PHYSICAL REVIEW C VOLUME 25, NUMBER 1 JANUARY 1982

Monte Carlo calculation of the deexcitation of fission fragments in the spontaneous fission of ^{252}Cf

M. Kildir and N. K. Aras
Department of Chemistry, Middle East Technical University, Ankara, Turkey
(Received 26 December 1979)

The consequences of the evaporation of prompt neutrons and γ rays from primary fission fragments in the spontaneous fission of ^{252}Cf have been examined by a Monte Carlo method. A semiempirical mass equation and experimental values of total kinetic energies are used to obtain the sum of excitation energies of complementary fragments. The division of this energy between complementary fragments is determined by the two-spheroid model for the scission configuration. The prompt neutron multiplicity distribution gives a value of 3.68 for the average number of prompt neutrons in ^{252}Cf(sf). Calculated values of the average number of prompt neutrons as a function of fragments mass are in good agreement with the experimental values. Calculated results for the charge dependence of total γ-ray energies show an odd-even effect, whereas no such effect is apparent in the charge dependence of the average number of neutrons. Mass and charge distributions of secondary fission fragments are also well reproduced in the calculations.

[NUCLEAR REACTIONS, FISSION ^{252}Cf(sf); calculated prompt neutrons and γ rays. Pairing effect on γ-ray energy.]

INTRODUCTION

The emission of prompt neutrons and gamma rays from individual fragments in ^{252}Cf(sf) has been the subject of detailed experimental investigations.[1–3] Measurements of the charge dependence of the total gamma ray energy of complementary fragments have shown the existence of an odd-even effect, whereas no fine structure was found in the measurements of the charge dependence of the total number of neutrons emitted from the complementary fragments in ^{252}Cf(sf).[1] However, the fine structure observed in the recent measurement on the mass dependence of the number of prompt neutrons has been taken as evidence of odd-even charge effects.[2,3]

Statistical theory has been used to calculate the characteristics of prompt neutrons and gamma rays emitted from primary fragments in ^{252}Cf(sf).[4–6] Owing to the assumptions used in the foregoing studies, it was not possible to determine the effect of charge on prompt neutron or gamma ray emission. In the present study, the neutron and gamma ray emission from all fission fragments with appreciable yields were carried out using a Monte Carlo method.[7] The average number of prompt neutrons and gamma ray energies were obtained as a function of charge and mass of primary fission fragments. The general trends of several experimental quantities in ^{252}Cf(sf) are well reproduced in this calculation.

MODEL ASSUMPTIONS AND CALCULATIONS

In order to calculate the neutron and gamma ray emission from fission fragments, the sum of the excitation energies of complementary fragments must be known. We assume that the average sum of the excitation energies E_x of complementary light (A_l,Z_l) and heavy (A_h,Z_h) fragments from fission of ^{252}Cf is given by

$$E_x = Q - \overline{\text{TKE}} , \quad (1)$$

where $\overline{\text{TKE}}$ and Q are the total kinetic energy and total energy release for a given fragment pair, respectively. The total energy release is given by $Q = \Delta M(^{252}\text{Cf}) - (\Delta M_l + \Delta M_h)$ and calculated with the values of mass excesses taken from Myers and Swiatecki.[8] The average total kinetic energy as a function of fragment mass is taken from time-of-

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flight measurements.[9]

The division of E_x between light and heavy fragments is determined with the two-spheroid model for the scission configuration. Here, the scission point configuration is approximated by two deformed fragments in contact. If the deformation energy of a fragment is taken as

$$E_{\rm def}=\alpha(D-R_0)^2\ , \tag{2}$$

the total potential energy of the system will be

$$V=\frac{Z_l Z_h}{D_l+D_h}+\alpha_l(D_l-R_{0l})^2+\alpha_h(D_h-R_{0h})^2\ , \tag{3}$$

where D is the distance measured along the major axis, R_0 is the radius, and Z is the charge of the light (l) and heavy (h) fragments. Here α is related to the stiffness parameter C_2 of the nucleus by

$$\alpha=\frac{2\pi}{5}\frac{C_2}{R_0{}^2}\ . \tag{4}$$

If we assume that there is a minimum in the potential energy at the scission point, then the deformation energy of fission fragments will be

$$E_{\rm def}=\frac{1}{\alpha}\frac{(\overline{TKE})^4}{4Z_l{}^2 Z_h{}^2}\ . \tag{5}$$

