FEB 24 1965

MASTER

PATENT CLEARANCE OBTAINED. RELEASE TO
THE PUBLIC IS APPROVED. PROCEDURES
ARE ON FILE IN THE RECEIVING SECTION.

THE ENERGY BALANCE IN FISSION AND
MONTE CARLO CALCULATIONS ON PROMPT-NEUTRON EMISSION¹

G. E. Gordon (Assoc. Professor of Chemistry)

and

N. K. Aras² (NATO Predoctoral Fellow)

Department of Chemistry and Laboratory for Nuclear Science

Massachusetts Institute of Technology

Cambridge, Massachusetts, USA

ABSTRACT

We have treated the evaporation of neutrons from primary fission fragments by the Monte Carlo method. The total energy release E_T for fission of U^{236*} into a given pair of complementary, primary fragments is obtained from semi-empirical mass formulas. The average kinetic energy, $\bar{E}_K$, for a given mass ratio of division is obtained from the results of instrumental studies with

¹ This work was in part supported by the U.S. Atomic Energy Commission under contract AT(30-1)905

² Holder of a Fellowship from the N.A.T.O. Scientific Committee, Ankara, Turkey. Present address: Oak Ridge National Laboratory, Oak Ridge, Tennessee.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

the average sum of the fragment excitation energies, $\bar{E}_x$, being given by $E_T - \bar{E}_K$. The division of $\bar{E}_x$ between light and heavy complementary fragments is determined by a completely free parameter. The latter is adjusted to yield the experimental ratio of average number of prompt neutrons emitted by the light fragment, $\bar{\nu}_L$ to that of the heavy fragment, $\bar{\nu}_H$. Dispersion of the excitation energies is determined by dispersion in kinetic energies.

Primary mass yields are taken from instrumental fission studies. Charge distribution is assumed Gaussian (with $\sigma = 0.62$) about the most-probable value, Z_p , for each mass. The latter are adjusted to give agreement with experimental results.

Nuclear level densities are assumed proportional to $\exp[2a^{1/2}(E-\delta)^{1/2}]$, with $a = A/13$; δ is a correction for odd-even effects. Monte Carlo iterations are performed by a computer for all primary fragments of appreciable yield.

We have obtained good agreement with several important experimental quantities: the dependence of $\bar{\nu}_A$ upon mass number, the neutron kinetic energy spectrum, and the average prompt γ -ray energy release. In regions of large $\bar{\nu}_A$ the secondary charge-distribution curves are much broader than for the primary fragments and sometimes asymmetric, with the low-Z side broader than the high-Z side. Dispersions in ν 's are so large as to suggest that our assumption that $\bar{E}_K$ is the same for all charge divisions for a given mass division must be relaxed to obtain agreement with experimental data.

THE ENERGY BALANCE IN FISSION AND
MONTE CARLO CALCULATIONS ON PROMPT-NEUTRON EMISSION ¹

G. E. Gordon (Assoc. Professor of Chemistry)
and
N. K. Aras ² (NATO Predoctoral Fellow)

Department of Chemistry and Laboratory for Nuclear Science
Massachusetts Institute of Technology
Cambridge, Massachusetts, USA

1. Introduction

The initial aim of these calculations was to investigate the effect of prompt-neutron emission upon the shape of the charge-distribution curve, i.e. independent fractional chain yield (designated fcy) as a function of nuclear charge Z for a given mass number A . The charge distributions of the primary fragments (i.e. prior to prompt-neutron emission) are of interest for

¹ This work was in part supported by the U. S. Atomic Energy Commission under contract AT(30-1)905

² Holder of a Fellowship from the N.A.T.O. Scientific Committee, Ankara, Turkey. Present address: Oak Ridge National Laboratory, Oak Ridge, Tennessee.

comparison with theoretical predictions, but at present there are no measurements of primary distributions. A considerable body of data on the charge distributions of secondary fragments has been amassed, principally by radiochemical measurements of the independent yields of shielded and quasi-shielded nuclides [1,2]. Unfortunately, it would not be possible, even if the neutron multiplicities of each of the primary-fragment nuclides were known, to infer directly the primary charge distributions from those of the secondary fragments. Thus we have taken the approach of assuming reasonable primary distributions, performing Monte Carlo neutron-evaporation calculations to estimate neutron multiplicities for each primary nuclide, and computing the resulting secondary mass and charge distributions for comparison with experimental data. We were interested in the magnitude of difference between most-probable charge, Z_p , for the primary and secondary fragments of a given A . Also, we wanted to learn whether or not the primary-to-secondary transformation introduces distortions into the charge-distribution curve (e.g. broadening, skewing, etc.).

