Model for Calculating Independent Radiochemical Yields from the Thermal-Neutron Fission of U^{235}

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An empirical model was devised to predict independent radiochemical yields from the thermal-neutron fission of U^{235}. As much as possible, experimental data were used as a basis for the calculation. The resulting model is best described as a method of correlating the available mass, energy, and yield data in order to predict other quantities, particularly independent radiochemical yields. Calculated radiochemical mass yields, independent yields, kinetic energies, and neutron emission probabilities are compared with experiment. The agreement is generally good. However, the calculation shows a large discrepancy in the energy balance at symmetric fission. Also, it indicates that less than half of the observed gamma-ray energy per fission is accounted for by gamma-ray emission following neutron emission. The results also suggest that even-even fragments are preferentially formed.

INTRODUCTION

THE object of this work is to calculate the independent radiochemical yields from the thermal-neutron fission of U^{235}. Previous work has given a good idea of the general features of the charge distribution of the fragments. The available data suggest that the frequency distribution of the isobars of a given mass is a narrow Gaussian.[1] The center of this Gaussian gives the most probable charge of the fragments of a given mass. Wahl used the independent yield data and an assumed Gaussian distribution to derive a curve giving the most probable charge versus fragment mass.[2] His curve shows a small but significant deviation from the charge distribution obtained by setting the charge proportional to the fragment masses.

We decided to try to calculate the independent yields by a method which would take advantage of the above information and also would take advantage of the extensive data now available on semi-empirical fragment binding energies,[3–6] neutron emissions as a function of mass,[7,8] and the initial mass yields of the fragments.[9–11] Instead of assuming a Gaussian distribution for the *radiochemical* charge distribution (for a given mass), we assumed that the *initial* charge distribution is Gaussian. We also assumed that the excitation energy of each fragment has a Gaussian probability distribution. We combined these assumptions with the initial mass yield and neutron emission data to calculate the independent radiochemical yields. We also tried calculating the most probable charges from the binding energy data, but found it more fruitful to use Wahl's prescription.

Throughout the calculation we used empirical or experimental data wherever possible, and introduced simplifying assumptions only when necessary. The resulting model is best described as a method of correlating the available mass, energy, and neutron yield data for the purpose of predicting other quantities, particularly independent radiochemical yields.

The calculation reproduces the observed radiochemical yields as well as or better than other methods. The calculation also predicts fragment excitation energies and kinetic energies. The calculated kinetic energies agree well with experiment, except in the region of symmetric fission.

The semiempirical masses of Cameron,[3] the initial yield data of Milton and Fraser,[9] and the neutron emission data of Apalin[7] were used as input in the calculation. These are not necessarily the only available data, but appeared to the authors to be the most reasonable choice at the time the calculation was made. The calculation was also run using Seeger's semi-empirical mass data,[4] and essentially the same results were obtained (see text).

Before going further we review the nomenclature. The preface *initial* refers to fragment parameters before emission of any neutrons, and the preface *radiochemical* refers to fragments after neutron emission. Yields for all nuclides with a given mass number are called *mass* yields, and yields of a particular nuclide are called *independent* yields. An independent radiochemical yield, for example, is the number per 100 fissions of a particular nuclide produced after neutron emission but before beta decay.

[1] See for example C. D. Coryell, M. Kaplan, and R. D. Fink, Can. J. Chem. **39**, 646 (1961).
[2] A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962).
[3] A. G. W. Cameron, Can. J. Phys. **35**, 1021 (1957); and also A. G. W. Cameron, At. Energy Can. Ltd., AECL-690 (1957).
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[5] H. B. Levy, Lawrence Radiation Laboratory Report No. UCRL-4588, 1955 (unpublished); and J. Riddell, At. Energy Can. Ltd., CRP-854, 1962 (unpublished).
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PROCEDURE

The independent radiochemical yield of the nucleus (Z,M) is given by

$$Y(Z,M)=\sum_{\nu} p_\nu(Z,M+\nu)y(Z,M+\nu). \quad (1)$$

The quantity $p_\nu(Z,M)$ is the probability of the nucleus (Z,M) emitting ν neutrons, and $y(Z,M)$ is the initial independent yield of (Z,M). To calculate $Y(Z,M)$ we must estimate the quantities on the right side of Eq. (1). First we consider the initial independent yields $y(Z,M)$.

We assumed that the initial charge distribution is Gaussian:

$$y'(Z,M)=[y(M)/(2\pi\sigma_Z^2)^{1/2}] \times\exp[-(Z-Z_0)^2/2\sigma_Z^2]. \quad (2)$$

The quantities $y(M)$ are the experimental initial mass yields. The initial independent yield $y(Z,M)$ is obtained by integrating Eq. (2) from $Z-1/2$ to $Z+1/2$. The parameters $Z_0(M)$ and σ_Z were chosen to fit, within the limits of the model, the available data on independent radiochemical yields. In particular, σ_Z was taken to be 0.55 charge units, and the quantities $Z_0(M)$ are a slight modification of those deduced by Wahl from radiochemical yield data. The choice of the parameters $Z_0(M)$ is discussed in detail in Appendix A.

Next, we consider the neutron emission probabilities, $p_\nu(Z,M)$. Our method of estimating these quantities is more involved. We assumed a two-parameter probability distribution for the excitation energy of each fragment before neutron emission. The neutron emission probabilities were calculated from this excitation energy distribution. First, we discuss the method of going from the excitation energy distribution to the neutron distribution, and then we will discuss the method of determining the two parameters in the excitation energy distribution.

The excitation energy of each fragment was assumed to have a Gaussian probability distribution:

$$n(W)=[1/(2\pi\sigma_W^2)^{1/2}]\exp[-(W-\bar{W})^2/2\sigma_W^2]. \quad (3)$$

The quantity $n(W)dW$ is the probability that the fragment is formed with excitation energy between W and $W+dW$. The excitation energy is W. The variance $\sigma_W^2(M)$ and the average energy $\bar{W}(M)$ are parameters to be determined. We assumed that these parameters are the same for every isobar of a given mass number. However, they were allowed to vary with mass number. The Gaussian distribution is suggested by the experimental kinetic-energy distribution[9] (see Appendix B).

