-6.74	1.669	.1279	.3192	3522	1498	0523	0046	0025	0010	0005	
	`0 '	2.528	.0158	.1295	.3660	3158	1354	0405	0035	0010	0005	.0000
	0.70	2.615	.0121	.1115	3536	3296	1490	0457	.0022	0037	0013	0004
	7.00	3,365	.0017	.0215	1853	3516	2988	1139	.0040	0038	0014	0004
	14.00	4.204	.0002	0035	.0428	.2142	.3761	.2557	.0976	.0222	0030	0012 0029
226	∫ (~Spontaneous)]											
U ²³⁰	-6.50	1.676	.1217	.3425	.3256	.1511	0576	0071	- 0033	0017	0006	0001
	-4.00	1.974	.0696	.2755	.3621	.1985	.0826	0176	- 0025	- 0022	0000	0001
	-2.00	2.215	.0425	.2173	.3683	.2402	.1073	.0291	- 0004	- 0022	0009	0002
	0	2.458	.0250	.1630	.3523	.2810	1372	0433	0034	- 0031	0012	0003
	1.00	2.581	.0188	.1385	.3368	.2994	.1544	0516	0061	- 0032	0010	0004
	4.00	2.956	.0075	.0780	.2693	.3374	.2140	0821	0176	- 0024	0018	0005
	8.00	3.464	.0017	.0282	.1631	.3291	.2993	1389	0418	0024	0025	0009
	14.00	4.209	.0001	.0014	.0477	.2123	.3566	.2566	.1037	.0255	0010	0015
040	((Spontaneous))											
Pu^{240}	-6.30	2.083	.0545	.2169	4095	2132	1014	0157	- 0041	0040	0010	0004
	-1.50	2.681	.0191	.1062	3118	3313	1788	0585	0041	0049	0018	0004
	0	2.889	.0131	.0764	.2660	.3536	2162	0750	.0050	0003	0032	0011
	7.00	3.761	.0026	.0041	.0886	.2969	3888	1612	.0112	0004	0038	0014
	14.00	4.556	.0009	0069	.0091	.1481	.3578	.3034	.1503	.0421	.00053	0033 0054

TABLE III. CALCULATED NEUTRON EMISSION PROBABILITIES FOR FISSION OF NUCLIDES FOR WHICH ADEQUATE FRAGMENT KINETIC ENERGY DISTRIBUTIONS ARE NOT AVAILABLE

Results are not normalized to a measured $\overline{\nu}$ value, but are based on excitation energies 2.29 Mev less than the computed excitation energy. Effects of (n, n'f) have not been included.

Compound Nucleus	Neutron Energy Inducing Fission (Mev)	$\overline{\nu}$	р ₀	P ₁	^р 2	P ₃	P4	Р ₅	P ₆	P ₇	P.8	Р ₉
Th^{233}	2.00	1.713	.1152	.3305	.3329	.1604	.0586	.0091	0038	0021	0007	0001
	14.00	3.207	.0043	.0507	.2124	.3458	.2557	.1052	.0304	0001	0032	0012
บ ²³⁸	{-5.98 }	2.043	.0591	.2542	.3722	.2149	.0854	.0208	0025	0029	0010	0002
	2.00	3.023	.0063	.0678	.2544	.3469	.2226	.0864	.0218	0022	0029	0010
U ²³⁹	2.00	2.672	.1654	.1232	.3169	.3151	1655	.0586	.0105	0036	0022	0007
	14.00	4.226	.0001	.0023	.0491	.2128	.3494	.2506	.1077	.0303	.0007	0030

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been used in the present calculations. However, also of interest are neutron emission probabilities for fission of other compound nuclei for which only single chamber measurements of the kinetic energy have been made.

Calculations have been made for the fission of three such compound nuclei, U^{238} (Ref. 25), U^{239} (Refs. 1, 26), and Th²³³ (Refs. 26, 27). In these cases the variation of kinetic energy with mass ratio obtained from the fission of U^{236} is used with the average energy of fission obtained from the measurements of these nuclei. The binding energy of neutrons to the fission products of these nuclei is computed on the basis of the computed mass surface. In this calculation, the charges of the primary fragments are determined on the basis of the Glendenin¹⁶ hypothesis. The masses are determined on the basis of a fixed heavy fragment mass distribution¹⁴ independent of the mass of the compound nucleus undergoing fission.