In the course of work, we have found that the calculations are useful for a detailed accounting of the energy released in the fission process. Also, it appears that with the calculations it may be possible to determine the correct choices (or, at least, discard some incorrect ones) among sets of a priori equally reasonable assumptions about features of the fission process which are not amenable to direct experimental study. For example, it may be possible to find out if there is positive, negative, or no correlation between the excitation energies of complementary fragments of a given event.

Some of the calculations reported here have been used previously in the interpretation of experiments on isomer-yield ratios [3] and ranges of fission fragments [4].

2. Description of the calculations

In constructing a model for the calculations, one finds a number of points about which one can make any one of several a priori equally reasonable assumptions. Often there are no direct experimental data upon which to base a choice among the set of reasonable assumptions. We have chosen a set of reasonable assumptions and compared the results of the calculations with a wide variety of experimental data, finding generally good agreement with most. In some cases, we have also performed the calculations with alternate choices of assumptions and we discuss below the results of such variations. In later works, we will investigate the implications of additional variations of the assumptions. It is necessary to investigate these other assumptions to determine if a given set which gives reasonably good agreement with most experimental data is unique or merely one out of several sets which give agreement.

2.1 Organization of the calculations

In the calculations we treat a region of primary fragments up to 18 mass units long and 10 charge units wide. For each mass chain, we first calculate the primary fractional chain yield, fcy, of each isobar according to

$$fcy = \frac{1}{\sqrt{\pi C}} \exp [-(Z - Z_p)^2 / C] , \quad (1)$$

where C is the charge-dispersion parameter. All isobars having f_{cy} below some given limit, generally 10^{-6} , are rejected as giving negligible contribution to total neutron emission, etc., and the f_{cy} 's of remaining isobars are renormalized to a sum of unity. The independent yield of each isobar is computed as $f_{cy} \cdot Y(A)$, the primary yield of mass chain A as obtained from instrumental fission studies.

A given number of iterations is performed for each of the isobars which has not been rejected. The first step of each iteration involves selection of an excitation energy of the primary fragment, via the Monte Carlo rejection technique [5], from the distribution obtained as discussed in Sect. 2.2. The evaporation of neutrons is followed to completion as described in Sect. 2.3. After each neutron is emitted and at the end of each iteration, various quantities, e.g. neutron kinetic energies, γ -ray energy release, number of neutrons emitted, are weighted by the independent yield of the primary fragment and stored in appropriate histograms for later treatment and inclusion in the output data. When primary fragment (Z, A) emits n neutrons in an iteration, the secondary yield of species $(Z, A-n)$ is increased by $f_{cy}(Z) \cdot Y(A)$. At the end of each mass chain, the spectrum of neutron kinetic energies is transformed to the laboratory system (assuming isotropic emission from a fragment of mass A having a uniform velocity corresponding to the average kinetic energy release for that chain), weighted by $Y(A)$, and added to the overall laboratory-system neutron kinetic energy spectrum.

After all mass chains in the region have been treated, the various stored results are appropriately grouped and renormalized, average values

and dispersions are computed, and the results printed. It requires about 1.2 min to treat an 18 mass unit region for 50 iterations per primary nuclide on an IBM-7094 computer.

2.2 Excitation-energy distributions

We assume that the average sum of the excitation energies, $\bar{E}_x$, of complementary light (A_L, Z_L) and heavy (A_H, Z_H) fragments from fission of U^{236*} is given by

$$\bar{E}_x = E_T - \bar{E}_K (A_H/A_L), \quad (2)$$

where the total energy release, E_T , is given by

$$E_T = \Delta M(U^{236*}) - (\Delta M_L + \Delta M_H). \quad (3)$$

The mass excesses, ΔM , are obtained from mass equations and the average total kinetic energy release as a function of mass ratio, $\bar{E}_K (A_H/A_L)$, is taken from time-of-flight, semiconductor-detector, or range measurements of fragment kinetic energies. Here we have had to make a choice among reasonable assumptions in the absence of direct experimental observations. From mass equations one finds that E_T for a given A_H/A_L has approximately parabolic dependence on Z_L (or Z_H) with the maximum value generally occurring at a Z_L value near Z_{p_L} as shown for mass ratio 139/97 in the lower part of Fig. 1. The assumption of Eq. (2) implies that $\bar{E}_K$ is the same for all charge division, Z_H/Z_L , for a given mass division. Changes in E_T with changing Z_H/Z_L thus require corresponding changes in $\bar{E}_x$, as illustrated in Fig. 1. We could have made the extreme opposite assumption, namely that $\bar{E}_K$

changes with Z_H/Z_L and $\bar{E}_x$ remains approximately constant.