Here again, $\overline{\rm TKE}$ is the total kinetic energy and it is assumed to be independent of charge for a given fragment pair.[10] From Eqs. (4) and (5) and the relation $R_0=r_0A^{1/3}$, the ratio of deformation energies of complementary fragments is obtained as

$$\frac{E_{\rm def}(l)}{E_{\rm def}(h)}=\frac{C_{2h}A_l{}^{2/3}}{C_{2l}A_h{}^{2/3}}\ . \tag{6}$$

The stiffness parameters of some nuclei have been deduced and listed by Wong.[11] Neutron-rich nuclei with very short half-lives are formed in ^{252}Cf(sf). Hence, the stiffness parameters of these nuclei are not known. We have used the correlation between the experimental values of the stiffness parameters[11] and the Myers-Swiatecki shell energies[8] to estimate the stiffness parameters of fission fragments. The correlation was obtained using the functions

$$\frac{C_2}{C_2(\mathrm{LD})}=\frac{k-\delta W}{k+\delta W}\ , \tag{7}$$

where δW is the calculated value of shell correction energy and C_2(LD) is the stiffness parameter of a nucleus in the liquid drop model. The value of k was determined from the best fit to experimental data shown as a solid curve in Fig. 1 and found to be $k=8.0\pm0.2$ MeV.

According to Eq. (7), if the shell correction energy of a nucleus (δW) is zero, the stiffness parameter of that nucleus will be equal to its value in the liquid drop model. On the other hand, the nuclei near the closed shells have negative values for their shell correction energies and as a result their stiffness parameters calculated by Eq. (7) will be larger than that of the liquid drop model. This indicates their higher resistence against deformation. With the use of Eqs. (6) and (7) and the Myers-Swiatecki formulation of the stiffness parameters of nuclei, an equation for the ratio of deformation energies of complementary fragments is obtained as

$$\frac{E_{\rm def}(l)}{E_{\rm def}(h)}=\frac{(8.0-\delta W_h)(8.0+\delta W_l)\left[5.908-10.256\dfrac{(N_h-Z_h)^2}{A_h{}^2}-0.114\dfrac{Z_h{}^2}{A_h}\right]}{(8.0+\delta W_h)(8.0-\delta W_l)\left[5.908-10.256\dfrac{(N_l-Z_l)^2}{A_l{}^2}-0.114\dfrac{Z_l{}^2}{A_l}\right]}\ , \tag{8}$$

where N is the neutron number of a fragment. Equation (8) is used to determine the division of E_x between light and heavy fragments with the assumption that this division is the same as that of the deformation energies. The variances of the excitation energies of complementary fragments, $\sigma_{l,h}{}^2$, were obtained from the dispersion in the total kinetic energies[9] assuming no correlation between the excitation energies of fragments.

In the calculations, the independent yields of fission fragments are estimated using the experimental values of chain yields of primary fission fragments[9] and the primary fractional chain yields.[12] Mass chains with the mass number between 93 and 161 are considered. In each mass chain 10 isobars around the most probable charge are taken into account.