In view of the similarity of the kinetic energy distributions for all the nuclei for which double chamber measurements have been made, such a use of the data of both U^{236} and the nuclei in question is expected to give reasonably accurate data. It is, of course, necessary to take into consideration different conditions of the even-odd parameter δ for the cases of the odd A nuclei U^{239} and Th²³³.

With the approximately 1-Mev uncertainties in the masses used and the approximately 2-Mev uncertainties in the energies used (except for Th²³³, see discussion on page 35), it is not unreasonable to expect an uncertainty of several Mev in the excitation energies derived in the above analysis. Because of this uncertainty, the results of the fission neutron emission calculations for U^{234} , U^{236} , and Pu^{239} have been normalized to the recent $\bar{\nu}$ measurements²⁸ of thermal-neutron induced fission, the $\bar{\nu}$ accuracies are about one percent. As indicated in the caption for Table II, this normalization requires a reduction of the calculated excitation energies E_X by several Mev. The close agreement of the amount of this reduction for the three cases indicates that the consistency (from one fissioning nuclide to another) in the mass and energy analysis is accurate to about 3 Mev.

No $\overline{\nu}$ determinations of comparable accuracy for fission of U^{238} , U^{239} , and Th²³³ are available. Consequently, the 2.29-Mev correction to the excitation energy of the fission of U^{236} is made to apply to these nuclides by the following: The average kinetic energies^{25,1,27}

²⁷J. L. Fowler and L. Rosen, Phys. Rev. 72, 926 (1947).

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²⁵W. J. Whitehouse and W. Galbraith, Phil. Mag. <u>41</u>, 429 (1950).

²⁶W. Jentschke, Z. Physik 120, 165 (1943).

²⁸M. D. Goldberg, J. A. Harvey, D. J. Hughes, and V. E. Pilcher, Brookhaven National Laboratory Report BNL-250, August 1954.



used in these calculations are from measurements which have accompanying U^{236} measurements. Each average U^{236} energy is normalized to that of Brunton and Hanna. The same normalization is then applied to the measurements of U^{238} , U^{239} , and Th²³³.

It is to be noted in the results in Tables II and III that negative emission probabilities for the larger values of ν are obtained. Again, these negative probabilities are not physically possible, but have meaning in the mathematical sense. To test whether these negative probabilities have a large influence on the variation of $\overline{\nu}$ and $\overline{\nu}^2$, these quantities were recalculated after dropping the negative probabilities and renormalizing. Although the absolute value of $\overline{\nu}$ and of $\overline{\nu}^2$ for each value of E_b increased by about one percent, the variation of these quantities with E_b changes very little.

It appears from fission cross section measurements as compiled by Barschall and Henkel²⁹ that the onset for (n, n'f) is between 6 and 7 Mev of incident neutron energy and that the (n, n'f) cross section is of comparable magnitude to the (n, f) cross section up to 14 Mev. The present calculations are only for the (n, f) process, but are adaptable (with reduced accuracy) to the (n, n'f) process by shifting the E_b axis to zero at $E_n \cong 6.5$ Mev. Furthermore, ν is increased by 1 and the nuclide under consideration changes, for example, from U^{236} to U^{235} . It will be seen in the comparison of the results from U^{239} and U^{238} that the neutron emission characteristics of neighboring isotopes are essentially the same.

(a) Fission of U^{236}

Since more experimental data of neutron emission exist for fission of U^{236} than for other nuclides, the fission of this nuclide was investigated most thoroughly in the calculations. In Table IV are the results of the neutron emission calculations for three conditions of temperature and for three conditions of dispersion in the kinetic energy data.

For each parameter, the lower and upper values are considered to be limiting values on the basis of the available data. It is to be noted that changes in the dispersion have considerably less effect on the change of $\overline{\nu}$ and $\overline{\nu^2}$ with E_n than do changes in the nuclear temperature.

In Figs. 6 and 7 are plotted the results from Table II for $\overline{\nu}$ and ν^2 , respectively. Data in addition to those in Table IV show that the linear relation between $\overline{\nu}$ and E_n in Fig. 4 also applies for the various conditions of T and u discussed above. In Fig. 8 are plotted the results of $P_{\mu}(E_n)$ of Table II.

²⁹H. H. Barschall and R. L. Henkel, LA-1714, August 1954.

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Fig. 6. Plot of the $\overline{\nu}$ data of Table II.









Fig. 8. Plot of the fission neutron emission probabilities of Table II for U^{236} fission.

