Peculiarities of cluster formation in true ternary fission of $^{252}$Cf and $^{236}$U*

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Abstract
The existence of a new type of cluster decay called 'collinear cluster tri-partition' (CCT) is discussed by an analysis of the landscape of the potential energy surface (PES). The total energy of the ternary system is found as a sum of the binding energies of fragments and nucleus–nucleus interaction between them. The pre-scission state of the ternary system is assumed to be arranged as a chain of the three fragments along a straight line. Minima and valleys of the PES are determined by variation of the proton and neutron distributions between them. Pre-scission prompt emission of neutrons is assumed and PES is calculated for the cases of emission of two neutrons. The presence of the valley corresponding to the formation of the isotopes of Sn with masses $A = 130$–$136$ is inherent for all PES calculated for CCT for spontaneous fission of $^{252}$Cf and fission induced by neutrons of $^{235}$U. There are local minima indicating the formation of Ca, Fe, Ni, Ge and Se isotopes having magic proton or/and neutron numbers, such as 20, 28 and 50. The analysis shows that the experimentally observed $^{68}$Ni is formed as the edge fragment of the ternary system connecting to Sn by Si and Ca isotopes at fission of $^{236}$U and $^{252}$Cf, respectively.

Keywords: collinear ternary fission, potential energy surface, clusters, fission barrier

(Some figures may appear in colour only in the online journal)

1. Introduction

The role of the nuclear shell structure in the formation of fission products appears in the observed asymmetric mass distributions depending on the excitation energy of the system undergoing fission. References [1, 2] discuss the yields of very asymmetric Ni and even Fe isotopes in the cold binary fission of $^{236}$U analyzed as a fission channel around magic proton number $Z = 28$ with the mass number 70. The yield of the very asymmetric products was not observed in coincidence with conjugate heavy product and, therefore, the authors of the above-mentioned paper did not consider the possibility of those fission fragments as CCT products. This scenario of the yield of the very asymmetric products of the Ni isotopes with $A = 68$ and 70 was studied in the $^{235}$U($n_{th}$, $f$) reaction and at spontaneous fission of $^{252}$Cf by the FOBOS collaboration [3–7].

The observation of two and more nuclear fission products in the fission of $^{235}$U with thermal neutrons and in the spontaneous fission of $^{252}$Cf has opened a new area of study in nuclear reactions. This phenomenon is connected with the appearance of cluster states in nuclear reactions and it is the manifestation of the shell structure which is responsible for the production of isotopes with magic numbers of neutrons and protons. When a massive nucleus loses its stability and goes to fission, first of all clusters are formed as future fragments having the neutron or/and proton number close to the
magic numbers 28, 50, 82 and 126. In the case of ternary fission one observes fragments with the charge numbers 28, 50, 82 and 126. This kind of ternary fission is known as true ternary fission, and it produces fragments with comparable masses flying in the same line but opposite directions (see figure 1), which is different from the emission of alpha-particles or light-charged nuclei with mass numbers $A < 16$ in the plane perpendicular to the fission axis binary fission [8].

The probability of multicluster fission of the U, Pu and Cf isotopes is less than one per cent of the corresponding cross sections of binary fission. The cross section of CCT is comparable with that of the well-known ternary fissions with the emission of an alpha-particle [9, 10]. The emission of the light-charged particles from the neck region on the plane perpendicular to the fission axis is the main characteristic of ternary fission, although the light charged particles can be emitted in direction close to the momentum of the fission fragments [9]. The probability of the yield of light-charged particles heavier than alpha-particles decreases with increasing charge and mass number. The observed yield of the group of neutron-rich isotopes of Ni and Ge in coincidence with the heavy fragment with mass number $A = 136–140$ is an unexpected phenomenon. Therefore, theoretical interpretation of these processes is required for a full understanding of the mechanism. The ternary fission fragmentation of $^{255}$Cf for all possible third fragments using the recently proposed three-cluster model [11] was studied in [12]. The authors concluded that the theoretical relative yields imply the emission of $^{14}$C, $^{34,36,38}$Si, $^{46,48}$Ar and $^{48,50}$Ca as the most probably third particle in the spontaneous ternary fission of $^{255}$Cf.

