Canadian Journal of Physics

Transformations of actinides fission product yields due to post scission emission of nuclear particles:²³²Th

Journal:	<i>Canadian Journal of Physics</i>
Manuscript ID	cjp-2020-0356.R4
Manuscript Type:	Article
Date Submitted by the Author:	06-Mar-2021
Complete List of Authors:	Maslyuk, V.T.; Institute of Electron Physics National Academy of Sciences of Ukraine, Photonuclear processes Parlag, O.O.; Institute of Electron Physics National Academy of Sciences of Ukraine Romanyuk, M.I.; Institute of Electron Physics National Academy of Sciences of Ukraine, Photonuclear processes Lendyel, O.I.; Institute of Electron Physics National Academy of Sciences of Ukraine, Photonuclear processes Pop, O.M.; Institute of Electron Physics National Academy of Sciences of Ukraine, Photonuclear processes
Keyword:	post-scission approach, fission-products yield, beta particles, neutrons, emission
Is the invited manuscript for consideration in a Special Issue? :	Not applicable (regular submission)

SCHOLARONE™
Manuscripts

Transformations of actinides fission product yields due to post scission emission of nuclear particles:²³²Th

V. T. Maslyuk, O. O. Parlag, M. I. Romanyuk, O. I. Lendyel and O. M. Pop
*Institute of Electron Physics - Universitetska str., 21, 88017, Uzhhorod, Ukraine**
 (Dated: March 5, 2021)

The “many ensembles” method was proposed to investigate the influence of nuclear particles’ post-scission emission on mass and charge distributions of fission products. The post scission approximation had been used; each of these ensembles consists of the fission fragments after emission of chains of different lengths, both the beta ($\beta^\pm$) particles and neutrons. The theory allows one to find the most probable two fragment clusters of fission products and study their evolution after the post-scission emission of nuclear particles. The isotope ²³²Th was chosen as an example, the fission fragments of which are intensively studied in the experiment. It is shown that the post-scission emission of nuclear particles eventually leads to the convergence of the asymmetric peaks, which looks like enhanced symmetric fission mode over asymmetric one for fission product yields. A comparison of the theoretical results and experimental data for the ²³²Th fission fragments indicates their satisfactory matching.

PACS numbers: 21.10.Gv; 24.10.Pa; 64.60.Ej

Keywords: post-scission approach, fission-products yield, beta particles, neutrons, emission, time evolution, ²³²Th

I. INTRODUCTION

The mass/charge spectra of fission-fragment (MCSFF) yields are important observable parameters that are dependents on the nature of the inter-nuclear interaction and the influences of filled nuclear (proton, neutron) shells [1]. The study of MCSFF provides valuable information about the nature of nuclear matter [2, 3], and is essential for the physics of nucleosynthesis [4], studying the heavy nuclei stability [5, 6]. The applied aspect of the MCSFF study is vital for terrestrial radioecology, nuclear power tasks, and radioactive waste utilization [7]. The heavy nuclei’s fission fragments are the basis for forming the chemical and isotopic composition of the Earth’s crust and space objects. However, in practice, it is difficult to establish the MCSFF because a post-scission ensemble is a dynamic system that is influenced by the nuclear particle emission from the fission fragments. The number of either neutron (prompt, delayed) or beta ($\beta^\pm$) depends on the time after nuclear fission: more nuclear particles will be emitted for a longer time interval. The time scale of such processes includes ultra-short-lived and almost stable nuclei-fragments. Therefore, for the experimental study of MCSFF, the abilities of the widely used experimental methods from radiochemical and gamma-spectrometric [8] to the capabilities of the large-acceptance magnetic spectrometer [9] should be carefully analyzed. Investigating the time evolution of the MCSFF ensemble when considering the post-scission emission of elementary particles is also a difficult task for theoretical studies [10, 11]. Now, there are different theoretical approaches to describe the for-