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TABLE IV.DEPENDENCE OF CALCULATED NEUTRON EMISSION PROBABILITIESUPON NUCLEAR TEMPERATURE AND UPON DISPERSION

Calculations are approximately normalized to $\overline{\nu} = 2.46$ for $E_n = 0$ by shifting the calculated excitation energy distribution. The parameter u is the width term in the Gaussian expression of the dispersion (Eq. 5). Data are for fission of U²³⁶.

Т	u	En			$d\overline{\nu}/dE_n$
(Mev)	(Mev)	(Mev)	$\overline{\nu}$	ν^2	(Mev ⁻¹)
1.0	7.2	0	2.4593	7.3580	0 1468
210	112	8	3.6341	14.7369	0.1400
1.0	7.9	0	2.4648	7.1403	0 1150
1.0	7.2	8	3.3897	12.5834	0.1156
1 4	5.0	0	2.4631	7.3567	0 1040
1.4	5.9	8	3.4595	13.3069	0.1246
1 /	05	0	2.4557	7.0716	0 1969
1.4	0.0	8	3.4661	13.1056	0.1263
1 4*	F 9+	0	2.4581	7.2139	0 1055
1.4*	1.2*	8	3.4636	13.2096	0.1257

*Conditions used for other calculations in this report.

(b) Fission of
$$U^{234}$$
 and Pu^{240}

In Figs. 6 and 7 are the plots of $\overline{\nu}$ and $\overline{\nu}^2$, respectively, for the fission of U²³⁴ and Pu²⁴⁰. These data are in Table II.

In Table III are listed the fission neutron probabilities for the fission of Th^{233} , U^{238} , and U^{239} . As discussed on page 25, these results are not normalized to a $\overline{\nu}$ value, but are based on the same excitation energy correction applied to fission of U^{236} .

(d) Effects of (n, n'f)

In Fig. 9 is shown the variation of $\overline{\nu}$ when the effects of (n, n'f) are taken into account, as discussed on page 26. In this analysis it was assumed that the variation of $\overline{\nu}$ as a function of E_b was the same for U^{234} as for U^{233} , for U^{236} as for U^{235} , and for Pu^{240} as for Pu^{239} . This assumption is based upon the results of U^{238} and U^{239} listed in Table III. The difference between the $\overline{\nu}$ results of these two isotopes is satisfactorily explained by the difference in the kinetic energy used in the calculation of these two nuclei. On the basis of the Coulomb origin

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Fig. 9. Plot of $\overline{\nu}$ data of Table II corrected for the effects of (n, n'f).

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of the kinetic energy, the same kinetic energy is expected from the two nuclei instead of the measured 3 Mev less for U^{238} compared²⁵ to U^{236} and 0.7 Mev less for U^{239} compared¹ with U^{236} . It is to be noted that the 2.3-Mev difference between the average kinetic energies of U^{239} and U^{238} used in the calculations accounts for 0.30 in $\overline{\nu}$ on the basis of the calculated $d\overline{\nu}/dE_n$. This is to be compared with the 0.35 difference in $\overline{\nu}$ calculated with these masses and energies.

DISCUSSION

The variation of $\overline{\nu}$ with E_n obtained from these calculations is to be compared with the experimental measurements by sphere multiplication by Graves³⁰ and Beyster,³¹ with the preliminary results from scintillator tank work by Martin et al.,³² with the chamber measurements by Fowler,³³ and with a combination of counter and radiochemical determinations by Terrell.³⁴ Similarly, the results of spontaneous fission measurements by Carter³⁵ and by Segre³⁶ for spontaneous fission of Pu²⁴⁰ and of Segre³⁷ and Littler³⁸ for U²³⁸ are to be compared with the calculated results for spontaneous fission. In addition, the liquid scintillation tank has been used by Martin et al.³² and Hammel and Kephart³⁹ to obtain preliminary results on the spontaneous fission of Pu²⁴⁰. In Table V is a comparison of these experimental results with the present calculated results, and in Table VI is a similar comparison of the P_v data.

- ³²Martin, Terrell, and Diven, LASL, P-3-76, 1954 and P-Division Progress Report, January 20, 1955 (not available).
- ³³J. L. Fowler, Reactor Science and Technology <u>4</u>, 141 (1954) and Oak Ridge National Laboratory Report ORNL-1715, July 1954.

- ³⁶E. Segrè, LA-491, November 1946.
- ³⁷E. Segrè, Phys. Rev. <u>86</u>, 21 (1952).
- ³⁸D. J. Littler, Proc. Phys. Soc. London A65, 203 (1952).
- ³⁹J. E. Hammel and J. F. Kephart, as reported in DIR 1003, LASL, 1954 (not available).