The assumption of Eq. (2) requires the shape of the E_x distributions to be those of the E_K distributions for each mass division. As the latter appear to be approximately Gaussian [8], we assume that the E_x distributions are Gaussian with dispersions given by

$$\sigma_{E_x} = \sigma_{E_K} \quad (4)$$

Assuming no correlation between the excitation energies, E_L and E_H , of the light and heavy fragments, respectively, in each event we have

$$\sigma_{E_L}^2 + \sigma_{E_H}^2 = \sigma_{E_x}^2 \quad (5)$$

On general statistical grounds we assume that

$$\frac{\sigma_{E_H}^2}{\sigma_{E_L}^2} = \frac{\bar{E}_H}{\bar{E}_L} \equiv F(A_H/A_L) \quad (6)$$

Again, in the case of Eq. (5), we have no direct experimental check on the assumption. If there is a negative correlation between E_L and E_H , the σ_{E_L} and σ_{E_H} values could be much larger than indicated by Eq. (5). On the other hand, violations of the assumption of Eq.(2) as discussed above would cause the σ_{E_L} and σ_{E_H} values to be smaller than those obtained from Eqs. (4) and (5). In principle, the excitation-energy-splitting parameter, $F(A_H/A_L)$, should be obtained from a complete theory of the fission process. However, in the absence of any completely satisfactory theory, we have left $F(A_H/A_L)$ as a free parameter to be varied until approximate agreement is obtained with the experimental value [9] of $\bar{\nu}_H/\bar{\nu}_L$, the ratio of average number of prompt neutrons emitted by the heavy fragments to that of the complementary light fragments as a function of mass ratio, A_H/A_L .

2.3 Treatment of Neutron Evaporation

After the excitation energy of a given primary fragment has been picked, de-excitation by the emission of neutrons and γ rays is treated by the Monte Carlo method described by Dostrovsky, Fraenkel, and Friedlander [10], except that we ignore charged-particle emission. According to this treatment, level-densities of the residual nuclei as a function of excitation energy, E , are assumed to be given by

$$w(E) = \text{Const.} \exp \left[2\sqrt{a(E-\delta)} \right], \quad (7)$$

in which δ is the "characteristic level" or pairing-energy correction to account for reduced level density when the number of protons and/or neutrons in the residual nucleus is even. In most of the calculations we have assumed a to be given by A/γ , where γ is a parameter which is generally found to be in the range 8 to 13 MeV in other types of experiments.

This treatment assumes that a neutron is emitted whenever possible except that no neutron emissions may leave the residual nucleus with an excitation energy less than δ if δ is non-zero. Thus, the "effective" neutron separation^{energy} for nuclide Z, A is $S_n(Z, A) + \delta(Z, A-1)$. Any excitation energy remaining after all possible neutrons have been emitted is assumed to be given off as prompt γ rays.

3. Results of Calculations

3.1 Choice of input parameters

The parameters for most of the calculations whose results are given

below are as follows:

(1) Kinetic energy release and dispersion. Values for $\bar{E}_K$ and σ_{E_K} over most of the fission-product region were taken from the time-of-flight data of Milton and Fraser [8] and the semiconductor-detector measurements by Gibson et al. [7], respectively. In the low-yield, symmetric region, where instrumental studies are most subject to error, we have used kinetic energies based on ranges of specific fission products in aluminum [4]. Note, however, the special problem of kinetic energies discussed below in Sect. 3.2.

(2) Primary yield-mass curve. Over most of the fission-product region primary-mass yields were taken from the work of Milton and Fraser [8]. In the low-yield regions, where we were unable to read the yields from Milton and Fraser's linear plot, values were taken from preliminary results (not corrected for mass-dependent pulse-height defect) of semiconductor-detector experiments by Gibson et al. [11]. The latter were used for mass chains 75 to 79, 112 to 124, and 157 to 161. The values used are shown below in Fig. 10.

(3) Primary charge-distribution curves. We have assumed that f_{cy} as a function of Z for primary fragments of mass A is given by Eq. (1), with C a constant for all masses, equal to 0.94 for most calculations, and equal to 0.4 for one calculation. As starting values we used the primary Z_p values suggested by Wahl et al. [2]. Later we made slight adjustments of the primary Z_p values to improve the agreement between calculated and experimental Z_p values for the secondary fragments. The final values used are shown below in Fig. 7.

(4) Mass excesses and pairing-energy corrections. All mass-excess values were taken from Seeger's semiempirical mass formula [6]. The level-density pairing correction, δ of Eq. (7), was taken as 0 for odd-odd, δ_Z for even-Z, odd-N, δ_N for odd Z, even N, and $\delta_Z + \delta_N$ for even-even nuclides. The values of δ_Z and δ_N were assumed to be a smooth function of A and were taken from the evaluation of pairing effects on masses by Seeger [6].