mation of fission fragments ensembles, the topology of their mass-charge spectra based as a rule on various modifications of the liquid drop model. Macroscopic approaches widely exploit the mechanical ideas as size, shape, and viscosity and the kinetics characteristics of the nuclei-scission processes [12, 13]. Recently, significant progress has been made in combining macro- and microscopic approaches to describe a fissile system’s potential energy surface [14, 15]. A number of microscopic dynamic self-consistent quantum transport theories have also been proposed, which provide ample opportunities to study the nucleus’s stability using from ab initio to self-consistent mean-field approaches and allow one to take into account the dynamics of the fission process [16, 17]. The obtained surfaces of the fissile nuclei’s potential energy, depending on their deformation, allow us to study the nucleus’ decay trajectories within multidimensional Langevin models and establish as the mass distribution of fission fragments and also their total kinetic energy characteristics [18, 19]. Within these approaches, the nucleus’s post-scission state is considered the binuclear fission fragments system only; of their subsequent nuclear transformations are neglected. However, the importance of post-fission neutron and beta particle emission for the nuclear fission process is widely discussed in the literature [20, 21]. There are current tendencies to go beyond the simple two-fragments scission scheme and consider the effects caused by the emission of nuclear particles from the fission fragments [22]. Such theories would allow one to investigate the time evolution of the MCSFF and explain the experimental data when the fission fragments yields are carried out after some time after the nucleus fission (the cooling time). In the present work, we propose the many ensembles method for studying the post-scission transformation of MCSFF with an account of nuclear particles’ emission. It is done within the framework of a statistical approach that allows one to study

* <http://www.iep.org.ua>;
 vmasliuk@nas.gov.ua; volodymyr.maslyuk@gmail.com

the ordering of the post-scission ensemble of fission fragment clusters [23]. The prospects of thermodynamics in the nucleus fission were reported in [24, 25].

II. THEORY

The basics of the post-scission approach and the proposed statistical method for the systematization of fission fragments are described earlier [22, 26, 27]. A similar ideology is also implemented in the SPY (Scission Point Yields) model, which focuses on [28] studying the yields and energy characteristics of fission fragments, based on all statistical nucleon analysis configurations that can be realized at the nuclear scission point. The study's subject is the post scission configurations or the ensemble of fission fragments of the fissioning nucleus with atomic mass A_0 and the charge Z_0 obtained due to thermodynamic ordering. This ensemble's single nuclear cluster, denoted by i , comprises two fission fragments $j=1, 2$ with protons and neutrons when nuclear particles are emitted. Each cluster satisfies the following conservation conditions:

$$\begin{aligned} \sum_{j=1,2} (N_{p,j}^i + N_{n,j}^i + n_j^i) &= A_0, \\ \sum_{j=1,2} (N_{p,j}^i + m_j^i) &= Z_0. \end{aligned} \quad (1)$$

The positive sign m_j^i has the meaning of the number of emitted β^+ and a negative one, respectively, the number of β^- particles. There are several options for constructing ensembles of fission fragments, taking into account post scission emission of nuclear particles. In the first one, such an ensemble can be built for fixed values of n, m in (1), i.e., $n_1^i = n_2^i = n$, $m_1^i = m_2^i = m$. In this paper, considering the statistical nature of the emission of nuclear particles, we used an extended version, when n, m only sets the limits within which the emission of nuclear particles can occur. In other words, the ensemble will contain all the fission fragments obtained, taking into account both the neutron emission in the range (0:n) and for beta particles, respectively, in the range (0:m). Only the conservation conditions (1) must be hold. By changing the sets n, m , it is possible to realize a different number of ensembles, each of which contains all the fission fragments obtained after emission of the nuclear particles within the range of values (0, 1, ..., n) and (0, 1, ..., m). Further calculation in the case of many ensembles based on the following assumptions [29]:

- nucleons with different binding energy for different nuclei fragments must be considered as statistically non-equivalents;
- the thermodynamic ordering of the nuclei fragment ensembles determines the probability of realization and the topology of the fission fragments yields;

- the thermodynamic parameters of all fission fragments obtained without or taking account post scission nuclear particles emission are determined by the initial state of the fissile nucleus. The last one may be considered as the thermostat.