The neutron emission probabilities were calculated from these excitation energy distributions. We assumed that all neutrons are emitted with 1.21 MeV kinetic energy, the average center-of-mass energy. (This oversimplification in the kinetic-energy distribution is partially corrected for in the excitation energy distribution, see Appendix B.) We also assumed that each nucleus emits as many neutrons as its excitation energy allows. (We therefore neglected gamma-ray emission preceding or competing with neutron emission. This point is discussed in more detail later.) With these assumptions the probability of the nucleus (Z,M) emitting ν neutrons is given by

$$P_\nu(Z,M)=\int_a^b dW(2\pi\sigma_W^2)^{-1/2} \times\exp[-(W-\bar{W})^2/2\sigma_W^2]. \quad (4)$$

In this equation,

$$a=\sum_{i=0}^{i=\nu-1}[S_n(Z,M-i)+1.21] \quad \text{for} \quad \nu\neq 0,$$

$$a=-\infty \quad \text{for} \quad \nu=0,$$

and

$$b=\sum_{i=0}^{i=\nu}[S_n(Z,M-i)+1.21].$$

The quantity $S_n(Z,M)$ is the energy required to separate a neutron from the nucleus (Z,M). These quantities were taken from the semiempirical mass values of Cameron.[3] (Additional calculations using Seeger's mass values[4] indicated that the different choice of mass values had little effect on the final results.) The quantity a is the energy needed to separate ν neutrons (each with 1.21-MeV kinetic energy) from the nucleus, and the quantity b is the energy required to separate $\nu+1$ neutrons.

We now take up the determination of the parameters $\sigma_W^2(M)$ and $\bar{W}(M)$ in the excitation energy distribution, Eq. (3). The variances $\sigma_W^2(M)$ were allowed to vary with mass number. They were chosen so that the variances for corresponding light and heavy fragments are equivalent to the measured variance of the kinetic-energy distribution for that mass pair. (Also, a small correction was made for the neutron-kinetic-energy spread.) The estimates of the excitation energy variances are discussed in more detail in Appendix B.

The average excitation energies $\bar{W}(M)$ were also assumed to be a function only of mass number M. The quantity $\bar{W}(M)$ was chosen to make the nucleus emit the correct average number of neutrons for that mass number. Thus the average numbers of neutrons emitted as a function of mass number were needed for input data. The data of Apalin *et al.* were used.[7] Given $\bar{\nu}(M)$, $\bar{W}(M)$ is found by an iterative process. For each mass number, a value of $\bar{W}$ is inserted in Eq. (4), and an average number of neutrons calculated from the values of $p_\nu(Z,M)$ for a given value of M. The value of $\bar{\nu}(M)$ so obtained was compared with the experimental value of $\bar{\nu}(M)$, $\bar{W}$ was readjusted, and the process repeated until the calculated and input values of $\bar{\nu}(M)$ agree within 1%.

Thus Eqs. (2) through (4) gave us estimates of the initial independent yields and the probabilities of

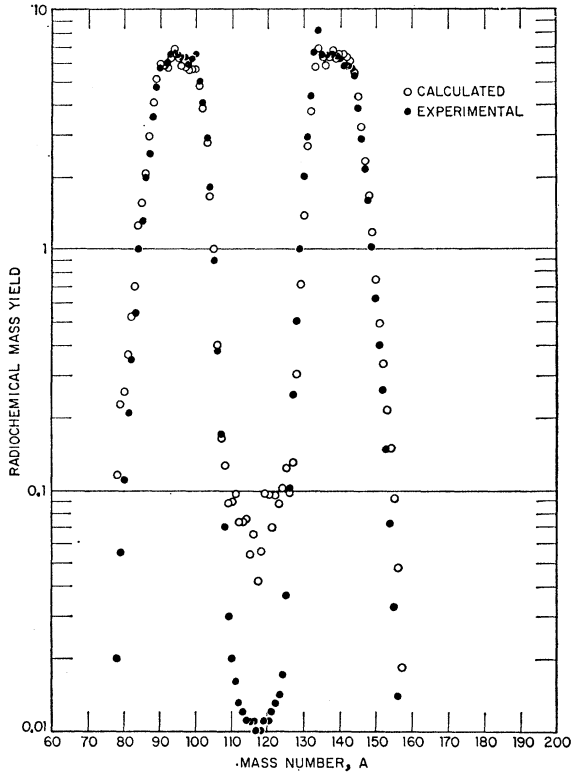

FIG. 1. Comparison of calculated and measured radiochemical mass yields. The open circles are the radiochemical mass yields calculated in this paper, and the solid circles are measured and interpolated values taken from the compilations of Farrar *et al.*[12] The discrepancies are attributed to the finite resolution of the initial mass yield and neutron measurements which were used as input in the calculation.

emitting ν neutrons from each fission fragment. The independent radiochemical yields were then calculated from Eq. (1). To keep the computer calculation within reason, the summation over ν was cut off at $\nu=6$, and only yields greater than 10^{-4} were calculated.

The above indicates how initial yields and excitation energies were obtained as part of the calculation. Some other quantities also were calculated from this model. The excitation energies were combined with semi-empirical mass calculations to estimate the fragment kinetic energies. Also, after the excited fragment has emitted its maximum number of neutrons, it still generally has energy to lose by gamma-ray emission. We calculated this (residual) energy available from gamma-ray emission for each fragment mass. (The gamma-ray energy obtained in this manner does not include gamma rays emitted before or in competition with neutron emission. Since there is evidence that either or perhaps both of these processes are significant, our calculated average gamma-ray energies are expected to be low. This was found to be the case, and is discussed in the next section.)

Finally, if we assume that the excitation energies of a pair of fragments are statistically independent, the probability of emitting n neutrons from the pair is

$$Q_n=\sum_{\nu} p_\nu(Z,M)p_{n-\nu}(92-Z,236-M), \qquad (5)$$

and the probability of emitting n neutrons per fission is given by

$$P_n=\sum_{Z,M} Q_n. \qquad (6)$$

These and all the other quantities discussed above were calculated with the aid of a computer, and the results are described in the following sections.