(5) Level-density parameter. The level-density parameter, a of Eq. (7), was assumed equal to A/γ . Initially a value of $\gamma = 8$ MeV was used, but slightly better agreement between calculated and experimental neutron kinetic energy spectra was obtained with $\gamma = 13$ MeV, which was used for all calculations reported here.

3.2 Excitation-energy cutoff - effect on kinetic energies.

It is necessary to cut off the $E_{L,H}$ distributions at some energies above and below the most-probable E_L (or E_H) at which the probability becomes very small. We have applied the cutoff at $\bar{E}_{L,H} \pm 3\sigma_{L,H}$ at which the probability is about 1% of the peak value. However, the cutoff on the low-energy side often falls in the region of negative excitation energies as shown for the example of $A = 97$ in Fig. 2. Since we reject all negative excitation energies, the histogram of "acceptable" excitation energies may be quite asymmetric.

The zero-cutoff causes no difficulty in the evaporation calculations, but implies an E_K distribution different from that assumed for the input data. As excitation energy is cut off on the low-energy side, the implied E_K distribution is cut off on the high-energy side. Both the $\bar{E}_K$ and σ_{E_K} of the implied distributions are smaller than the input values

as shown at the right of Fig. 2. We have exaggerated the sharpness of the E_K cutoff in Fig. 2 by assuming E_H always has the value $\bar{E}_H$. In the actual calculations E_L and E_H are assumed to be uncorrelated; thus, the E_K cutoff will be rounded off as shown below in Fig. 4. Fortunately, the cutoff problem is made less serious by the fact that, in general, the most-probable charge division is the one having the highest E_T and, thus, the least serious cutoff. This may be seen by considering the fcy curve for mass 97 in Fig. 1 and the E_L distributions of Fig. 2.

The program was modified to allow it to store and print out information on the implied E_K distributions. As expected, the resulting $\bar{E}_K$ and σ_{E_K} values were generally lower than the input values, in the worst cases by as much as 3 and 2 MeV, respectively. In order to cause the implied E_K distributions to agree with our choice of experimental values we have raised the input values of $\bar{E}_K$ and σ_{E_K} as shown in Fig. 3. The implied $\bar{E}_K$ and σ_{E_K} values agree well with the experimental values in most mass regions, but we have allowed the σ_{E_K} values in the symmetric region to remain below the curve obtained by Gibson et al [7], as most kinds of experimental errors in this region would tend to increase the observed values.

In Fig. 4 we show histograms of the implied E_K distributions for three different mass splits. For mass 97, we compare the calculated curve with the experimental data obtained by Milton and Fraser [8]. The histogram for $A = 104$ illustrates the case of a large difference between the input and experimental $\bar{E}_K$ values and that for $A = 119$ shows

a typical result in a region of high σ_{E_K} . The histogram for mass 97 is in good agreement with the experimental data. Many of the calculated histograms show asymmetries of the type present in that for mass 104. We are unable to determine whether or not this asymmetry is in disagreement with experimental results as, aside from the mass-97 data shown, there are no recent data on kinetic energies presented in this form. In older work on spontaneous fission of Cf^{252} [12], some of the E_K distributions for various ranges of mass ratio of division show asymmetries of this type.

3.3 Gamma-ray energy release

In Fig. 5 we plot the average γ -ray energy release, $\bar{E}_{\gamma L, H}$, per fragment as a function of A and the sum, $\bar{E}_{\gamma \text{sum}}$ of $\bar{E}_{\gamma L}$ and $\bar{E}_{\gamma H}$ for complementary mass numbers as a function of A_L . There have been no studies of $\bar{E}_{\gamma \text{sum}}$ as a function of A_H/A_L for fission of U^{236*} . In the case of Cf^{252} spontaneous fission, there does not appear to be any strong dependence of $\bar{E}_{\gamma \text{sum}}$ on mass number [12]. In regions where the number of γ rays is small, the γ ray energies are high. Our overall weighted value of $\bar{E}_{\gamma \text{sum}}$ is 7.66 MeV in excellent agreement with the experimental value of 7.2 ± 0.8 MeV [13]. The calculated value of 7.66 MeV includes the energy of long-lived isomeric states having half lives so long that their decay energies are not included in the 7.2-MeV experimental value.

Previous calculations on the dissipation of fragment excitation energies have predicted $\bar{E}_{\gamma \text{sum}}$ values of 3.8 [14] and 4.9 MeV [15], considerably smaller than the experimental value. The main reason for the

larger $\bar{E}_{\gamma\text{sum}}$ values obtained in our work is the use of the pairing-energy correction for level densities. The excitation energy required for neutron emission is $S_n + \delta$ rather than merely S_n . Thus, with the use of δ 's, more of the excitation energy appears as γ rays and less as neutron evaporation, improving the calculated fit to $\bar{E}_{\gamma\text{sum}}$ and $\bar{\nu}_A$. In one calculation in which all δ values were set equal to zero, the overall average $\bar{E}_{\gamma\text{sum}}$ value was 5.03 MeV.