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³⁰E. R. Graves, LASL P-Division Progress Report, June 20, 1954 (not available).

³¹J. R. Beyster, private communication, 1954.

³⁴N. J. Terrell, Jr., Reactor Science and Technology 4, 141 (1954).

³⁵W. W. Carter, LA-1582, July 1953.

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TABLE V.COMPARISON OF EXPERIMENTAL AND CALCULATED RESULTSOF AVERAGES OF FISSION NEUTRON EMISSION

The calculated results are of Figs. 7 and 9. Zero energy neutrons and the subscript "th" indicate thermal-neutron induced fission. Effects of (n, n'f) are included.

Compound Nucleus	Neutron Energy Inducing Fission (Mev)	Quantity	Calculated	Experiment	Experiment Reference
U ²³⁶	1.25	$\overline{\nu}$	2,61	$2.59 \pm .06$	32
	4.8	$\overline{\nu}$	3.06	$3.12 \pm .07$	32
	4.0	$\overline{ u}$	2.95	$3.12 \pm 10\%$	31
	4.5	$\overline{ u}$	3.01	$3.28 \pm 10\%$	31
	Fission Spectrum	$\overline{\nu}/\overline{\nu_{th}}$	1.10	$1.05 \pm .03$	31
	0.7	$\overline{\nu}/\overline{\nu}_{th}$	1,034	$1.02 \pm .02$	34
	0.5	$\overline{\nu}/\overline{\nu}_{th}$	1.02	$1.14 \pm .13$	33
	2.7	$\overline{\nu}/\overline{\nu}_{th}$	1.13	$1.42 \pm .26$	33
	3.7	$\overline{\nu}/\overline{\nu}_{th}$	1.19	$1.63 \pm .23$	33
	5.0	$\bar{\nu}/\bar{\nu_{th}}$	1.29	$1.71 \pm .19$	33
	14.0	$\overline{\nu}/\overline{\nu_{th}}$	1.79	$1.99 \pm .23$	33
	14.0	$\overline{\nu}$	4.29	3.95	30
Pu ²⁴⁰	Spontaneous	$\overline{ u}$	2.08	$2.20 \pm .05$	32
	Spontaneous	$\overline{ u}$	2.08	$2.22 \pm 5\%$	35
	Spontaneous	$\overline{ u}$	2.08	$2.37 \pm .3$	36
U^{238}	Spontaneous	$\overline{ u}$	2.04	$2.2 \pm .3$	37
	Spontaneous	$\overline{ u}$	2.04	$2.5 \pm .2$	38
	Spontaneous	$\overline{\nu^2}/\overline{\nu}$	2.57	$3.26 \pm .2$	40
	Spontaneous	$\overline{\nu^3}/\overline{\nu}$	7.28	$12.73 \pm .9$	40
U^{239}	1.5	$\overline{\nu}$	2.61	$2.58 \pm .09$	32
	4.5	$\overline{\nu}$	2,99	$3.31 \pm \sim 10\%$	31
	14.0	$\overline{ u}$	4.30	3.5	30





Reference	Compound Nucleus	Neutron Energy Inducing Fission (Mev)	Р ₀	Р ₁	P ₂	P ₃	P4	P ₅	Р ₆	P ₇
32	U^{236}	0.08	.014	.165	.357	.306	.115	.046	006	002
			±.005	±.016	±.022	±.024	±.020	±.013	±.005	±.002
Calc.	U ²³⁶	0	.025	.163	.352	.281	.137	.043	.003	003
32	Pu ²⁴⁰	Spontaneous	.050	.215	.352	.263	.110	.013	005	.003
			±.009	±.021	±.029	±.031	±.026	±.015	±.008	±.004
39	Pu ²⁴⁰	Spontaneous	.008	.26	.42	.15	.11	.05		
Calc.	Pu ²⁴⁰	Spontaneous	.055	.217	.410	.213	.101	.016	004	005

TABLE VI. COMPARISON OF EXPERIMENTAL AND CALCULATED RESULTS OF PROBABILITIES OF NEUTRON EMISSION P $_{\nu}$

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The calculated increase of $\overline{\nu}$ with E_n is seen from Table V to be very close to the measured increase, perhaps being somewhat larger than the measured increase. As seen by Table IV, the difference between the 14-Mev values of Graves compared with the calculated values in Table V and, similarly, the corresponding spontaneous fission values for Pu^{240} , can probably be explained on the basis of temperature or dispersion alone. As an alternative explanation, these differences can possibly be explained by a true mass surface in the region of the fission products that is less steep than predicted by the parabolic surface used.