RESULTS

Radiochemical yields were calculated for mass number 78 through 158. The calculated mass yields are compared with the experimental compilation of Farrar *et al.*[12] in Fig. 1. The agreement is good except for the small yields on the wings and in the valley of the curve. This disagreement is undoubtedly due to the finite resolution of the initial mass yield measurements and neutron measurements which were used as input data. The known fine structure of the radiochemical mass yield data is reproduced, at least qualitatively, although the peaks at masses 100 and 134 are underestimated. This again is probably due to the finite resolution of the input data. The agreement between the calculated and measured radiochemical mass yields does not test our model, but rather demonstrates the consistency between the initial mass yield measurements of Milton and Fraser[9] and the neutron yield measurements of Apalin *et al.*[7]

Independent radiochemical yields were calculated also for mass numbers 78 through 158. Yields were calculated for the five most abundant isobars of each mass number. The numerical results are too extensive to include here. They may be obtained from the authors on request. Also available are calculated initial independent yields for mass numbers 78 through 158.

Experimental results are available to compare with 47 of the calculated independent yields. The experimental data were taken from the compilation of Wahl *et al.*[2] and Coryell *et al.*[1] The more recent data of Troutner *et al.*,[13] Love *et al.*,[14] and Wahl[15] also were included. Figure 2 shows the ratios of calculated to experimental yields. The comparison is actually made for fractional yields—the ratio of the independent to the mass yield. Although the comparison shows disagreements as large as a factor of 20, the over-all agreement is better than that obtained by other means of estimating independent yields. The rms average of the natural logs of the ratios of calculated to measured

[12] H. Farrar and R. H. Tomlinson, Nucl. Phys. **34**, 367 (1962); and H. Farrar, H. R. Fickel, and R. H. Tomlinson, Can. J. Phys. **40**, 1017 (1962).

[13] D. E. Troutner, A. C. Wahl, and R. L. Ferguson, Phys. Rev. **134**, B1027 (1964).

[14] D. L. Love and P. O. Strom (private communication).

[15] A. C. Wahl, Progress Report No. TID-14466, Washington University, St. Louis, Missouri, 1962 (unpublished).

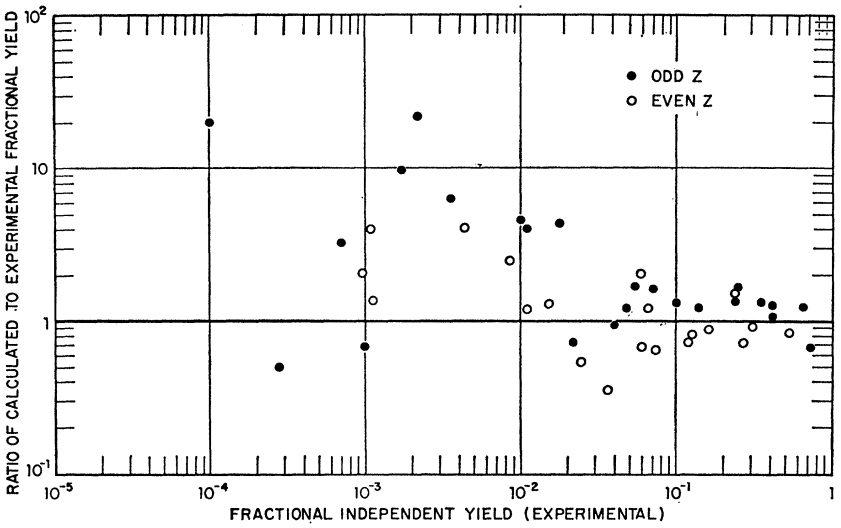

FIG. 2. Comparison of calculated and measured independent yields. The fractional independent yield, given on the abscissa, is the ratio of the independent radiochemical yield to the radiochemical mass yield. (In some cases the measured quantity we wish to compare with is a fractional chain yield—the yield of all isobars of a given mass with atomic number less than a specific value. In these cases, the comparison was made in the following manner. If the fractional chain yield was greater than one-half, the comparison was made with the fractional yield subtracted from unity. This quantity is the fractional yield for all isobars with atomic number *greater* than the specified one. For such cases, the latter number is more sensitive to errors and therefore is a better measure of the success of the calculation.) The points represent experimental measurements taken from Refs. 2, 13, 14, and 15. Even-Z nuclides are represented by open circles and odd-Z nuclides by solid circles. The ordinate gives the ratio of the fractional yield calculated in this paper to the reported measured yield.

yields is 1.0, which corresponds to a factor of 2.7 disagreement. The average log of similar ratios for the estimates in Refs. 16 and 17 is 1.8, 1.3, and 1.5. The estimates in Ref. 16 were based on the theories of Present[18] and Glendennin,[19] and those in Ref. 17 on the equal charge displacement rule. It is to be expected that the results presented in this paper would agree better with experiment, since we used an extensive amount of experimental data as input. Our work is more of an empirical nature than the previous works.

The calculation tends to underestimate the yields of even-Z nuclides, and to overestimate the yields of odd-Z nuclides (see Fig. 2). Although the correlation is not nearly strong enough to draw firm conclusions, these results suggest that even-even fragments are formed preferentially.

The average excitation energies as a function of mass number were also calculated. The results are shown in Fig. 3. These excitation energies may be combined with the neutron emission numbers to calculate the average energy used per neutron emitted. The average value is 8.4 MeV per neutron. This number includes the 1.2-MeV kinetic energy per neutron, and the energy emitted as gamma radiation following neutron emission.

The calculated excitation energies were combined with the fragment binding energies calculated by Cameron to compute the kinetic energy liberated as a function of fragment mass number. The energy balance equation is

$$T=(M-A)_{236}-(M-A)_L-(M-A)_H-\bar{W}_L-\bar{W}_N. \quad (7)$$

In this equation the quantities $(M-A)$ are the mass excesses of the light and heavy fragments, and of U^{236}, in MeV. The neutron binding energy is included in $(M-A)_{236}$. Our calculation neglects gamma-ray emission in competition with neutron emission, so this energy is neglected in Eq. (7). This point is discussed below.