Charge distributions for all mass chains are assumed to be Gaussian with the charge-dispersion parameter of 0.84 about the most probable

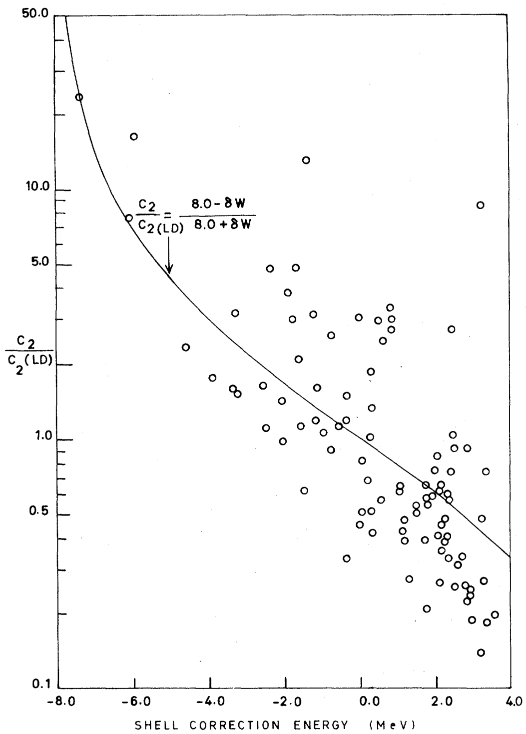

FIG. 1. The correlation between the stiffness parameters and shell correction energies for various nuclei.

charges.[12] The emission of prompt neutrons and gamma rays from a fragment with a given average energy is treated by a Monte Carlo method.[13] A given number of iterations is performed for all isobars having primary fractional chain yields larger than 10^{-6}. A Monte Carlo rejection technique is used to select the excitation energy of a given fragment and the kinetic energy of an emitted neutron. Here, the energy distribution for the primary fragment is assumed to be Gaussian in the range of $\bar{E}_{l,h} \pm 3\sigma_{l,h}$ and the emission probability of a neutron is taken to be as described by Dostroksky *et al.*[7] After the emission of the first neutron, the residual nucleus may have enough excitation energy to evaporate another neutron. This evaporation process continues until no more neutron emission is possible. Any excitation energy remaining after all possible neutrons have been emitted is assumed to be given off in the form of gamma radiation. In this treatment it is assumed that deexcitation of fission fragments by the emission of a neutron is always possible if its effective excitation energy is larger than the neutron separation energy. Note that except for the very first step of each iteration, only the kinetic energy of evaporated neutron is selected by a Monte Carlo rejection technique. The Fermi gas model of level densities is used in the calculations of emission probabilities of neutrons. Here, the level density as a function of excitation energy, $\rho(E)$, is given by

$$\rho(E) = C\exp\{\,2[a(E-\delta)]^{1/2}\,\}\ , \tag{9}$$

where C is a constant, δ is the pairing energy, and a is the level density parameter. The pairing energy is taken to be $22/A^{1/2}$ for even-even nuclei and $11/A^{1/2}$ for odd-A nuclei.[8] The level density parameter of the fragment with mass A is taken[12] as $A/13$.

After each neutron is emitted and at the end of each iteration, various quantities (neutron kinetic energies, gamma ray energy release, number of neutrons emitted, etc.) are weighted by the independent yield of the primary fragment and stored for later treatment and inclusion in the output data. After all mass chains have been treated, the various stored results are appropriately grouped and renormalized and average values and dispersion are computed.

RESULTS

The average sum of the excitation energies of complementary fragments calculated with Eq. (1) depends on the charge and mass of the fission fragments. As an example, the charge dependence of E_x for the mass division of $\frac{147}{105}$ is shown in Fig. 2(b). As is seen, the total energy release has a parabolic dependence on Z. Since this mass division produces two odd-A fragments the pairing energy does not show any fine structure in the total energy curve. In the case of the formation of two even-A fragments, there will be fine structure in the total energy curve as a function of Z due to the pairing effect. The average total kinetic energy of the mass ratio $\frac{147}{105}$ is equal to 184.4 MeV (Ref. 9) and is also indicated in Fig. 2(b). The difference between the two curves Q and $\overline{\mathrm{TKE}}$ gives the average sum of the excitation energies of complementary fragments. The primary fractional chain yields of isobars, $P_A(Z)$, of $A=147$ are shown in Fig. 2(a). They are calculated with $Zp=56.80$ and $C=0.84$. Using the values of $P_A(Z)$ and Q, the weighted average of the total energy release is calculated and shown as a function of A_h/A_l together with $\overline{\mathrm{TKE}}$ in Fig. 3. The energy difference between the two curves gives the weighted sum of the excitation energies of complementary fragments. As is seen in Fig. 3 the energy difference has a maximum around the symmetric division.