The use of δ 's, although not intended for that purpose, has in part the same effect on $\bar{E}_{\gamma\text{sum}}$ as a correction for angular momentum of the fragments. Prompt γ -ray angular distributions [16] and isomer-yield ratios [3] indicate high average spins (5 to 8 $\hbar$) of primary fragments, even in thermal-neutron fission. High angular momentum tends to favor γ -ray emission relative to neutron evaporation at excitation energies not far above S_n [17]. If we use the simple assumption that the angular momentum is present as collective rotation, "unavailable" for neutron evaporation, and emitted after evaporation as γ rays, the increase in $\bar{E}_\gamma$ per fragment would be about 0.4 to 1.2 MeV for assumed fragment moments of inertia of the rigid-body value (J_{rig}) or $0.3 J_{\text{rig}}$, respectively. Thus, the rotational effects could be large enough to give agreement with the experimental value of $\bar{E}_{\gamma\text{sum}}$, but they would not have the odd-even properties of the δ values that seem to be required to give smooth charge-distribution curves for even-mass secondary fragments as discussed below.

3.4 Neutron kinetic energy spectrum

In Fig. 6 we compare the calculated and experimental [18] laboratory-system kinetic energy spectra. The agreement between is generally good,

although in the high-energy, linear portion, the calculated spectrum drops off slightly faster with energy than the experimental one. The curve of Fig. 6, obtained with $\gamma = 13$ MeV, is in better agreement with the experimental data than that calculated with $\gamma = 8$ MeV for which the downward slope at high energies was greater. Better agreement with the experimental curve could probably be obtained by use of a γ value greater than 13 MeV, but we felt constrained to the range $8 \leq \gamma \leq 13$ MeV indicated by most analyses of the spacings of neutron-capture resonances. The value of $\gamma = 13$ MeV is in reasonable agreement with Terrell's analysis of the gross fission neutron spectrum which yielded the result $a = 12 \pm 2 \text{ MeV}^{-1}$ corresponding to $\gamma = 10.5 \pm 1.5 \text{ MeV}$ [15].

3.5 Neutron multiplicities and charge-distribution curves

In the lower part of Fig. 7 we show the calculated points for $\bar{\nu}_A$ as a function of A as compared with the experimental values [9]. The agreement is quite good except that the calculated points are slightly low in the region $100 \leq A \leq 106$ and generally high by about 0.15 neutron for $A \leq 138$. Improvements could perhaps be obtained by adjustment of $F(A_H/A_L)$ values, but we felt the fit was good enough for our purposes. Fluctuation of $\bar{\nu}_A$ values and related quantities is mainly due to statistical fluctuations in the Monte Carlo calculations. For most calculations, we ran only 50 iterations per primary nuclide.

Between $A = 108$ and 129 there are no experimental values of $\bar{\nu}_A$ for U^{236*} fission. Milton and Fraser [8] assumed a $\bar{\nu}_A$ curve in that region based largely on Cr^{252} data. Their assumed curve, which rises to ~ 3.7 at $A = 120$, was in thermodynamic agreement with their $\bar{E}_K$ data and E_T from mass equations. In previous work [4], we found the E_K deficit at symmetric fission to be only 21 MeV instead of 36 MeV as determined by Milton and Fraser. Thus, the $\bar{\nu}_A$ values thermodynamically consistent with our

measurements of $\bar{E}_K$ are considerably lower than those used by Milton and Fraser.

In Fig. 7 we show the primary and secondary Z_p values from the calculations and all known experimental secondary Z_p values. As shown, we have adjusted slightly the primary Z_p values suggested by Wahl et al. [2] to obtain better fits to the experimental secondary values. The calculated secondary Z_p values are actually weighted average charges, $\bar{Z}$, for each chain. As no serious asymmetries have been found in the secondary charge-distribution curves, we did not feel it necessary to do curve fitting at each mass number to obtain true Z_p values. Experimental secondary Z_p values were taken from the paper by Wahl et al. [2], with additional values for $A = 112$ [19], 127 [20], and an upper limit for $A = 121$ [21]. The agreement between calculated and experimental values is very good except for $A = 96, 97$, and 98 . The experimental points may be in error because of isomerism in Nb^{96} and Nb^{97} , and the large uncertainty for Tc^{98} (see reference [2]).