The calculated kinetic energies are shown in Fig. 4. They are compared with the experimental measurements of Milton and Fraser.[9] In the region of symmetric fission there is a discrepancy between the excitation energies indicated by the neutron experiments and the kinetic energies. Since experiments are difficult to perform in this region, it is possible that some of the discrepancy is experimental. Other possible explanations are a distribution of charge radically different from the assumed one, or a very large release of gamma-ray energy in symmetric fission.

In the region where the initial yields are high, the calculated kinetic energies are 3 or 4 MeV high. The discrepancy is almost certainly due to the lack of accounting for gamma rays emitted before, or in competition with, the neutrons. The measured gamma-

[16] R. C. Bolles and N. E. Ballou, U. S. Naval Radiological Defense Laboratory Report No. TR-456, 1956 (unpublished).
[17] L. E. Weaver, P. O. Strom, and P. A. Killeen, U. S. Naval Radiological Defense Laboratory Report No. TR-633, 1963 (unpublished).
[18] R. D. Present, Phys. Rev. **72**, 7 (1947).
[19] L. E. Glendennin, C. D. Coryell, and R. R. Edwards, Nat. Nucl. Energy Ser., Div. IV, **9**, (1951) Paper 52.

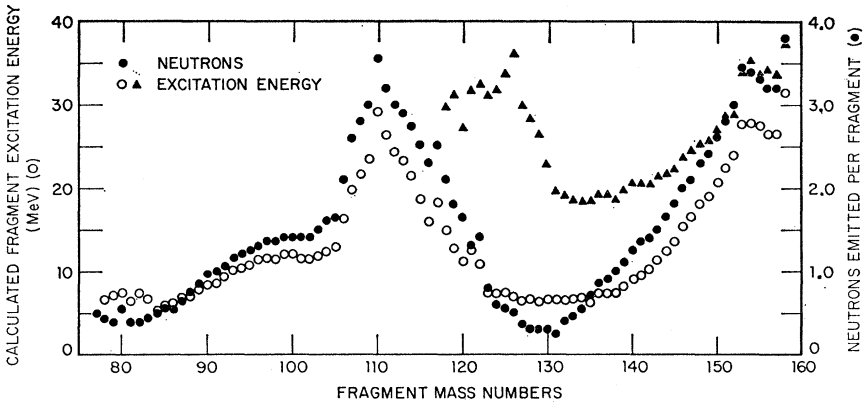

FIG. 3. Calculated fragment excitation energies. The open circles represent the average excitation energy for each fragment mass, as calculated in this paper. The triangles give the total excitation energy for each fragment pair, given for the mass of the heavy fragment. Shown for comparison (solid circles) are the neutron emission numbers measured by Apalin (Ref. 7). These numbers served as input in the calculation of the excitation energies.

ray yield 7.2±0.8 MeV per fission.[20] On the other hand our model, calculating only the gamma rays emitted after neutron emission, gives 1.4 MeV per fission. Therefore we have not accounted for 5.8 MeV of gamma rays per fission. The average calculated kinetic energy is 171.9 MeV, compared to 168.0 MeV measured.[9] The discrepancy is 3.9 MeV, which is over-accounted for by the 5.8 MeV of missing gamma-ray energy. (The discrepancies for individual fragment masses, of course, depends on how the gamma-ray energy is distributed.) This remaining discrepancy is within the errors expected in the calculation, except in the symmetric region. The errors in the calculated binding energies may be as high as 1 or 2 MeV. An error of 0.2 in the number of neutrons, which is reasonable, would cause an error of about 1 MeV in the excitation energy.

Thus, except for the symmetric region, the discrepancies in the energy balance are quite compatible with the uncertainties in the input data. It appears, however, that one must take into account the gamma rays emitted before, or in competition with, the neutrons to account for either the measured kinetic energy release or the measured gamma-ray energy release.

The last results to be compared with experiment are the probabilities P_n of emitting n neutrons per fission.

TABLE I. Probabilities P_n of emission of ν neutrons per fission event.

	Calculated	Measured (Ref. 21)
P_0	0.070	0.027
P_1	0.170	0.158
P_2	0.269	0.339
P_3	0.242	0.305
P_4	0.164	0.133
P_5	0.060	0.038
P_6	0.019	−0.001

[20] F. C. Maienschein, R. W. Peelle, R. W. Zobel, and T. A. Love, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Paper P/670.

The quantities P_n were calculated from Eqs. (5) and (6), and the results are given in Table I. The calculated results are compared with the measured values of Diven *et al.*[21] The measured values correspond to 80 keV incident neutrons, but are not expected to be substantially different from those for thermal neutrons.

The calculated probabilities give an average of 2.50 neutrons emitted per fission, compared to 2.49 for the measured probabilities. The calculated average depends only on the measurements of Milton and Fraser, and of Apalin *et al.*, and the agreement again demonstrates the accuracy of the measurements.

However, the difference between the spreads of the calculated and measured distributions is significant. The variance of the calculated distribution is 1.92, compared to 1.21 for the measured distribution. This point is discussed further in the next section.

CONCLUSIONS

In the preceding section the discrepancies in the independent yields are characterized by the logs of the ratios of the calculated to the measured yields. It is more instructive to express the discrepancies in terms of errors in those input parameters which determine the charge and energy distributions. In principle we could do this by rerunning the problem several times with small variations in the parameters. However, the length of the computer program prohibits this approach. Instead we have devised an approximate method for estimating the error in Z_0 or σ_Z needed to reproduce the observed discrepancy. For this purpose, we assume that the *radiochemical* charge distribution is approximately Gaussian:

$$f'(Z,M)\simeq[1/(2\pi\sigma_Z')^{1/2}]\exp[-(Z-Z_0')^2/2\sigma_Z'^2]. \quad (8)$$

(The primes indicate that the parameters refer to the radiochemical distribution.) With this assumption, an

[21] B. C. Diven, H. C. Martin, R. F. Taschek, and J. Terrell, Phys. Rev. **101**, 1012 (1956).

error in Z_0' appears as

$$\delta Z_0' \cong (\sigma_Z')^2 \frac{\ln(f_c/f_m)}{(Z-Z_0')}, \quad (9)$$

and an error in σ_Z appears as

$$\delta \sigma_Z' \cong (\sigma_Z')^3 \frac{\ln(f_c/f_m)}{(Z-Z_0')^2}, \quad (10)$$

where f_c and f_m are the calculated and measured fractional independent radiochemical yields. These equations give us the means to interpret the observed discrepancies in terms of errors in the midpoints or widths of the charge distributions. In the right side of Eqs. (9) and (10) we use $\sigma_Z' \simeq 0.6$ charge units, and $Z_0'(M) \simeq Z_0(M+\bar{\nu}(M))$.