The proposed thermodynamic method contains no time variables and cannot consider the kinetics of the fission process. It is focused on investigating only the final state of the fission fragments ensemble realized after the complex radioactive-decay processes of neutron/beta particle emission both as prompt and delayed. However, the time evolution of MCSFF can be considered indirectly by constructing many nuclei ensembles resulting after the emission of chains of nuclear particles of different lengths. Increasing the size of the chains of emitted particles allows one to realize the state of the system of fission fragments after a long time since the scission of the initial nucleus. Such a procedure can simulate the conditions for post-fission experiments when measurements are carried out after a long "cooling" time of an irradiated sample [30, 31]. The parameters that determine the equilibrium condition for each of the investigated ensembles of nuclear fragments are subsequently deduced from the state of the minimum thermodynamic potential of constant pressure [32], which for the i -th nuclear cluster has the form:

$$G_i = \varepsilon_i - TS_i + PV_i. \quad (2)$$

For the i -th cluster here ε_i is the internal energy, and V_i , respectively, the volume of a two fragments cluster, $V_i = V_1^i + V_2^i$. The ε_i values can be determined by the binding energy of the two-fragment cluster and have the negative values corresponding to the bound states of the nucleons. For different nuclear clusters, its spectrum $\{\varepsilon_i\}$ is an additive quantity in the binding energy, $B(N_{p,i}^j, N_{n,i}^j)$, of the j -th nucleus fragment from the i -th cluster:

$$\varepsilon_i = \sum_{j=1,2} B(N_{p,i}^j, N_{n,i}^j). \quad (3)$$

Here according to (1), $N_{n,1}^i + N_{n,2}^i = A_0 - n_1^i - n_2^i$, $N_{p,1}^i + N_{p,2}^i = A_0 - m_1^i - m_2^i$. Binding energies, $B(N_{p,i}^j, N_{n,i}^j)$, used in (3) are tabulated in [33, 34], and their extrapolation is given, for example, in [35, 36]. The term $P \cdot V_i$ can be written as $P \cdot N^i \cdot \Delta v$, where N^i – the number of nucleons nuclei-fragments of the i -th cluster, $N^i = N_{n,1}^i + N_{n,2}^i + N_{p,1}^i + N_{p,2}^i$, and Δv is the elementary volume of the nucleus per nucleon, which is 40-50 times larger than the size of the nucleon [37]. The constant $P \cdot \Delta v$ is a parameter of the theory. It can be evaluated through the ideal gas law: $P \cdot \Delta V = \Delta N \cdot T$, and $\Delta N \approx 1$, the value of $P \cdot \Delta v$ must be in the same order as T . More detailed value based on neutron emission spectra gives for $P \cdot \Delta v$ range 3-5 MeV [23]. The configurational entropy S_i in (2):

$$S_i = \ln(\omega_i) \quad (4)$$

is calculated through the degeneracy factor ω_i with the account of the statistical nonequivalence of nucleons with different specific binding energy:

$$\omega_i = A_0! / \left(\prod_{j=1,2} K(n_j^i) (N_{p,j}^i! (N_{n,j}^i - n_j^i)!), \quad (5)$$

where n_j^i - number of fission neutrons of the j -th fission fragment from the i -th cluster, $K(n_j^i) = 1/n_j^i!$, $\prod_{j=1,2} x_j! = x_1! x_2!$. From the (5), one can see that with increasing the nuclear temperature T entropy term in eq. (2) is responsible for the increase the symmetry of fission fragments yield. This is since the maximum ω_i is achieved at $N_{p,1}^i = N_{p,2}^i$, $N_{n,1}^i = N_{n,2}^i$. Formulas (1)-(4) allow one to find the probability of realization or yield of two-fragments nuclear clusters of the i -sort. In the isothermal-isobaric ensemble, the probability of a microstate, here two-fragments cluster i , is defined by the term $\exp(-G_i/T)$ [31] and has the form:

$$Y(N_{p,1}^i, N_{p,2}^i, N_{n,1}^i, N_{n,2}^i) = \omega_i \exp \{ -(\varepsilon_i + P \cdot V_i)/T \} / z, \quad (6)$$

where ε_i - is the spectrum values (3) of the two-fragments cluster, ω_i - determines its degeneracy by the formula (5), the partition function of the system z is defined as:

$$z = \sum_i \omega_i \exp \{ -(\varepsilon_i + P V_i)/T \}. \quad (7)$$

The expression (6) refers to the probability of realization only of two-fragment clusters.

The next step is calculating the cumulative fission yields: $F(A, m, n)/F(Z, m, n)$ which is the yield of the post scission fission fragment with mass number A or charge Z produced after at most n neutron and m beta particles emission. The functions $F(Z, m, n)/F(A, m, n)$ are obtained by summing (6) over A/Z for each Z/A for clusters (i) satisfying the conditions:

$$\begin{aligned} N_{p,j}^i &= Z \quad (j = 0 \text{ or } 1) \\ N_{p,j}^i + N_{n,j}^i &= A \quad (j = 0 \text{ or } 1) \\ 0 &\leq \sum_{j=1}^2 n_j^i \leq 2n, \\ 0 &\leq \sum m_j^i \leq 2m \text{ if } m \geq 0, \\ 2m &\leq \sum m_j^i \leq 0 \text{ otherwise.} \end{aligned}$$

The cumulative fission yields distribution is normalised to 200%.

III. RESULTS AND DISCUSSION

The method's possibilities were tested by the example of the even-even isotope ^{232}Th , whose MCSFF has been

well established and recently studied [30, 31]. The procedure of the simulation of the experimental conditions required the building of many ensembles. Each ensemble contains the ^{232}Th isotope fission fragments after the nuclear particles' emission within their length chains. For the j -th nucleus from the i -th cluster, the result of the emission of m_j^i beta particles leads to the transformation $N_{p,j}^i \Rightarrow (N_{p,j}^i - m_j^i)$, where the positive sign is for β^+ and the negative for β^- particles, respectively. After the emission of the neutrons n_j^i : $N_{p,j}^i + N_{n,i}^i \Rightarrow N_{p,j}^i + N_{n,j}^i - n_1^i - n_2^i$. The calculation carried out for the ^{232}Th shows that both the beta particles and the neutron's emission may change the most probable pairs of fission fragments' nucleon composition. It also favors the formation of most probable clusters with closed-shell fragments, having a greater specific binding energy. The neutrons emission leads to an increase in the configuration entropy (5) of the nuclear fission cluster. The most probable are two fragments fission clusters obtained from the minimum of the thermodynamic potentials of Gibbs (2) for a constant pressure ensemble. The results of calculations are given in Tables I and II, containing from $M=1$ to five of the most probable two fragments clusters, both without and when taking into account the emission of nuclear particles.

The calculation was given in the "cold" fission assumption with the value of the nuclear temperature, which we consider as a characteristic of the thermostat, $T = 0.5$ MeV and $P \cdot \Delta v = 3$ MeV. The sign before n is always positive, while the sign before m indicates the sort of beta particles emitted: minus suggests that there is the chain of β^- particles from the $[-m:0]$ range; positive m values relate to β^+ particles with their emission range $[0:m]$. As shown in Tables I and II, some probable nuclear clusters retain their composition for different sets n, m . This is because the probability of yield of the i -th cluster primarily depends on the thermodynamic potential value (2). Some nuclear clusters obtained under the pre-emission of nuclear particles have small G_i values and may prevail even for extended ensembles of fragment nuclei containing all the nuclei fragments formed by the emission of neutron chains $0, 1, \dots, n$ and $0, 1, \dots, m$ beta-particles. The emission of nuclear particles leads to the gain of specific energy ε_i in (2) but can be compensated by a decrease in the entropy factor S_i . The competition of energy and entropy factors determines the probability of realization of the i -th cluster. These conditions explain the stability of the proton/neutron composition of some probable nuclear sets in tables I and II with increasing values of the chains n and m . However, as a trend, it can be seen that increasing the length of the chain of emitted nuclear particles leads to the dominance of more stable isotopes among the most probable clusters. Figure 1 shows the dependencies of the thermodynamic potentials (2), normalized to A_0 , on the length of the chains of emitted nuclear particles for the most probable nuclear clusters $M=[1:5]$, Tables I and II. Such