The results discussed in this paper are based on two different experiments with binary coincidences of fission fragments and measurements of the masses and energies of the two fragments [4]. In two other experiments [7] for the study of spontaneous ternary fission of $^{255}$Cf, events in coincidence with neutrons are reported. The prompt emission of neutrons from the neck region (scission neutron source) is inherent to the spontaneous fission of the actinides [13]. The authors of [7] reported on the two registered products of CCT in coincidence with the neutron multiplicity emitted from the neck region of the fissioning nucleus. The ‘neutron belt’ was assembled in a plane perpendicular to the symmetry axis of the spectrometer, which serves as the mean fission axis at the same time. The third fragment of CCT was blocked by the entrance structure of the detectors.

The relatively high yield of the CCT-effect (more than $10^{-7}$/binary fission) is likely due to the collective motion through very elongated (hyper-deformed) pre-scission shapes and a large phase space covering a larger number of mass partitions with high $Q$-values [14]. The formation of the third cluster occurs in the neck region between the main binary fragments during the pre-scission stage of the splitting. The fact that the formation of binary fragments is the main channel is seen from all PES figures as a wide and deep valley at $Z_3 < 2$, where $Z_3$ is the charge number of the middle cluster.

The case of alpha-cluster formation has been well studied both experimentally and theoretically. But the CCT process needs to be studied in detail taking into account conditions leading to the formation of a ternary system with comparable masses and dynamics of the rupture of two necks connecting border fragments to the middle nucleus.

The aim of the calculation is to explain the possibility of the population of the Ni isotopes as a fragment in the ternary system. The relatively large cross section of the yield of fission products with the given charge and mass numbers is the consequence of the population of the states corresponding to the minima of PES. This is a necessary condition and the cluster can be emitted from the system if it is able to overcome the pre-scission barrier of the nucleus–nucleus interactions connecting the ternary system. Therefore, at first, it is important to analyze the PES landscape calculated for the considered fissioning system under consideration (figure 1).

2. Outline of theoretical approach

The calculations are performed based on an assumption that the third cluster has appeared in the neck region of the binary fragments due to fluctuation of the proton and neutron transfer between them during descent from the saddle point before scission point. The difference between the total energy of the ternary system and fissioning nucleus is used as the PES, which shows an effect of the nuclear shell structure on the nascent fission products. The PES is found as a sum of the energy balance of the interacting fragments and
nucleus–nucleus interaction between them

\[
U(R_i, R_j, Z_i, Z_j, A_i, A_j) = V_i(R_i, Z_i, Z_j, A_i, A_j) + V_j(R_j, Z_j, Z_i, A_j, A_i) + V_{ij}^{(Coul)}(Z_i, Z_j, R_i + R_j) + Q_{ij}
\]

(1)

Here \( Q_{ij} \) is the balance of the fragments binding energy at the ternary fission; \( V_i \) and \( V_j \) are the nucleus–nucleus interaction of the middle cluster \('3'\) (\(A\) and \(Z\) are its mass and charge numbers, respectively) with the left \('1'\) (\(A_1\) and \(Z_1\)) and right \('2'\) (\(A_2\) and \(Z_2\)) fragments of the ternary system; \( V_{ij}^{(Coul)} \) is the Coulomb interaction between two border fragments \('1'\) and \('2'\) nuclei and vice-versa; the last interaction affects \( V_{jk} \). The effect of the third fragment is important in the case of short time between ruptures of two necks because the depth of the potential well will be smaller, i.e. the barrier against fission will be smaller. Consequently the probability of the ternary fission increases. The values of the binding energies of all considered possible fragments, as constituents of the ternary system, were obtained from the table of masses by Audi et al [18].

The procedure of calculations of PES by using equation (1) has been made by following steps: i) We find positions \( R_{m1} \) and \( R_{m2} \) of the edge fragments \( Z_{A1} \) and \( Z_{A2} \) relative to the middle cluster \( Z_{A3} \), respectively, providing the minimum value of \( U \) by variations of values of \( R_1 \) and \( R_2 \). ii) The cluster mass number \( A_3 \) for each charge number \( Z_3 \) is changed from the minimum value \( A_3^{\text{min}} \) to the maximum value \( A_3^{\text{max}} = 2.6Z_3 \), leading to a strong increase of \( U \). iii) The charge number of the right edge fragment \( Z_2 \) is found from the conservation law for the proton numbers \( Z_2 = Z_{A2} - Z_3 - Z_1 \). iv) The neutron distribution between constituents of the ternary system at the given charge distribution is varied in order to find the minimum of \( U \) as a function of \( A_1 \) for the given mass and charge numbers \( A_3 \) and \( Z_3 \) of the middle cluster and charge numbers \( Z_1 \) and \( Z_2 \). We vary \( A_1 \),...