The calculated results of $\overline{\nu}$ and $\overline{\nu^2}$ for U²³⁶ in Figs. 6 and 7, respectively, were integrated over the fission neutron spectrum and the resulting values for fission neutron induced fission are $\overline{\nu} = 2.70$ and $\overline{\nu^2} = 8.48$.

Multiplicity measurements have been made by counter techniques for spontaneous fission of U^{238} by Geiger.⁴⁰ It is to be noted in Table V that satisfactory agreements between most of the more recent experimental data and the calculated data exist, but the multiplicity measurements of Geiger are somewhat larger than the present calculated results. This discrepancy can be partially explained by the apparently incorrect normalization for the U^{238} data, as evidenced by the low calculated results of $\overline{\nu}$ of spontaneous fission of U^{238} compared with the experimental value. Such a possibility of the calculated $\overline{\nu}$ being low is, however, difficult to explain on the basis of the energies used in the calculation. As discussed on page 32, the measured kinetic energy of fission used in the calculated $\overline{\nu}$ is about 0.36 greater than what would be obtained if the "true" Coulomb energy were used in the calculations.

Of all the calculated results, those of the compound nucleus Th²³³ are most uncertain. As seen in Table III, the use of the E_C values of Fowler and Rosen²⁷ in the calculations yields surprisingly low values of $\bar{\nu}$. These 1947 data of Fowler and Rosen²⁷ were used in preference to the 1943 data of Jentschke²⁶ as being more recent. However, a surprisingly large discrepancy exists between these two sets of experimental data when each is normalized to the accompanying U²³⁶ measurements. Fowler and Rosen find 2.6 Mev less energy for Th²³³ than U²³⁶ while Jentschke finds 9.5 Mev less. On the basis of the calculated $d\bar{\nu}/dE_n$, $\bar{\nu}$ values 0.85 greater than those of Table III would be obtained if the data of Jentschke were used. In view of these large discrepancies for Th²³³, the results for Th²³³ are considered to be of less accuracy than the other data.

The present calculations can also be used to determine the neutron emission probabilities from the several mass groups of the fission products used in the calculations. These results are contained in Table VII. It is seen that no correlation of $\overline{\nu}$ with R_m can be determined

⁴⁰K. W. Geiger and D. C. Rose, Can. J. Phys. <u>32</u>, 498 (1954).

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with certainty. This is in contrast to the radiochemical determinations,¹⁵ which indicate that $\overline{\nu}$ is lower for R_m values near unity than for larger R_m values. Thus, radiochemical determinations indicate a relatively smaller probability of neutron emission from the fragments in the symmetrical fission region. It should be emphasized that the E_C data for the low and high R_m values for each fission case are of relatively low accuracy compared with the E_C data of the most probable mass ratio value. This is because of poorer statistics and the effects of dispersion.⁴ As a result, the $\overline{\nu}$ values calculated for these low and high R_m values are subject to error.

TABLE VII. CALCULATED NEUTRON EMISSION PROBABILITIES OF VARIOUS GROUPS

These data correspond to thermal-neutron induced fission when the calculated excitation energies are normalized as in Table II. Mass ratios are the center of each of the three mass ratio groups. The results for all mass ratio groups (properly weighted) are referred to as "all."

Compound Nucleus	Mass Ratio	$\overline{\nu^{L}}$	$\overline{\nu^{\mathrm{H}}}$	$\overline{\nu}$	$\overline{\nu^{L}}/\overline{\nu^{H}}$
U ²³⁴	133/101	0.828	0.695	1.539	
	141/93	1.218	1.286	2.504	
	149/85	2.475	2.365	4.839	
	A11	1.328	1.327	2.655	1.001
U ²³⁶	131/103	1.440	1.152	2.592	
	141/95	1,212	1.244	2.456	
	149/87	1.045	0.970	2.015	
	A11	1,248	1.185	2.433	1.053
Pu ²⁴⁰	131/109	1.341	1.105	2.446	
	139/101	1.523	1.607	3.131	
	147/93	1,458	1.453	2.912	
	A11	1.449	1.410	2.859	1.027

It is of interest to compare the calculated $\overline{\nu^L}/\overline{\nu^H}$ values of Table VII with the measured values found by Fraser⁴¹ by the angular correlation of fragments and neutrons in a coincidence

⁴¹J. S. Fraser, Phys. Rev. <u>88</u>, 536 (1952).