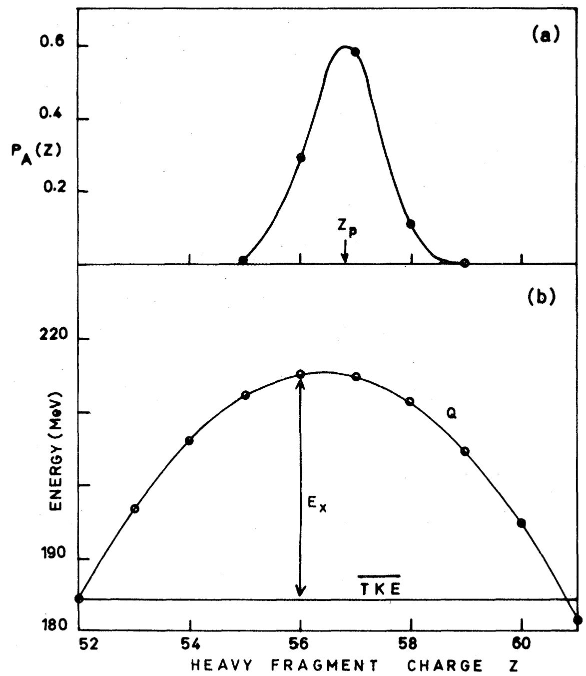

FIG. 2. (a) The primary fractional chain yields of isobars with mass 147. (b) Total energy release in ^{252}Cf(sf) as a function of heavy fragment charge for mass ratio of $\frac{147}{105}$.

If we assume the ratio of excitation energies of complementary fragments is the same as that of deformation energies, then Eqs. (1) and (8) can be used to determine the average excitation energies of fission fragments. These values of excitation energies of individual fragments are used in the calculation of characteristics of prompt neutrons and gamma rays by a Monte Carlo method. The ratio of the weighted average of the excitation energies of complementary mass chains are shown in Fig. 4. It has a maximum at the mass ratio of $\frac{134}{118}$.

The mass dependence of the average number of prompt neutrons, $\overline{\nu}(A)$, is shown in Fig. 5. The saw-tooth structure is well reproduced for the mass dependence of prompt neutrons from fission fragments. The sum of the average number of neutrons from complementary mass chains, $\overline{\nu}_T(A)$, has a maximum at the symmetric division which is consistent with the experimental result of Walsh *et al.*[2] The weighted average of $\overline{\nu}_T(A)$ over the chain yields of primary fragments gives a value of 3.68 for the average number of neutrons per fission of ^{252}Cf(sf), also in agreement with the experimental value of 3.73.[14]

The calculated prompt neutron multiplicity distribution is compared with the experimental results[14] in Fig. 6. The variance of the distribution is calculated to be 2.49. This is larger than the experimental value of 1.57 ± 0.02. As is seen in Fig. 6, the calculated prompt neutron multiplicity distribution in ^{252}Cf(sf) is represented quite well by a Gaussian function with $\overline{\nu}_T = 3.68$ and $\sigma_{\nu_T}{}^2 = 2.49$. The prompt neutron multiplicity distribution is also calculated for light and heavy fragments yielding the values of $\overline{\nu}_l = 2.23$, $\sigma^2 l = 1.42$ and $\overline{\nu}_h = 1.45$, $\sigma_h{}^2 = 1.07$, respectively.