TABLE I. The evolution of five the most probable two-fragment clusters, $M=[1:5]$, after emission the neutrons of different chains' lengths: n defines all their sets from the ranges $[0:n]$. There is no beta emission.

M/n	0	+1	+2	+3	+4
1	$\{^{100}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$	$\{^{100}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$
2	$\{^{102}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{98}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$	$\{^{94}_{38}\text{Sr}, ^{134}_{52}\text{Te}\}$	$\{^{96}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$
3	$\{^{101}_{40}\text{Zr}, ^{131}_{50}\text{Sn}\}$	$\{^{100}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$	$\{^{98}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{94}_{38}\text{Sr}, ^{134}_{52}\text{Te}\}$	$\{^{94}_{38}\text{Sr}, ^{134}_{52}\text{Te}\}$
4	$\{^{104}_{40}\text{Zr}, ^{128}_{50}\text{Sn}\}$	$\{^{101}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{100}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{98}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$	$\{^{98}_{40}\text{Zr}, ^{130}_{50}\text{Sn}\}$
5	$\{^{99}_{39}\text{Y}, ^{133}_{51}\text{Sb}\}$	$\{^{100}_{40}\text{Zr}, ^{131}_{50}\text{Sn}\}$	$\{^{97}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$	$\{^{97}_{40}\text{Zr}, ^{133}_{51}\text{Sb}\}$	$\{^{97}_{40}\text{Zr}, ^{133}_{51}\text{Sb}\}$

TABLE II. The evolution of five the most probable two-fragment clusters, $M=[1:5]$, after emission of the beta-particles of different chains' lengths: m defines all their sets from the ranges $[-m:0]$ for β^- particles, $[0:m]$ for β^+ , respectively. Two cases are considered: $n = 0$ when the neutron emissions are absent, and for emitted m neutrons for the range $[0:3]$.