Secondary fcy values for three mass chains are shown in Fig. 8. For comparison we have drawn curves calculated according to Eq. (1) with $C = 0.94$ and Z_p values which best fitted the points. With the possible exception of mass 111, the points fall on approximately symmetric curves having $Z_p \approx \bar{Z}$, indicating that our use of $\bar{Z}$ instead of Z_p in Fig. 7 is not in serious error.

The fcy points for $A = 131$ are typical of regions of small $\bar{\nu}_A$ (here about 0.6). The secondary fcy values fall on a nearly symmetric curve having $C = 1.07$, only slightly greater than that of the primary fragments, 0.94. The point for Te^{131} is in reasonable agreement with the experimental

value of 0.124 ± 0.014 [3]. The points for $A = 111$ are typical of regions of large $\bar{\nu}_A$, in this case about 2.3. The calculated C , 1.58, is much greater than that of the primary fragments and the points fall on a somewhat asymmetric curve which is broader on the low- Z side. This type of asymmetry would probably escape experimental detection as most independent-yield measurements are of species on the high- Z side.

The fcy values for $A = 154$ show the effect on an even- A chain of setting all δ values equal to zero. The points obtained using the pairing-energy corrections fall on a smooth curve that is unusually narrow for this region (see Fig. 9). Points obtained with all δ 's = 0 show strong odd-even effects: fcy 's of e-e nuclides are generally greater and those for o-o species lower than those obtained with pairing corrections. There is no experimental evidence for any such odd-even variation. In an extensive investigation of this point, Herrington was unable to remove odd-even fluctuations without the use of pairing corrections [22]. Therefore, if our assumptions of smooth primary yield-mass and charge-distribution curves are correct, it is apparently necessary to include pairing corrections in the level-density expression to avoid odd-even fluctuations in the secondary fcy curves.

Thomas and Vandenbosch suggested that, because of pairing effects on E_T , the primary yield of e-e species may be greater than for neighboring odd- A species, which may in turn be greater than for o-o species [23]. Possibly an adjustment of primary yields as suggested would yield smooth fcy curves for secondary fragments without the use of pairing corrections. In the present formulation, no apparent odd-even fluctuations are introduced into the secondary fcy distributions for odd- A , as each member of the chains has about the same value of δ .

Setting δ 's equal to zero also raises the $\bar{\nu}_A$ values to generally about 0.3 neutron above the experimental values and lowers the average γ -ray energy release per fission from 7.66 to 5.03 MeV.

In Fig. 9 are shown the calculated C values of the secondary fragments. The scatter of points results from the strong dependence of C on the secondary yields of species far from Z_p , whose yields are very sensitive to fluctuations of the Monte Carlo process. Clearly, however, there are observable trends of C with changing A.

The C values obtained in the calculation are probably generally too large. Independent-yield data analyzed by Wahl et al. [2] give $C = 0.94 \pm 0.15$ based mainly on mass chains 139 to 143, where we find $C \approx 1.15$. Part of the increase of our C values results from dispersion in $\bar{\nu}_A$ larger than consistent with experimental data as discussed below. Also, the input C of the primary fragments, 0.94, may be too large. An input C of 0.4 gave a secondary C dependence on A having approximately the same features as those of Fig. 9, but generally displaced ~ 0.5 charge unit lower.

There are only six mass chains for which independent yields are known for more than one isobar [2]. The five from A = 139 to 143 give $C = 0.94 \pm 0.15$; the sixth, at A = 91, gives $C = 1.48 \pm 0.87$. From our results in Fig. 9 it seems likely that there are large variations in C with A unless there are variations in C of the primary fragments in the opposite sense and of about the same magnitude as those shown for the secondary fragments in Fig. 9.

The value of C of the secondary fragments is a complicated function of C of the primary fragments, the $(\delta Z_p/\delta A)$ of the primary fragments, $\bar{\nu}_A$ and its derivatives with respect to A and with respect to Z_H/Z_L for constant A_H/A_L , the dispersion σ_{ν_A} , and, in regions of rapid change, the slope of the primary yield-mass curve. With C of the primary fragments kept constant with A , the next most important effects on secondary C values are probably σ_{ν_A} and $(\delta Z_p/\delta A)$. In Fig. 9 we see that the general trend of C parallels that of σ_{ν_A} , with perturbations caused by sudden large changes of $(\delta Z_p/\delta A)$. As noted below, the calculated σ_{ν_A} values are too large. If they can be reduced, the secondary C values will likewise be reduced.