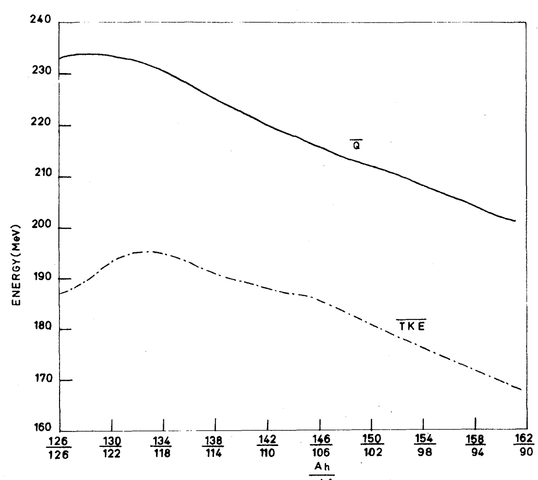

FIG. 3. Variation of the weighted average of the total energy Q, and total kinetic energy $\overline{\text{TKE}}$ with mass ratio in ^{252}Cf(sf).

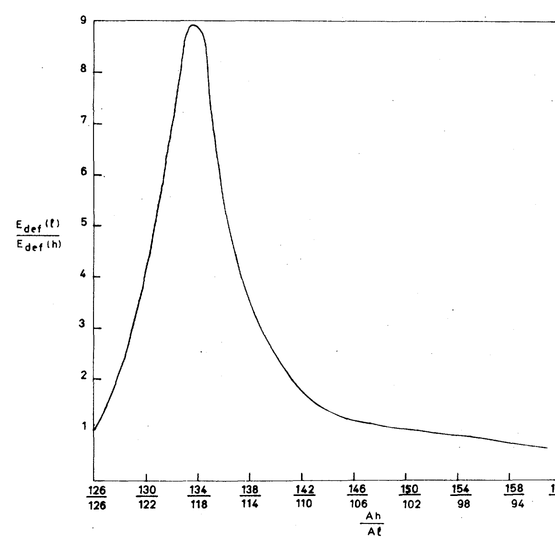

FIG. 4. Variation of the ratio of deformation energies of heavy and light fragments with mass ratio in ^{252}Cf(sf).

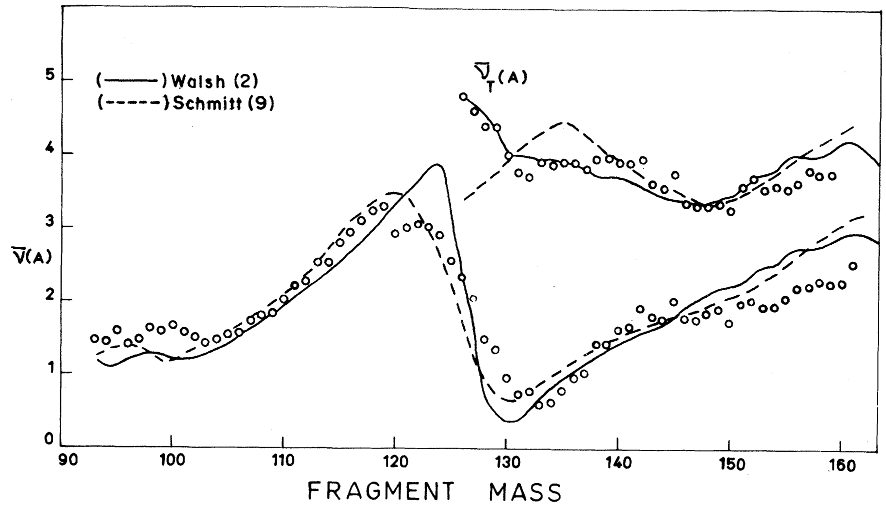

FIG. 5. Experimental and calculated prompt-neutron yields in ^{252}Cf(sf).