**Figure 2.** Potential energy surface (\( U \)) calculated for the formation of fragments of the ternary system formed after emission of two neutrons from \(^{252}\text{Cf}\) as a function of the charge numbers of middle cluster and left edge fragment.


from $A_1 = 2Z_1$ up to $A_1^{\text{max}}$ corresponding to the strong increase of PES. The mass number of the right edge fragment $A_2$ is found from $A_2 = A_\text{tot} - A_3^{\min} - A_1$. The above-mentioned $Z_\text{tot}$ and $A_\text{tot}$ are the total charge and mass numbers of the fissioning system.

The results of calculations $U(R_{m1}, R_{m2}, Z_1, Z_2, A_1, A_2)$ can be presented as a matrix of size $(\Delta Z_3 \times \Delta Z_1)$, where $\Delta Z_3$ and $\Delta Z_1$ are the intervals of variation of $Z_3$ and $Z_1$, respectively.

Probably the constituents are not in their ground state after formation and before their escape from the ternary system, but a procedure of calculation by considering mass and charge numbers as variables, changing in the wide range of values, will be cause very time-consuming calculations.

Therefore, shell effects in the binding energies do not depend on their deformation.

### 3. Results of calculation

Results of the PES for the ternary fission of $^{252}$Cf are presented by a contour map in figure 2 as a function of the charge numbers of the middle cluster $Z_1$ and one, $Z_2$, the right edge fragment. It can be seen as a valley corresponding to the formation of the cluster $^{132}$Sn for different values of $Z_1$ and $Z_2$. This fact reflects the long tail in the mass–mass distribution of the experimentally registered products, which is parallel to the $M_1$ and $M_2$ axes (see figure 4 in [4] and figure 5(a) in [7]). Those tails demonstrate the persistence of the shell structure in the double magic nucleus $^{132}$Sn in the formation of the fission fragments. The vertical line at $Z_1 = 28$ shows local minima corresponding to the formation of Ni isotopes as fragments of the ternary system. This line crosses the line on the valley of formation $^{132}$Sn isotope at $Z_1 = 20$ where there is a local minimum. The probability of formation of the cluster configurations $^{132}$Sn + $^{48}$Ca + $^{68}$Ni after the emission of two neutrons is large because the proton or neutron numbers of the three fragments are equal to the magic numbers whether 28, 50 and 82. The dependence of the driving potential $U_d$ extracted from PES $U(R_{m1}, R_{m2}, Z_1, Z_2, A_1, A_2)$ at $R_1 = R_{m1}$ and $R_2 = R_{m2}$ on the charge and mass numbers of the right edge fragment is shown in the upper and lower figures of figure 3, respectively. The comparison of results obtained for the mass numbers 48 (dotted curve), 50 (solid curve) and 52 (dashed curve) of Ca being the middle cluster in figure 3, demonstrates that minimal values of driving potential corresponds to the formation of the $^{68}$Ni isotope, which was observed with sufficiently large probability (see [4, 7]), when $^{48}$Ca is formed as a middle cluster and $^{132}$Sn is the right edge fragment. On the contour map of the PES there are local minima showing the favored population of $^{132}$Sn + $^{38}$S + $^{92}$Ge, $^{132}$Sn + $^{36}$Si + $^{84}$Se, $^{138}$Ba + $^{72}$O + $^{86}$Ge, and others. We found that the middle cluster is more neutron rich than edge fragments. A much smaller energy minimum in the PES (by 10 MeV) for the alternative configuration, the $^{132}$Sn + $^{72}$Ni + $^{48}$Ca channel, gives for this reaction a much smaller probability; the difference is due to the changed Coulomb repulsion forces. This effect is observed in the yields observed in the experiment [4].