M/m	-4	-3	-2	-1	0,+1,+2,+3,+4
$n=0$	1	$\{^{106}_{44}\text{Ru}, ^{126}_{50}\text{Sn}\}$	$\{^{105}_{43}\text{Tc}, ^{127}_{50}\text{Sn}\}$	$\{^{104}_{42}\text{Mo}, ^{128}_{50}\text{Sn}\}$	$\{^{102}_{41}\text{Nb}, ^{130}_{50}\text{Sn}\}$
	2	$\{^{108}_{44}\text{Ru}, ^{124}_{50}\text{Sn}\}$	$\{^{104}_{43}\text{Tc}, ^{128}_{50}\text{Sn}\}$	$\{^{102}_{42}\text{Mo}, ^{128}_{50}\text{Sn}\}$	$\{^{101}_{41}\text{Nb}, ^{131}_{50}\text{Sn}\}$
	3	$\{^{104}_{44}\text{Ru}, ^{128}_{50}\text{Sn}\}$	$\{^{101}_{42}\text{Mo}, ^{131}_{51}\text{Sb}\}$	$\{^{98}_{40}\text{Zr}, ^{134}_{52}\text{Te}\}$	$\{^{100}_{40}\text{Zr}, ^{132}_{50}\text{Sn}\}$
	4	$\{^{96}_{40}\text{Zr}, ^{136}_{54}\text{Xe}\}$	$\{^{100}_{42}\text{Zr}, ^{132}_{51}\text{Sb}\}$	$\{^{106}_{42}\text{Mo}, ^{126}_{50}\text{Sn}\}$	$\{^{100}_{41}\text{Nb}, ^{132}_{50}\text{Sn}\}$
	5	$\{^{114}_{46}\text{Pd}, ^{118}_{48}\text{Cd}\}$	$\{^{106}_{42}\text{Mo}, ^{126}_{50}\text{Sn}\}$	$\{^{100}_{42}\text{Mo}, ^{132}_{50}\text{Sn}\}$	$\{^{99}_{41}\text{Nb}, ^{133}_{51}\text{Sb}\}$
$n=3$	1	$\{^{106}_{44}\text{Ru}, ^{126}_{50}\text{Sn}\}$	$\{^{105}_{43}\text{Tc}, ^{126}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{134}_{52}\text{Te}\}$	$\{^{96}_{40}\text{Zr}, ^{133}_{51}\text{Sb}\}$
	2	$\{^{106}_{44}\text{Ru}, ^{124}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{135}_{53}\text{I}\}$	$\{^{102}_{42}\text{Mo}, ^{128}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{131}_{51}\text{Sb}\}$
	3	$\{^{108}_{44}\text{Ru}, ^{124}_{50}\text{Sn}\}$	$\{^{103}_{43}\text{Tc}, ^{128}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{132}_{52}\text{Te}\}$	$\{^{96}_{40}\text{Zr}, ^{131}_{51}\text{Sb}\}$
	4	$\{^{107}_{44}\text{Ru}, ^{124}_{50}\text{Sn}\}$	$\{^{96}_{40}\text{Zr}, ^{134}_{53}\text{I}\}$	$\{^{100}_{42}\text{Mo}, ^{128}_{50}\text{Sn}\}$	$\{^{95}_{39}\text{Y}, ^{134}_{52}\text{Te}\}$
	5	$\{^{96}_{40}\text{Zr}, ^{134}_{54}\text{Xe}\}$	$\{^{105}_{43}\text{Tc}, ^{127}_{50}\text{Sn}\}$	$\{^{97}_{40}\text{Zr}, ^{134}_{52}\text{Te}\}$	$\{^{97}_{40}\text{Zr}, ^{133}_{51}\text{Sb}\}$

dependencies for different beta particle emission chains, Fig. 1 a), were obtained without ($n = 0$), and with the neutron emission account with a chain length up to $n = 3$. Fig.1 b) shows the same behavior G_i when considering only the neutrons' chains emitted in the ranges $[0:4]$. As can be seen, the emission of electrons β^- in comparison with β^+ particles or fission neutrons significantly reduces the values that increase the probability of such cluster yield. The probabilities of the emergence of new compositions of probable nuclear clusters are also appearing. The positron β^+ emission almost does not lead to changes and redistribution of MCSFF, since practically all ^{232}Th fission fragments are neutron-proficient. The emission of β^- particles causes a more significant effect than the emission of the same number of neutrons. As can be seen from Fig. 1 a) taking into account neutron emission ($n=3$) reduces the G_i value that leads to increasing the probabilities of new nuclear clusters. The dependencies on the beta particle emission are changes too. In particular, the yields of probable nuclear clusters, $M=[1:5]$, taking into account the neutron emission become comparable, Fig. 1 a). The nuclear particles emission also leads to domination in the MCSFF of more stable nuclei. The range of their half-life are changes from several seconds at $n = 0$, $m = 0$ to 371.8 days and $2.30 \cdot 10^5$ years if $n=0$, $m = 4$, Table II. The same tendency occurs to account for the emission of neutron fission. As can be seen from Tables I and II, the heavy fission fragments