The charge dependence of the neutron and gamma ray emission has been calculated in order to examine the effect of pairing on these emissions. The isotopic fractional yields $P_Z(A)$ are determined with the use of independent chain yields and isobaric fractional chain yields. The weighted average of the numbers of prompt neutrons and the energies of gamma rays as a function of the charge of primary fragment is calculated by the use of $P_Z(A)$. The results are shown in Fig. 7 along with the experimental values of Nifenecker *et al.*[1] The general trends are well reproduced in each case. Results on the charge dependence of the total gamma ray energy as a function of the charge of the primary fragments show an odd-even effect. The weighted average of the total gamma ray energy for fragments of even Z is about 1.7 MeV larger than that of odd-Z fragments [Fig. 7(a)]. In the case of the average number of prompt neutrons as a function of fragment charge [Fig. 7(b)], no fine structure has been observed. This result is consistent with the experimental values of Nifenecker *et al.*[15]

Independent yields of secondary fission fragments are also calculated with the use of independent yields of primary fragments in the study. They are used to obtain mass and charge distributions in ^{252}Cf(sf). The calculated and experimental mass yields of secondary fission fragments[16] are compared in Fig. 8. The general features are well reproduced but in the near symmetric fission the

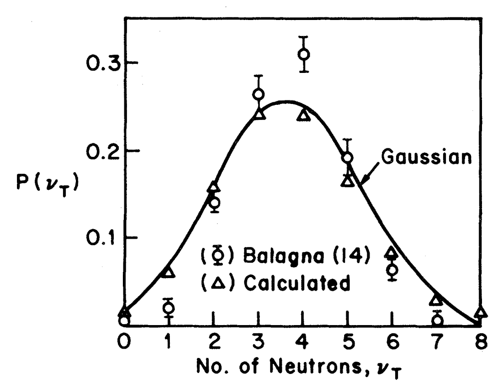

FIG. 6. The prompt-neutron multiplicity distribution in ^{252}Cf(sf).

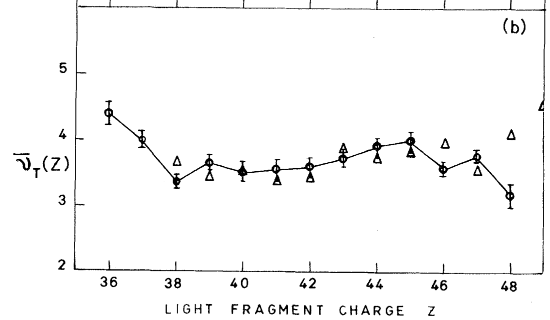

FIG. 7. (a) Variation of total energy released by γ rays with fragment charge. (b) Variation of total prompt-neutron yields with fragment charge in ^{252}Cf(sf).

calculated yields are too high. This may be a result of errors in the chain yields of the primary fragments. The chain yield of the primary fragment with $A = 126$ is 0.12%. The yields of mass chains of $A > 126$ are larger than this value. Hence, we do not expect to obtain a value much smaller than 0.12% for the yield of secondary fission fragment with $A = 126$. This is larger than the experimental result for this mass chain; however, one would expect a large error due to the small yield and resolution problems in instrumental measurements.

The effect of prompt-neutron emission on the shape of the charge distribution curve has been studied. Charge distribution curves of secondary fragments for three different mass chains are shown in Fig. 9 along with the experimental data.[17–19] Their shapes are found to be Gaussian with the dispersion parameters almost always greater than that of the primary fragments.