The quantities $\delta Z_0'$ and $\delta \sigma_Z'$ were calculated for the 47 cases where independent yields have been measured. The rms error in the midpoint of the charge distribution is 0.21 charge units, if we attribute all the error to the midpoint of the charge distribution. If we attribute all the error to the width of the charge distribution, we get an rms error in σ_Z of 0.14 charge units.

We conclude that the discrepancies in the yields, shown in Fig. 2, can be accounted for by variations in Z_0 and σ_Z of the order of 0.2 charge units. Of course the errors may be combinations of variations in the midpoint and widths, or may be due to departures from the assumed Gaussian shape.

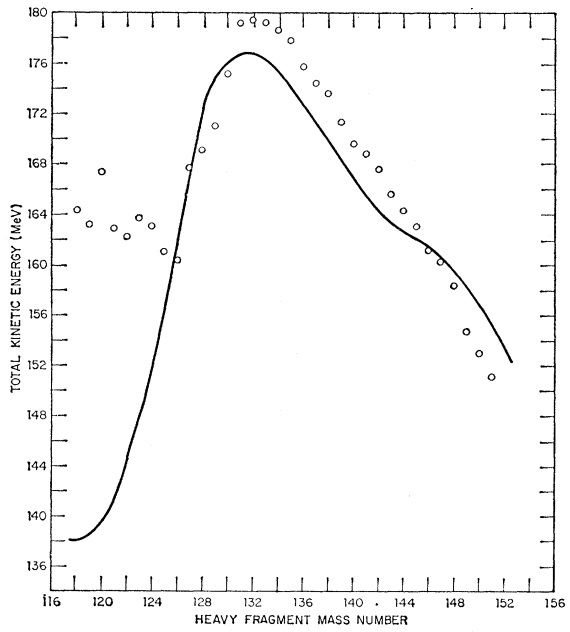

FIG. 4. Comparison of the calculated and measured kinetic energies. The open circles are the average kinetic energies of each fragment pair, plotted against the mass of the heavy fragment. The solid line represents the measurements of Milton and Fraser.[9] The calculated values were obtained by combining the excitation energy data from Fig. 3 with the semiempirical mass results of Cameron.[3]

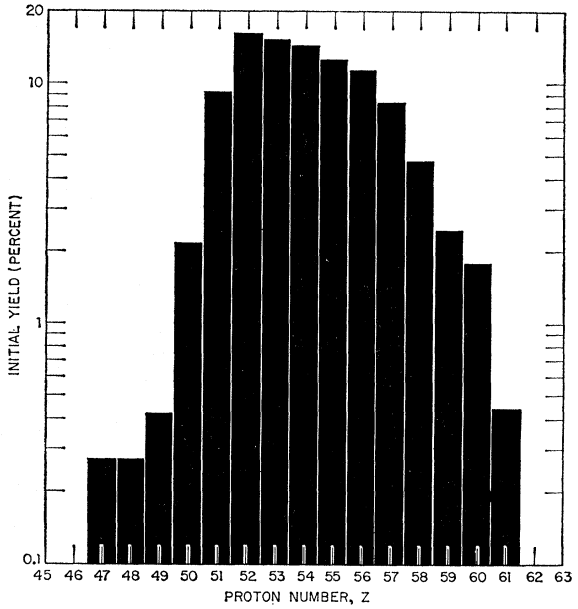

FIG. 5. Calculated initial yields versus atomic number. The ordinate gives the calculated initial yields, in percent, for fragments with each proton number from $Z=47$ to $Z=61$. The calculation did not include $Z=46$ or $Z\geq 62$. The light-fragment yields are of course the mirror image of the heavy-fragment curve.

Since variations of the order of tenths of charge units in the input parameters account for the error, it seems clear that the charge distribution given by Eq. (2) and Fig. 7 must be fairly close to the real charge distribution. While calculations of this sort cannot show the assumed distribution *uniquely* fits the data, it is difficult to see how the initial charge distribution could be substantially different from the assumed one and still give the observed independent yields.

Some particular yields are worthy of comment. We refer to the yields of As^{78}, Br^{82}, Rb^{86}, and Pm^{150}. These are represented by the four points to the right in Fig. 7, and are the basis for the estimates of Z_0 for very asymmetric fission. The calculated values for all of these numbers indicated that the dashed curve in Fig. 7 would have given better results for all four nuclides, so the dashed curve is probably a better guess for Z_0. Such an adjustment would improve the results shown in Fig. 2 and would improve the rms deviation between calculated and experimental yields. However, neither the model nor the input data are accurate enough to justify such juggling of the parameters, and the calculation was not repeated for the dashed curve in Fig. 7.

One source of error not considered in the preceding paragraphs is the error in the neutron separation and nuclear binding energies, taken from Cameron's semiempirical mass formula.[3] Actually the calculation was run twice, using Seeger's semiempirical mass formula[4] the second time. Substantially the same results were obtained. The differences in the two calculations were small compared to differences between the calculations