The average number of prompt neutrons per U^{236*} fission (with 80-keV neutrons) is $\bar{\nu} = 2.47 \pm 0.03$, with $\sigma_{\nu} = 1.072 \pm 0.021$ [24]. In Fig. 9 we see that σ_{ν_A} for many mass chains is greater than the experimental value of σ_{ν} . We have calculated the neutron multiplicities of all pairs of complementary fragments assuming no correlation between ν_L and ν_H [as presumed by Eq. (5)], weighted each distribution by the primary yield and fcy for the division, and obtained the overall neutron number distribution. This distribution has $\bar{\nu} = 2.59$ and $\sigma_2 = 1.39$; thus, $\bar{\nu}$ is slightly too high, but σ_2 is more seriously in error. The latter problem cannot be avoided by assuming a negative correlation between ν_L and ν_H . This would imply a negative correlation between E_L and E_H , giving σ_{E_x} values too small for agreement with experimental σ_{E_K} values. A positive correlation between E_L and E_H would give $\sigma_{L,H}$ values smaller than those obtained from Eqs. (5) and (6), but when the ν_L and ν_H values were combined assuming positive correlations, σ_{ν} would again be too large.

The most obvious method of reducing σ_v is by relaxing the assumption implied by Eq. (2). If we were to allow $\bar{E}_K$ to vary with Z_H/Z_L for a given A_H/A_L , this could account for much of the observed dispersion in E_K rather than requiring a large σ_{E_x} as in our present treatment. In the limit, the change of E_T with Z_H/Z_L could be absorbed entirely by changing $\bar{E}_K$ and allowing $\bar{E}_x$ to remain constant and the distributions of E_x to be considerably narrower than in the present formulation. Thus, deviations from Eq. (2) cannot be directly determined from experiments, but seem inescapable in view of the inconsistency of our calculated σ_v with experimental data. In later works we hope to examine the results obtained by relaxing the assumption of Eq. (2).

In Fig. 10 we compare calculated and experimental [18] secondary mass yields. The general features are reproduced, but in low-yield regions, near symmetric fission and on the wings of the peaks, the calculated yields are too high. This is probably the result of errors in the primary yield-mass curve used. The calculated points do not reproduce some of the fine structure of the radiochemical curve, e.g. the "spike" at mass 134. This may be because of fine structure that was missed in the primary curve, but is more likely the result of improper treatment of neutron evaporation near shell edges.

4. Conclusions

The most important result is that the calculated σ_v is larger than the experimental value. This result suggests strongly that there is not a fixed

E_K distribution for all charge divisions for a given A_H/A_L , but that $\bar{E}_K$ varies with Z_H/Z_L , absorbing part or all of the variation of E_T .

The total energy release, E_T , obtained from Seeger's mass formula [6] is well accounted for by $\bar{E}_K$ and $\bar{E}_{\gamma\text{sum}}$ values and neutron kinetic energies in agreement with experimental data, and $\bar{\nu}$ higher than the experimental value by only 0.12 neutron, corresponding to an error in energy of ~ 0.8 MeV. The overall accounting for E_T is thus much better than one would expect from errors in the mass equation alone.

The charge-dispersion parameter, C , of the secondary fragments is generally greater than that of the primary fragments, but we do not know by what amount as our results are based on σ_{ν_A} values that are too large. The value of C of the secondary fragments probably varies appreciably with A . Any asymmetric distortions of the charge-distribution curve caused by neutron evaporation are not very serious. It is necessary to use pairing-energy corrections in the level-density expression to avoid odd-even fluctuations in the secondary charge-distribution curves for even- A fragments.

5. Acknowledgments

We wish to thank Miss Louise Wilson for her considerable help with the computer calculations. We acknowledge the assistance in the early phases of the work by Professor Demetrios G. Sarantites, Professor Richard D. Fink, and Dr. Arthur C. Herrington. We are grateful to Dr. Walter M. Gibson and Professor T. Darrah Thomas for the use of their data prior to publication. We thank Dr. James Terrell and Professor Charles D. Coryell for their helpful comments on this work. Calculations were performed on computers at the M.I.T. Laboratory for Nuclear Science, the M.I.T. Computation Center, and at the Oak Ridge National Laboratory.