![Figure 3](image1.png)

**Figure 3.** Comparison of the driving potentials calculated for the pre-scission state of the collinear ternary system $Z_1 + ^{48}$Ca + $Z_2$ formed in the spontaneous fission of $^{252}$Cf as a function of $Z_1$ (upper figure) and as a function of $A_2$ (lower figure).

![Figure 4](image2.png)

**Figure 4.** The mass–mass distribution of the fission-fragments of the spontaneous fission of $^{252}$Cf gated by 2n emission. Arrows with numbers 1-6 mark the positions of masses of magic nuclei; a line numbered 7 points to events with the loss of a $^{14}$C nucleus. The main intensity is with masses for the third fragments from 36-20. Reproduced with permission from [7] Copyright 2012 Springer Science+Business media.
In figure 4 obtained from [7] the events of yields of two products in the spontaneous fission of $^{255}$Cf registered in coincidence are presented. Intense yield of the fission products with $A_1 = 68–94$, $A_2 = 50–60$ and $A_2 = 128–146$ registered in coincidence is observed. Obviously we see the sufficient influence of the shell effects in nuclear matter in the formation of the ternary fission products.

The results calculated for $^{255}$Cf and presented in figure 5 show the valley of minimum values of the PES in the mass number regions $A_1 = 70–100$ and $A_2 = 124–144$. The population of the mass distribution of the edge fragments in the corresponding ranges of $A_1$ and $A_2$ should dominate in the ternary fission of $^{255}$Cf. The fission probability from these pre-scission states depends on the value of the pre-scission barriers $B_{s1}$ and $B_{s2}$, which is determined by the depth of the potential well. The splitting probability of $^{132}$Sn from the other part of the system is determined by the depth of the potential well, which can be considered as pre-scission barrier $B_s$. The value of $B_s$ depends on the charge distribution between fragments of the ternary system. For the collinear configuration of the ternary system, we have two necks in the connected fragments and, consequently, we have two barriers $B_{s1}$ and $B_{s2}$ for separation of the left and right edge fragments. The answer to the question, which of these necks breaks earlier, depends on the relation between $B_{s1}$ and $B_{s2}$. The dependence of the values of $B_{s1}$ and $B_{s2}$ on the charge number $Z_1$, for the case that the middle cluster is $^{34}$Si at CCT of $^{238}$U, is shown in the upper part of figure 6. The lower part of figure 6 presents the driving potential of the ternary system $Z_1 + ^{34}$Si + $Z_2$, being formed at CCT of $^{238}$U. The presented results for the driving potential allow us to conclude, that 1) formation of the $^{60}$Ni isotope in coincidence with $^{132}$Sn occurs with large probability, because this configuration of the collinear ternary system has lower potential energy; the rupture of the neck connecting $^{132}$Sn to $^{34}$Si + $^{60}$Ni system occurs more easily than the rupture of the neck connecting $^{60}$Ni to the $^{34}$Si + $^{132}$Sn system. Then the second neck connecting the $^{34}$Si + $^{60}$Ni system in the field of $^{132}$Sn breaks down. A similar situation takes place in the case of CCT in the spontaneous fission of $^{255}$Cf, where $^{60}$Ni is formed together with $^{132}$Sn and $^{50}$Ca.

4. Summary

A possibility of the formation of $^{60}$Ni isotope in coincidence with $^{132}$Sn occurs with large probability, because this configuration of the collinear ternary system has lower potential energy; the rupture of the neck connecting $^{132}$Sn to $^{34}$Si + $^{60}$Ni system occurs more easily than the rupture of the neck connecting $^{60}$Ni to the $^{34}$Si + $^{132}$Sn system. Then the second neck connecting the $^{34}$Si + $^{60}$Ni system in the field of $^{132}$Sn breaks down. A similar situation takes place in the case of CCT in the spontaneous fission of $^{255}$Cf, where $^{60}$Ni is formed together with $^{132}$Sn and $^{50}$Ca.
landscape. There are local minima indicating the formation of Ca, Fe, Ni, Ge and Se isotopes having magic proton or/and neutron numbers, such as 20, 28 and 50. The analysis shows that the experimentally observed $^{60}\text{Ni}$ is formed as the edge fragment of the ternary system connected to the formation of Sn by Si and Ca isotopes at fission of $^{236}\text{U}$ and $^{252}\text{Cf}$, respectively.

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