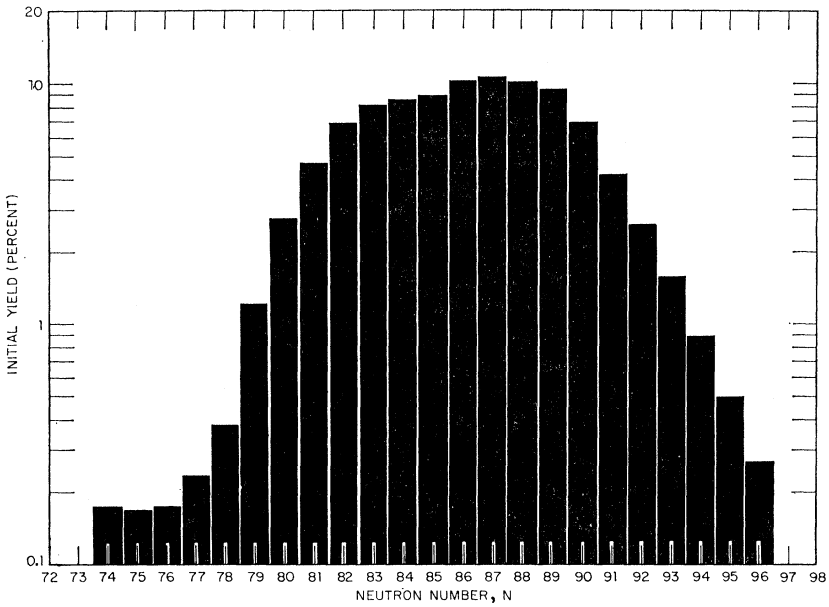

FIG. 6. Calculated initial yields versus neutron number. The ordinate gives the calculated initial yields, in percent, for fragments with neutron numbers ranging from 73 to 96. The calculation did not include neutron numbers $N=72$, 73, or $N\geq 97$.

and measurements. We conclude that the model is not sensitive to the errors in the mass formulas.

The calculated initial yields can be rearranged to give the initial yield as a function of either charge number or neutron number. These yields are presented in Figs. 5 and 6. It has often been proposed that the decrease in yield near symmetric fission is related to the shell closures at $Z=50$ and $N=82$. Figures 5 and 6 of course show decreases in this region, but they are not dramatic enough to prove the importance of the closed shells. If the yield curves have structure near closed shells, our calculation would tend to wash it out. However, Fig. 5 shows a more rapid change in yield at $Z=50$ than anywhere else in any of the yield curves. Thus there is an indication of a cutoff at $Z=50$, but certainly nothing conclusive.

The last data we discuss are the neutron emission probabilities per fission, P_n. The calculated neutron emission probabilities (Table I) give the correct mean number of neutrons per fission, but the spread about the mean is larger than the experimental spread. The calculated variance is 1.92 compared to 1.21 for the measured variance. The calculated standard deviation is therefore 1.38, about 25% larger than the measured standard deviation of 1.1.

Four simplifying assumptions could contribute to the discrepancy. First, we assumed that the variance of the excitation energy is equal to that of the kinetic energy. Second, we assumed that the ratio of the standard deviations of the light and heavy fragments is equal to the ratio of the excitation energies. Third, we assumed that the excitation energy distributions of corresponding light and heavy fragments are independent of each other. Fourth, we assumed that neutrons are emitted with a constant energy.

The third assumption is likely to be seriously in error. It is more reasonable to expect the excitation energies of a fragment pair to be correlated negatively. That is, if one fragment has an excess of excitation energy, the other fragment tends to be deficient in excitation energy. However, introduction of such a correlation alone will not resolve the discrepancy.[22]

The fourth assumption, concerning the energy distribution of the evaporated neutrons, is more likely to be the major cause of the discrepancy. The neutron kinetic-energy distribution is of course continuous, and should vary with excitation energy as well as with Z and M. A first-order correction was made to the variance of the excitation energy distribution for this effect (see Appendix B). However, the work of Leachman[23] and of Jackson[24] show that the kinetic-energy distribution will have a large effect on the number of neutrons emitted at a given excitation energy, and our simple correction is probably inadequate. We conclude that a more sophisticated treatment of the neutron evaporation process would be needed to predict the neutron multiplicities correctly with our model.

We note also that assumption four, above, may account partially for our low value for gamma-ray energy release. Leachman[23] calculated the neutron

[22] Quantitatively we could introduce such a correlation by replacing Eq. (B1) with $\sigma_{W_L}^2+\sigma_{W_H}^2+2C(W_L,W_H)=\sigma_T^2$. The quantity $C(W_L,W_H)$ is a correlation function which in this case would be negative. If Eq. (B1) were thus modified, Eq. (5) would have to be similarly modified. With such a correlation the variances $\sigma_{W_I}^2$ and $\sigma_{W_n}^2$ must be made larger to fit σ_T^2, and the modification of Eq. (5) will be offset by a greater spread in neutron emissions from the individual fragments. This is the basis of the statement above that a correlation between the excitation energies cannot in itself account for the observed discrepancy.

[23] R. B. Leachman, Phys. Rev. **101**, 1005 (1956).

[24] J. D. Jackson, Can. J. Phys. **34**, 767 (1956).

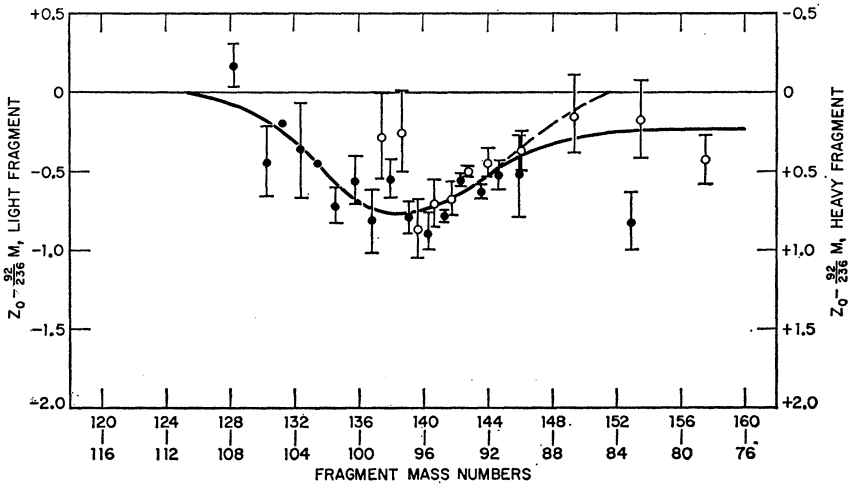

FIG. 7. Semiempirical values of the most probable charge Z_0. The quantity Z_0 is an estimate of the average number of protons for *initial* fragments of mass M. The values have been presented as $Z_0-(92/236)M$, in the manner of Wahl (Ref. 2), in order to emphasize the deviation from proportionate charge and mass division of the fragments. The points are the estimates of Z_0 given by Wahl (Ref. 2), except that they are adjusted to the mass values indicated by Apalin's neutron emission measurements (Ref. 7). The solid line represents the values of Z_0 used in the calculation in this paper. The dashed line gives a modification of the curve, which is suggested by the results of the calculations.

energy distribution using the statistical model, and calculated an average gamma-ray energy release of 3.8 MeV, a result more reasonable than our 1.4 MeV. The indication is that a better treatment of the neutron evaporation process would be needed to get accurate estimates of the gamma-ray energy release from our model.