peak of MCSFF under considering the neutron emission is retaining. This peak, especially for the charge spectra, formed mainly by $^{130,132}\text{Sn}$ isotopes, and only if $n = 3$, 4 in the spectra have appeared ^{134}Te isotopes. The peak of light fission fragments is mostly formed by $^{94-104}\text{Zr}$ isotopes and is sensitive to beta particles' post-fission emissions. Taking into account beta emission Zr isotopes are replaced by $^{100-102}\text{Nb}$, $^{100,102,106}\text{Mo}$, $^{104,106,108}\text{Ru}$, as well as the isotopes Tc and Pd. Moreover, the inclusion of longer chains of emitted nuclear particles leads to the convergence of the asymmetric peaks, which looks like enhanced symmetric fission mode over asymmetric one and the MCSFF's symmetrization's shape. This tendency has a non-monotonic behavior. There are some peculiarities for emitted β^- particles for m range $(-3, -2)$ where the peak of heavy fission fragments begins to forming by $^{132,134}\text{Te}$, $^{131,132}\text{Sb}$, $^{134,135}\text{I}$ isotopes. Figure 2 shows the results of the MCSFF calculations for the above cases. As one can see, the emission of β^+ particles has almost no effect on their MCSFF compared to β^- particles' emission. As mentioned above, it is because the asymmetric fission fragments of ^{232}Th are neutron-proficient nuclei. The emission of β^+ particles only increases the neutron excess in the nucleus. The corresponding nuclear clusters don't increase the probability of realization than before the emission of β^+ particles. Therefore, the effect of the emission of β^+ particles on the formation of MCSFF is insignificant. When we consider the emission of β^- par-

Fig.1 The dependencies of the reduced thermodynamic potential, G_i/A_0 , for the most probable nuclear clusters, $M = 1-5$, see Tables I, II on the length of the chains of emitted nuclear particles.

Here:

a) are the G_i/A_0 , dependencies on the chain length m of beta particle emission ($-m, -m+1, \dots, 0$) for β^- and $(0, 1, \dots, m)$, respectively, for β^+ particles. The dependencies are constructed when there is no neutron emission, $n = 0$, and when $n = 3$ (see table II);

b) G_i/A_0 dependencies on the chain length n when accounting the neutron emission only (see table I).

ticles, another nature of the MCSFF change will occur. In this case, there are significant changes in the MCSFF, Fig. 2a) and the redistribution among the most probable fission fragments, see Table II. All this causes a change in the topology of the MCSFF. At $n = 0$, the peak of the light distribution fragments is formed by Zr isotopes. If one considers the beta- particles' emission, this peak mainly became would be created by the isotopes Nb, Mo, Tc, Ru and Pd, with their most significant tendency to the filled proton shells $Z = 50$. If the post-scission neutron emission is only considered, some peculiarity of the MCSFF occurs for n range (2, 3), Fig. 2c), f). In this case, the light fission fragments' peaks are formed not by the Zr isotope only but also the Sr ones, see Table I. For the peak of the heavy fission fragments, if we consider the neutron's emission, only the isotopes of Sn can be changing on the mentioned isotopes Te. These all neutron emissions lead to growing the configuration entropy of the nuclear clusters, affecting MCSFF symmetrization.

Fig. 2 shows that nuclear particles' emission intensifies the convergence of the asymmetric peaks, which looks like enhanced the symmetry mode of the MCSFF yields. Herewith, the proportionality of two peaks shape for the light, and heavy nuclei-fragments can be violated. This state is because the peak of heavy fission fragments within the magic number 50 is formed by the most massive amount of two fission fragment clusters than the corresponding light ones with Z from the range (41, 43). For mass spectra, the peak of heavy fission fragments

near $A = 132$ is the dominant one and formed by also the most massive sets of fragments closer to the doubly magic nuclei Sn ($^{132}_{50}\text{Sn}$). Genetically related light fission fragments sets have an A range (96 - 100). As can be seen from Fig. 2d, the post scission emission of beta-particles affects the mass spectrum of the fission fragments and leads to their symmetrization. It is because the growth of the emission chain of β^- particles leads to an increase in the charge Z of light fragments of the most probable nuclear clusters, table II. Due to Unchanged Charge Density (UCD) Assumption or Hypothesis [38], their mass increases simultaneously (A_1). The mass of the heavy fragment (A_2) decreases from the conservation condition (1): $A_0 = A_1 + A_2$.