References

- [1] CORYELL, C. D., KAPLAN, M., and FINK, R.D., Can. J. Chem. 39, (1961) 646/663.
- [2] WAHL, A.C., FERGUSON, R. L., NETHAWAY, D. R., TROUTNER, D. E., and WOLFSBERG, K., Phys. Rev. 126 (1962) 1112/1127.
- [3] SARANTITES, D. G., GORDON, G. E., and CORYELL, C. D., Phys. Rev. (to be published).
- [4] ARAS, N. K., MENON, M. P., and GORDON, G. E., Nucl. Phys. (to be published).
- [5] KAHN, H., AECU-3259, Apr. 1954 (unpublished).
- [6] SEEGER, P. A., Nucl. Phys. 25 (1961) 1/135.
- [7] GIBSON, W. M., THOMAS, T. D., and SAFFORD, G., THIS CONFERENCE, Paper SM 60/36.
- [8] MILTON, J.C.D., and FRASER, J. S., Can. J. Phys. 40 (1962) 1626/1663.
- [9] TERRELL, J., Phys. Rev. 127 (1962) 880/904.
- [10] DOSTROVSKY, I., FRAENKEL, Z., and FRIEDLANDER, G., Phys. Rev. 116 (1959) 683/702.
- [11] GIBSON, W. M., private communication, 1962.
- [12] MILTON, J. C. D., and FRASER, J. S., in Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958. United Nations, Geneva (1958) Paper P/199.
- [13] MAIENSCHIEIN, F. C., PEELE, R. W., ZOBEL, W., and LOVE, T.A., in Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958, United Nations, Geneva (1958) Paper P/670.
- [14] LEACHMAN, R. B., Phys. Rev. 101 (1956) 1005/1011.
- [15] TERRELL, J., Phys. Rev. 113 (1959) 527/541.
- [16] HOFFMAN, M. M., Phys. Rev. 133 (1964) B714/B724.
- [17] GROVER, J. R., Phys. Rev. 123 (1961) 267/275.
- [18] HYDE, E. K., The Nuclear Properties of the Heavy Elements. III Fission Phenomena, Prentice-Hall, Englewood Cliffs, N. J. (1964). Contains most experimental data on fission with references to the original papers.

- [19] KIMURA, K. and CORYELL, C. D., M.I.T. Laboratory for Nuclear Science Progress Report, M.I.T.-2098-No. 64, May 1964, p. 34 (unpublished).
- [20] TROUTNER, D. E., WAHL, A. C., and FERGUSON, R. L, Phys. Rev. 134 (1964) B1027/B1029.
- [21] WAHL, A. C. and NETHAWAY, D. R., Phys. Rev. 131 (1963) 830/831.
- [22] HERRINGTON, A. C., Thesis, Dept. of Chem. Eng., Massachusetts Institute of Technology, 1955 (unpublished).
- [23] THOMAS, T. D. and VANDENBOSCH, R., Phys. Rev. 133 (1964) B976/B982.
- [24] DIVEN, B. C., MARTIN, H. C., TASCHEK, R. F., and TERRELL, J., Phys. Rev. 101 (1956) 1012/1015.

Figure Captions

- Fig. 1. Primary yield and E_T as a function of Z_L for mass division 139/97.
 Upper: Primary fcy calculated according to Eq. (1) with $Z_{p97} = 38.63$ and $C = 0.94$.
 Lower: E_T obtained from Seeger's mass formula [6], $\bar{E}_K$ for 139/97 divisions is 171.5 MeV, the input value which gives an implied E_K approximately equal to the experimental values, 166.4 [7] or 167.8 [8].
- Fig. 2. E_{97} and implied E_K (139/97) distributions for various Z_{97} values. Calculated from the data shown in Fig. 1, with $\sigma_{E_{97}} = 9.2$ MeV (input value), and the assumption that the mass-139 fragment always has its average excitation energy, $\bar{E}_{139} = 8.3$ MeV.
- Fig. 3. Input and output values of $\bar{E}_K$ and σ_{E_K} from the (Prompt) calculation compared with the experimental values.
- Fig. 4. Histograms of implied E_K distributions for several mass divisions. Obtained using the input data shown in Fig. 3. For $A = 97$, the points are experimental results of Milton and Fraser [8], as are the experimental values of σ and $\bar{E}_K$ given.
- Fig. 5. Calculated average γ -ray energy release per fragment and sum of average γ -ray energy release for complementary fragments as a function of A .
- Fig. 6. Calculated (Prompt) and experimental laboratory-system neutron kinetic energy spectra.
- Fig. 7. Calculated (Prompt) and experimental Z_p secondary and $\bar{\nu}_A$ (Terrell [9]) as a function of A . Also shown are input Z_p primary values and those suggested by Wahl et al. [2].
- Fig. 8. Calculated secondary fcy values (shown as points) as a function of Z for three mass chains. The curves drawn in were calculated from Eq. (1) with $C = 0.94$ and Z_p which best fit the secondary fcy points.
- Calculated using pairing-energy corrections (δ 's)
 x Calculated with all δ 's = 0
- Fig. 9. Upper: Calculated C of the secondary fragments
 Middle: Standard deviation σ_{ν_A}
 Lower: Derivative ($\delta Z_p / \delta A$) of the primary fragments.
- Fig. 10. Calculated (Prompt) and experimental [18] secondary yield-mass points and curve of input primary yield-mass curve [8, 11].