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experiment. Fraser found $\overline{\nu^L}/\nu^H$ to be 1.30 ± 0.08 for fission of the compound nuclei U^{234} , U^{236} , and Pu²⁴⁰. The corresponding ratios 1.00, 1.05, and 1.03 in Table VII indicate that the assumed equality of the excitation energy distributions X^L and X^H of the light and heavy fragments is in error and, instead, larger excitation energies of the light fragments should be used. On the other hand, the theory of fission proposed by Fong⁴ predicts $\overline{\nu^L}/\nu^H < 1$.

In later work on U^{234} with similar techniques, Fraser and Milton⁴² found $\overline{\nu}$ to be greatest for the most probable fission mode and to decrease 10 to 20 percent for symmetrical and approximately symmetrical fission modes. In addition, they found, for both the light and heavy fragments, an increase of $\overline{\nu}^{L}$ or $\overline{\nu}^{H}$ with mass of the fragment. As discussed above, the uncertainties of the E_C data for the less probable fission modes makes the corresponding data of Table IV unsuitable for comparison.

In addition to the data compared in Table V, two other determinations of $\overline{\nu}$ can be compared. Wahl⁴³ used the observed nuclear charge distribution of the fission products from 14-Mev neutron induced fission of the compound nucleus U^{236} and the Glendenin hypothesis¹⁶ of the nuclear charge of fission fragments and obtained a value of $\overline{\nu} = 5.2 \pm 0.5$ for 14-Mev neutron fission of the compound nucleus U^{236} . Using the same method with the same nuclide, Ford⁴⁴ found $\overline{\nu} = 3.1$ for $E_n = 5$ Mev and $\overline{\nu} = 4.8$ for $E_n = 14$ Mev. An analysis of recent sphere measurements with various energy neutrons inducing fission of the compound nucleus U^{236} has been made by Goad.⁴⁵ She finds the data fit best by $\overline{\nu} = 2.5$ for $E_n < 3.9$ Mev, $\overline{\nu} = 3.0$ for 3.9 Mev $< E_n < 6.5$ Mev, and $\overline{\nu} = 3.5$ for $E_n > 6.5$. Because of the less direct determinations of these values, they have not been included in Table V, even though the agreements with the present calculated results are reasonable.

- ⁴⁴G. P. Ford, private communication, 1952.
- 45 M. S. Goad, T-602, 1954, and revisions of this (not available).

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⁴²J. S. Fraser and J. C. D. Milton, Phys. Rev. <u>93</u>, 818 (1954).

⁴³J. S. Wahl, LAB-P-4 Memo, 1952 (not available).



APPENDIX

This appendix describes the method of solution of the integral equations 6 and 8 involving the empirical data of $I(E_1, R_m)$.

For convenience, the transposition of the E_I data by the 12.4-Mev ionization defect is first made by a replotting of the $I(E_I)$ data into $I'(E_I')$ data, where E_I' is $E_I' = E_I + 12.4$. With such a transposition of the original data, a transposition by the dispersion function is not necessary. The dispersion is thus redefined by

$$D'(E_{C}, E_{I}') \propto \exp\left[-\left(\frac{E_{C} - E_{I}'}{u}\right)^{2}\right] , \qquad (13)$$

where u is the width of the dispersion.

In the approximate method, it is assumed that $I'(E_{I}')$ can be fit adequately well by a series of Gaussian functions so that

$$I'(E_{I}') = \sum_{\beta=0}^{12} b_{\beta} \exp\left[-\left(\frac{E_{I}' - E_{0} + \beta d}{w}\right)^{2}\right], \qquad (14)$$

where b_{β} terms are amplitude coefficients, β is a summation parameter, E_0 is an energy constant, d is the spacing of the Gaussians, and w is the width term of the Gaussians. In Eq. 14 and the analysis to follow, the parameters R_m and δ are not written because the analysis applies for any condition of R_m and δ .

Since the $I'(E_{I}')$ data are fit by Eq. 14, a limited freedom of choice of the values of the E_{o} , w, and d terms exists. In practice, the solutions of the equations to follow were facilitated by the following choices: The width w is slightly less than the width of the $I'(E_{I}')$ distribution to be fitted, d is roughly 0.8 w, and $E_{o} = (E_{I}')_{max} + 7d$. The most probable E_{I}' is $(E_{I}')_{max}$. With the 13-Gaussian system used, these choices result in the seventh Gaussian as the dominant term and the other Gaussians tailor this curve to fit the empirical $I'(E_{I}')$ data.