We now summarize the main points of interest in the calculation. (i) The calculation indicates an average release of 1.4 MeV per fission following neutron emission. On the other hand, the measured gamma-ray yield is 7.2±0.8 MeV per fission. The indication is that about 6 MeV of gamma-ray energy is emitted before, or in competition with, the neutrons. This number is somewhat higher than the energy balance indicates, but is within the expected errors in the calculation. A comparison with the work of Leachman[23] indicates that our calculated gamma-ray energy release would be improved substantially with a more adequate treatment of the neutron evaporation process. (ii) The energy balance, as shown by comparing calculated and experimental kinetic energies (Fig. 4) shows some interesting discrepancies. In the region of symmetric fission, about 35 MeV is not accounted for. This deficit is too large to be covered by the normally expected errors in the measurements and the calculations. In the regions of the peaks of the yield curves, the calculation gives a surplus of about 3 MeV. Since we have not accounted for 5.8 MeV of gamma-ray energy per fission, this discrepancy is not unreasonable. Various input data can be expected to be in error by as much as a few MeV, so the over-all agreement is satisfactory, except in the region of symmetric fission. (iii) A comparison of the calculated and measured independent yields indicates that the errors in the assumed charge distribution are of the order of a few tenths of a charge unit. While the calculation does not prove that the assumed distribution is correct, it is difficult to see how it could be seriously in error. There is an indication that the dashed line in Fig. 7 more nearly represents the real charge distribution than does the assumed one (solid line). (iv) The comparison of independent and calculated independent radiochemical yields also suggests that even-even fragments may be preferentially formed in fission. However, the calculations are not nearly accurate enough to draw any firm conclusions.

Many of the features of the data already have been pointed out in the referenced papers. Also, many of the features of the energy balance and the discrepancies in the observed and estimated gamma-ray emission are given in Halpern's article.[25] Since the model uses various fragment, particle, and energy yields as input data, the results would no doubt be improved by better experimental data. Of particular interest is the puzzling discrepancy in the energy balance for near-symmetric fission, and the suggested preference for formulation of even-even primary fragments. The model will be revised and updated as better data become available.

APPENDIX A: COMPARISONS OF METHODS OF DETERMINING THE MOST PROBABLE Z

The values of Z_0, the most probable charge for a given initial mass number, were determined from Wahl's work, with a slight modification. In Wahl's method the initial mass corresponding to an empirically determined Z_0 is determined from the radiochemical mass and the neutron emission curve:

$$M(Z_0)-\bar{\nu}(M)=M_{\text{rad}}(Z_0). \qquad \text{(A1)}$$

[25] I. Halpern, Ann. Rev. Nucl. Sci. 9, 245 (1959).

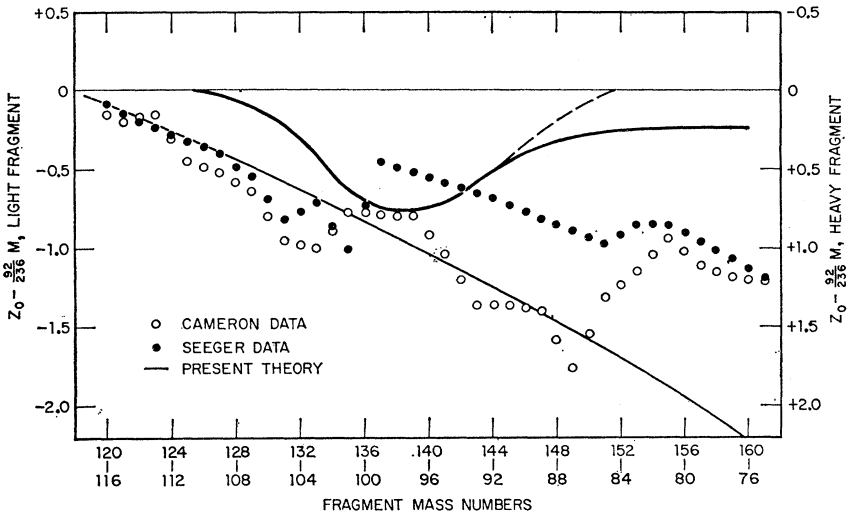

FIG. 8. Comparison of estimates of the most probable value of Z. The method of presentation is the same as in Fig. 7. The heavy solid line is the empirical Z_0 curve given in Fig. 7, and is included here for comparison. The points are estimates of Z_0 made from semiempirical mass data, assuming that the fragments divide in such a way as to maximize the total binding energies of the fragments. The open circles were obtained using Cameron's semiempirical mass data (Ref. 3), and the solid circles were obtained using Seeger's semiempirical mass data (Ref. 4). The light line represents the values of Z_0 estimated by Present.

The quantity $M_{\text{rad}}(Z_0)$ is the radiochemical mass corresponding to Z_0. Wahl used Terrell's values for $\bar{\nu}(M)$. In this work we used instead Apalin's values of $\bar{\nu}(M)$. The effect is to shift the points along the abscissa by a fraction of a mass unit. The revised curve is shown in Fig. 7. The points in Fig. 7 are the values of Z_0 given by Wahl,[2] and two values determined by Love's work.[4]

The above gives the method used to compute Z_0 for use in the computation of the main body of the paper. It is also possible to estimate Z_0 from the semiempirical mass data, using various prescriptions. We tried calculating Z_0 by two of these methods. The results are included here for comparison with the empirical values of Z_0.