DISCUSSION

The Myers-Swiatecki mass formula[8] combined with the kinetic energy values of Schmitt[9] gives a value of 3.68 for the average total number of neutrons in ^{252}Cf(sf). It is smaller by only 0.05 neutron (or about 0.4 MeV in energy) than experimental value. As is seen in Fig. 3, the weighted sum of the excitation energies of complementary fragments has

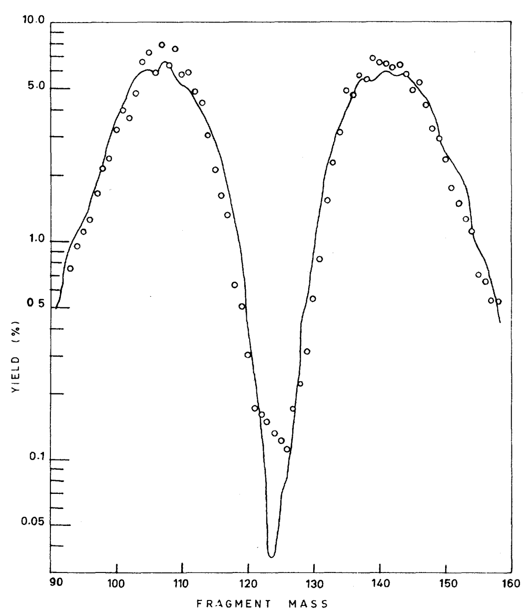

FIG. 8. Calculated (O) and experimental yield-mass curve for secondary fragments in ^{252}Cf(sf).

a maximum around the symmetric division. As a result the calculated values of the average number of neutrons from complementary mass chains gives a maximum at the symmetric division which is consistent with the experimental results of Walsh *et al.*[2] (Fig. 5). Also, the calculated values of $\bar{\nu}_T(A)$ as a function of heavy fragment mass are in good agreement with the same experimental results. Hence, it may be concluded that the total energy release Q in fission can be well accounted for by the liquid drop model of nuclei.

The division of the total excitation energy between the complementary fragments is obtained by the use of Eq. (8) and it is shown as a function of mass ratio in Fig. 4. It has a maximum at the mass ratio of $\frac{134}{118}$. This behavior can easily be understood when we realize that nuclei around the mass number of 132 contain numbers of protons and neutrons either equal or very close to the shell numbers of $Z = 50$ and $N = 82$. Hence, they are quite resistant to any type of deformations. As a result, they will take only a small part of the energy which is expended to produce large deformations in the fissioning nucleus. On the other hand, complementary fragments of mass 132 will contain numbers of protons and neutrons much different from magic numbers and their share of the deformation energy will be large at the scission point. After scission and separation of fragments, these deformation energies will be converted to the excitation energies of primary fragments. As a result the mass dependence of the average number of prompt neutrons will be dependent on the amount of energy stored in deformation at scission. As is seen in Fig. 5, the saw-tooth structure is well reproduced, which indicates that a reasonable model for the division of the total excitation energy between the complementary fragments is used in the calculations.

Calculated results for the charge dependence of total gamma ray energies show an odd-even effect, whereas no such effect is apparent in the charge dependence of the average number of neutrons (Fig. 7). Even though this result is consistent with the experimental values of Nifenecker *et al.*,[15] it is surprising that none of the excess energy in total energy release due to pairing passes into neutron emission. This may be understood if the effect of angular momentum is taken into account. The average spins of primary fission fragments are approximately $6-8\hbar$ higher than ground state spins. Neutron emission is expected to decrease the spin of fragments by about one unit of angular momen-