Two Gaussian expressions can be shown to fold as

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$$\exp\left[-\left(\frac{\mathbf{E}_{\mathbf{I}}' - \mathbf{E}_{\mathbf{0}} + \beta \mathbf{d}}{\mathbf{w}}\right)^{2}\right] \propto \int_{-\infty}^{\infty} d\mathbf{E}_{\mathbf{C}}\left\{\exp\left[-\left(\frac{\mathbf{E}_{\mathbf{C}} - \mathbf{E}_{\mathbf{0}} + \beta \mathbf{d}}{\mathbf{v}}\right)^{2}\right]\right\}\left\{\exp\left[-\left(\frac{\mathbf{E}_{\mathbf{C}} - \mathbf{E}_{\mathbf{I}}'}{\mathbf{u}}\right)^{2}\right]\right\}\right\}$$
(15)

where $w^2 = v^2 + u^2$. Thus, a substitution of the fitted I'(E_{I} ') expression of Eq. 14 into the convolution (Eq. 6) is,

$$\sum_{\beta=0}^{12} b_{\beta} \exp\left[-\left(\frac{E_{I}' - E_{o} + \beta d}{w}\right)^{2}\right]$$

$$\propto \int_{-\infty}^{\infty} dE_{C} \left\{\sum_{\beta=0}^{12} b_{\beta} \exp\left[-\left(\frac{E_{C} - E_{o} + \beta d}{v}\right)^{2}\right]\right\} \left\{\exp\left[-\left(\frac{E_{C} - E_{I}'}{u}\right)^{2}\right]\right\}.$$
(16)

From Eqs. 16 and 6, the fitted Coulomb energy distribution is seen to be

$$C(E_{C}) = \sum_{\beta=0}^{12} b_{\beta} \exp\left[-\left(\frac{E_{C} - E_{o} + \beta d}{v}\right)^{2}\right], \qquad (17)$$

where $w^2 = v^2 + u^2$ and v is the width of the Gaussian terms fitting $C(E_C)$.

The transformation from Coulomb energy E_C to the total excitation energy of the fragment pairs E_X is by Eq. 2. Thus, with $E_C = \phi - E_X$, the excitation energy distribution is written

$$X(E_{X}) = \sum_{\beta=0}^{12} b_{\beta} \exp\left[-\left(\frac{\phi - E_{0} - E_{X} + \beta d}{v}\right)^{2}\right].$$
 (18)

It is now required to find the excitation energy distributions of the individual fragments. As discussed in the text, these are considered identical, such that $x^{L} = x^{H}$, and are combined by

$$X(E_{X}) = \int_{-\infty}^{\infty} dE_{X}^{L} X^{L}(E_{X}^{L}) X^{H}(E_{X} - E_{X}^{L}) .$$
 (8a)

The substitution of $X(E_X)$ of Eq. 18 into Eq. 8a gives

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$$\sum_{\beta=0}^{12} b_{\beta} \exp\left[-\left(\frac{\phi - E_{o} - E_{X} + \beta d}{v}\right)^{2}\right] = \int_{-\infty}^{\infty} dE_{X}^{L} X^{L}(E_{X}^{L}) X^{H}(E_{X} - E_{X}^{L}) .$$
(19)

It can be shown that Eq. 19 and $X^{L} = X^{H}$ are satisfied by

$$\begin{bmatrix}
12 \\
\sum \\
B \\
\beta = 0
\end{bmatrix}
^{12} b_{\beta} \exp \left[-\left(\frac{\phi - E_{0} - E_{X} + \beta d}{v}\right)^{2} \right]$$

$$= \int_{-\infty}^{\infty} dE_{X}^{L} \left\{ \sum_{\alpha=0}^{6} a_{\alpha} \exp \left[-\left(\frac{\phi - E_{0}}{2} - E_{X}^{L} + \alpha d\right)^{2} \right] \right\}$$

$$\times \left\{ \sum_{\alpha=0}^{6} a_{\alpha} \exp \left[-\left(\frac{\phi - E_{0}}{2} - E_{X} + E_{X}^{L} + \alpha d\right)^{2} \right] \right\},$$
(20)

.