The first prescription we tried was to assume that the most probable Z_0 is that which maximizes the binding energy available for kinetic and excitation energy of the fragments. The Z_0's based on this prescription were estimated as follows. The semiempirical binding energies of odd-mass fragment pairs were summed and tabulated for each mass number. The sums of the binding energies of the pair Z, 92-Z are well approximated by a parabola in Z. The peak of the parabola represents the maximum binding energy. A parabola was fitted to the three fragment pairs with the highest binding energies, and Z_0 was taken to be the value corresponding to the highest binding energy. The situation for even-mass fragment pairs is complicated by pairing effects. We therefore determined Z_0 for even-mass pairs by linear interpolation of the odd-mass results. This corresponds to ignoring the pairing energy. The most probable Z_0's determined in this manner are shown in Fig. 8. Determinations were made both for Seeger's mass values and Cameron's mass values. As the figure shows, the two determinations do not agree well with each other or with the empirical values of Z_0.

The second prescription tried was to assume that the most probable Z is that which maximizes the total excitation energy of the fragment pair. The excitation energy is given by

$$W=B(236)-B(Z_1,Z_2)-T. \qquad \text{(A2)}$$

In this equation W is the excitation energy of the fragment pair, $B(236)$ is the binding energy of U^{236}, and $B(Z_1,Z_2)$ is in the total binding energy of the fragment pair. To maximize W with respect to Z we must first assume a reasonable form for the Z-dependence of the kinetic energy, T. We assumed that $T=kZ(92-Z)$, where k is a constant determined from the experimental kinetic-energy data. The assumption is equivalent to assuming that the kinetic energy is given by the Coulomb energy of two spherical fragments with charges Z, $(92-Z)$, separated by a constant distance. The expression for W is then maximized with respect to Z. While the expression for the kinetic energy is only approximate, it turns out that its effect on the position of Z_0 is small. Therefore the assumption is considered to be adequate. The values of Z_0 determined in this fashion are shown in Fig. 9. Again, the two calculations (for the two sets of mass values) do not agree well with each other or with the empirical curve. The general trend is more in line with the empirical values than is the first estimate, however.

Still another prescription for determining Z_0, based on theoretical differences between neutron and proton densities near the nuclear surface, is given by Present.[18] We have calculated Z_0 versus M using his formula, and the result is shown in Fig. 8 as a light line.

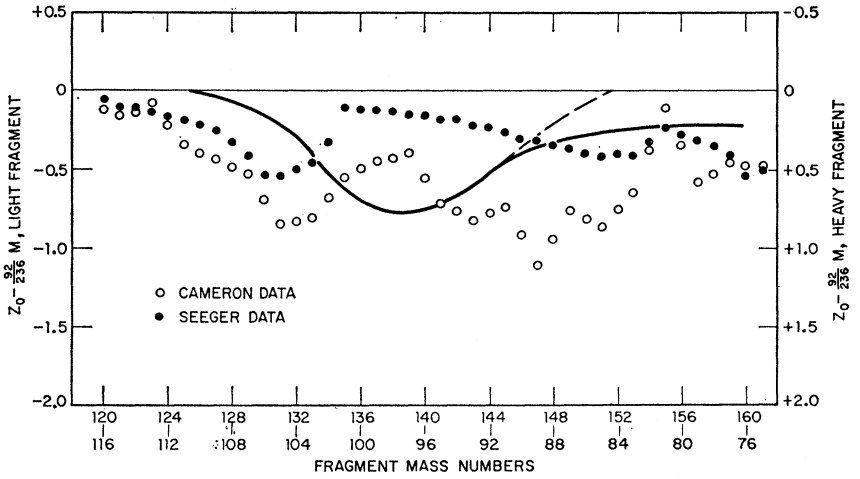

FIG. 9. Comparison of estimates of the most probable Z. The method of presentation is the same as in Fig. 7. The solid line is the empirical Z_0 curve given in Fig. 7, and is included here for comparison. The points are estimates of Z_0 made from semiempirical mass data, assuming that the fragments divide in such a way as to maximize the total excitation energies of the fragments. The open circles were obtained using Cameron's semiempirical mass data, and the solid circles were obtained using Seeger's semiempirical mass data.

To summarize, Figs. 8 and 9 show that none of the prescriptions reproduce the empirical curve. Comparison with the quantities $\delta Z_0'$ computed from Eq. (9) further indicates that the calculated values of Z_0 would increase the discrepancy between measured and calculated independent yields. Also, the methods based on maximizing the energy depend critically on which semiempirical mass formula is used. We conclude that the most probable values of Z cannot be predicted either by maximizing the binding energy or the excitation energy. The discrepancies do not necessarily mean that the basic assumption is wrong, but could be due to uncertainties in the semiempirical mass data.

APPENDIX B: CALCULATION OF EXCITATION ENERGY VARIANCES

We assumed that the total spread in excitation energy of a given pair of fragments corresponds exactly to their kinetic-energy spread. This assumption is not completely correct. Some of the spread in kinetic energy is due to the different binding energies of different isobars with a given mass number. However, the semiempirical mass data show that the spread due to binding energies is only of the order of 1 MeV, which is small compared to the total spread.

The kinetic-energy-distribution measurements of Milton and Fraser were used. Since the measured distributions are roughly Gaussian, the excitation energy distribution is assumed also to be Gaussian.

With the variance of the total excitation energy of each pair determined, it remains only to decide how the excitation energy is distributed between the fragments. This distribution is fixed by two assumptions. First, we assume that the variations about the mean of the light-fragment excitation energy are independent of those of the heavy fragment. With this assumption, the variances add to form the total variance:

$$\sigma_{W_L}^2+\sigma_{W_B}^2=\sigma_T^2. \tag{B1}$$

Second, we assume that the ratio of the standard deviations for the light and heavy fragments is proportional to the ratio of the average number of neutrons emitted. Since the number of neutrons emitted is roughly proportional to the excitation energy, this assumption is equivalent to setting the *fractional* standard deviations of the light- and heavy-fragment excitation energies equal to each other. We have no justification for this assumption, except that it is plausible. Also, some data on neutron emission versus kinetic energy for Cf^{252} (Ref. 26) seem to indicate that for complementary fragments the one with higher neutron emission accounts for most of the variation in kinetic energy, and therefore accounts for most of the variance.

Another correction to the variance was made to partially account for the neglect of the neutron kinetic distribution. Since the variance of the neutron kinetic-energy distribution is 1.27 $(MeV)^2$, this amount was added to each excitation energy variance used in Eq. (3).

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