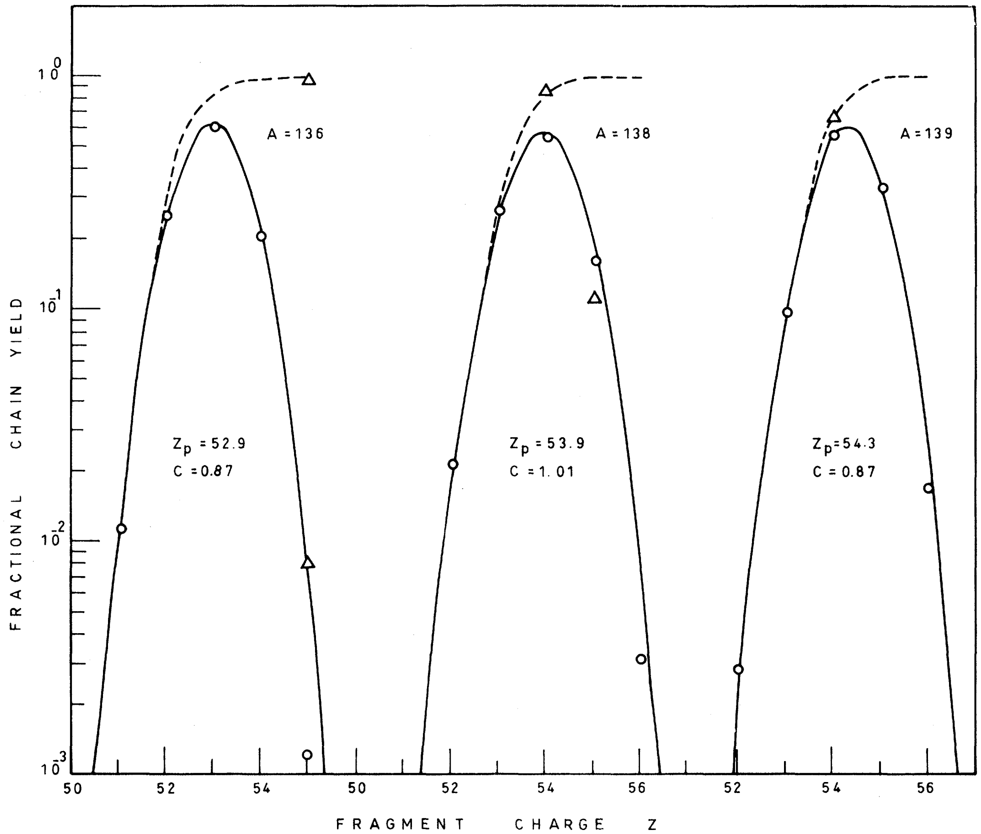

FIG. 9. Calculated (○) and experimental (Δ) charge distribution in ^{252}Cf(sf).

tum. After the emission of a few neutrons, the fragments are left with an excitation energy only slightly higher than the neutron binding energy but will still have from 5 to 7$\hbar$ units of angular momentum to dissipate. Further neutron emission, which would leave the residual nucleus in the vicinity of its ground state, is thus expected to be inhibited except for odd-odd nuclei. Hence, the relatively small amounts of energy will be dissipated by gamma ray emission in the case of odd-Z nuclei. Even though the effect of angular momentum is not taken into account in the present study, the pairing energy term in the level density expression produces the same effect as a correction for the angular momentum of the fragments. The difference between the calculated ($\Delta\bar{E}_\gamma = 1.7$ MeV) and the experimental values[20] ($\Delta\bar{E}_\gamma = 0.66 \pm 0.05$ MeV) of the odd-even effect on the total gamma ray energy will be reduced if smaller pairing energies are used in the calculations. In this case more of the excitation energy will be available for the neutron evaporation. Since the estimated energy cost per neutron evaporation is about 7 MeV, a 50% decrease in the pairing energy will increase the average number of prompt neutrons by only about 0.1. Even smaller effects on the other calculated quantities may be expected. Since our principal aim in these calculations was to reproduce the general trends of many important experimental results with a reasonable set of initial parameters and assumptions, we kept the value of the pairing energy as given in the Myers-Swiatecki mass formula.

As a result of the good agreement obtained between the calculated and experimental values of several important quantities in the spontaneous fission of ^{252}Cf, it is concluded that the emission of prompt neutrons and γ rays from primary fission fragments may be taken as a statistical process and treated by a Monte Carlo method. This study will be extended to other fissioning nuclei to investigate the validity of the model.

ACKNOWLEDGMENTS

We thank Prof. G. E. Gordon for the critical reading of this manuscript. This work was supported in part by Scientific and Technical Research Council (TUBITAK) of Turkey.

*Now on sabattical leave at Department of Chemistry, University of Maryland, College Park, Maryland.

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