if the following conditions between the amplitude parameters \mathbf{b}_β and \mathbf{a}_α are satisfied:

$$b_{0} = a_{0}^{2}$$

$$b_{1} = 2 a_{0}a_{1}$$

$$b_{2} = 2 a_{0}a_{2} + a_{1}^{2}$$

$$b_{3} = 2 a_{0}a_{3} + 2 a_{1} a_{2}^{2}$$

$$b_{4} = 2 a_{0}a_{4} + 2 a_{1} a_{3} + a_{2}^{2}$$

$$b_{5} = 2 a_{0}a_{5} + 2 a_{1} a_{4} + 2 a_{2}a_{3}$$

$$b_{6} = 2 a_{0}a_{6} + 2 a_{1}a_{5} + 2 a_{2}a_{4} + a_{3}^{2}$$

$$b_{7} = 2 a_{1}a_{6} + 2 a_{2}a_{5} + 2 a_{3}a_{4}$$

$$b_{8} = 2 a_{2}a_{6} + 2 a_{3}a_{5} + a_{4}^{2}$$

$$b_{9} = 2 a_{3}a_{6} + 2 a_{4}a_{5}$$

$$b_{10} = 2 a_{4}a_{6} + a_{5}^{2}$$

$$b_{11} = 2 a_{5}a_{6}$$

$$b_{12} = a_{6}^{2}$$

(21)

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When these conditions are satisfied the excitation energy distributions of the individual fragments are found as

$$X^{L}(E_{X}^{L}) = \sum_{\alpha=0}^{6} a_{\alpha} \exp\left[-\left(\frac{\phi - E_{0}}{2} - E_{X}^{L} + \alpha d\right)^{2}\right]$$
(22)

and the corresponding equation for the heavy fragment. Again, $w^2 = v^2 + u^2$.

It is now necessary to use the conditions given in Eqs. 21 with the fit of the $I'(E_{I'})$ experical data by Eq. 14 to solve for the a_{α} amplitude parameters required in the X^{L} and X^{H} expressions of Eq. 22. Equations 21 are a set of 13 simultaneous equations that are quadratic in a_{α} terms. To solve for the a_{α} values, the 13 expressions for the 13 b_{β} terms in Eqs. 21 are substituted in the equation of the fit to the empirical data, which is Eq. 14. The resulting 13 simultaneous equations, each of which is a quadratic of a_{α} terms multiplied by a Gaussian expression, are to be solved at 13 empirical values of $I'(E_{I'})$.

The method used for this solution was to take as the 13 values of $I'(E_0')$ the values at $E_I' = E_0, E_I' = E_0 - d, E_I' = E_0 - 2d, \ldots E_I' = E_0 - 12d$. Under these conditions and the previously stated conditions that result in b_6 being dominant, these 13 simultaneous, quadratic equations are solved by an iterative method. The first solution is assumed to be $a_3 = 1$ and $a_0 = a_1 = a_2 = a_4 = a_5 = a_6 = 0$. With this solution substituted, the resulting inequality in the fourth equation is used to find a_0 , the inequality in the fifth equation to find a_1 , the sixth for a_2 , the eighth for a_4 , the ninth for a_5 , and the tenth for a_6 . These solutions of a_{α} are substituted and the same corrections are applied. The process is repeated many times. The resulting fits of the empirical $I'(E_1')$ data are given in Table VIII.





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Compound Nucleus	R _m	w (Mev)	d (Mev)	E _O (Mev)	^a 0	^a 1	^a 2	^a 3	^a 4	^a 5	a 6
U ²³⁴	133/101	11.40	9.0	223	020	012	.196	1.000	.069	.170	105
	141/93	11.40	9.0	217	.019	048	.068	1.000	.016	.061	037
	149/85	9.48	7.5	201	.238	486	.642	1.000	.335	077	.006
U ²³⁶	133/103	13.80	11.0	241	001	011	.039	1.000	.066	.088	034
	141/95	11.40	9.0	220.5	.010	041	.120	1.000	.046	.066	.028
	149/87	9.48	7.5	204	022	.009	.127	1.000	.081	.069	042
Pu ²⁴⁰	131/109	13.80	11.0	245	.060	161	.339	1.000	.136	.129	009
	139/101	11.40	9.0	226	.101	228	.341	1.000	.074	.098	040
	147/93	9.48	7.5	209	029	.023	.120	1.000	.015	.090	038

TABLE VIII. PARAMETERS FOUND TO FIT THE EMPIRICAL E_I' DATA

The a values are relative to $a_3 = 1.00$ and are not absolute.

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