It is very useful to investigate how the MCSFF transformation after the post-fission emission of nuclear particles affects the isotopic spectra and values of the mean value of Z for a given A , $\langle Z \rangle = \langle Z \rangle(A)$. Similar dependencies for the ^{232}Th isotope were established in the experiment [39, 40]. In this work, the $\langle Z \rangle$ calculations were performed using isotope spectra for each chemical element with a fixed Z . They have shown that all such spectra are substantially varied, taking into account the post-fission emission of nuclear particles. Figure 3a) presents the results of such calculations for the Rh isotope spectra: $F(Z=45, A, m, n)/F(Z=45, m, n)$, which are a reliable marker of the fission of actinide nuclei. The sum of Y of the Rh isotopes yields were 100% normalized, and all data were fitted using the Gaussian distributions. As can be seen, the emission of nuclear particles shifts the isotopic spectra of Rh in Fig. 3a) to a value corresponding to beta-stable nuclei from (8). This distribution was

Fig. 2. The charge a), b), c) and mass d), e), f) fission fragments yields spectra of the ^{232}Th obtained with consideration only the beta particles a), d), and the neutrons c), f) with different emission chains length. The b) and e) are dependencies obtained for the more general case when considering both the beta particles and the neutron emission chains with a capacity of $n = 3$.

used to find value for $\langle Z \rangle$, considering the average characteristics of the isotope spectra of the Rh isotopes. The parameters for the beta particle and neutron chains were selected from Tables I, II. Fig. 3b) demonstrates the dependence of $\langle Z \rangle$ on the masses of the fission fragments A obtained with the same emission parameters as in Fig. 3a). As one can see, the most significant changes of $\langle Z \rangle$ from curve 1 to 2 occur for isotopes that do not contain magic numbers $Z = 50, 82$. The emission of nuclear particles shifts the dependences of $\langle Z \rangle = \langle Z \rangle(A)$ from the values realized in the pre-emission case, ($n=0, m=0$) and highlighted as 1, to dependence 2, determined from the liquid drop model for beta-stable isobars [38]:

$$Z_{LD} = \frac{A}{1.98 + 0.015 \cdot A^{2/3}}. \quad (8)$$

According to UCD hypothesis, the proton shell closure $Z=50$ play a role for primary fragments around $A=130$ since $(232/90) \cdot (50 + 0.5) = 130$. Thus the proton shell closure $Z=50$ prevents (A) to shift toward the curve 2 in the range $A = 120 - 140$. Moreover, such trends, Fig. 3b) are more pronounced, by taking into account the emission of beta particles than neutrons.

Finally, it is important to compare the obtained

theoretical data, Figs. 2, with the results of recent experiments where the ^{232}Th fission product yields were investigated [30, 31]. The nuclear experiment conditions should be taken into account: are the yields of fission fragments established immediately after irradiation of the fission target ^{232}Th , or afterward. In the latter case, the fission products contain isotopes that are formed after the emission of beta particles or neutrons. For the mentioned experiments for ^{232}Th fission, the irradiation/cooling times for the photonuclear (γ, f) reactions [30], were approximately 0.5/0.5 hour. The same for (n, f) reactions on fast neutrons were 1.0/1.0 hour [31]. Another feature is related to the conditions of gamma-spectrometric measurements during this experiment. The authors identified more than 48 fission products, the γ -ray abundance in which was not less than 10%. Ultra short-lived radionuclides were not mentioned, only fission products with a half-life of 15 min. As we say above, the proposed scheme of calculations allows one to indirectly simulate the conditions of the MCSFF study's experiments, since the increase in the chains' length after the emission of neutrons or beta particles can be realized over long time intervals. Also promising is the ability to simulate the

Fig. 3a) Isotope yield spectra $F(Z, m, n)$ of Rh nuclides obtained without and accounting the post-scission emission of nuclear particles. The m, n values for investigation were taken from tables I, II. LD marker obtained from equation (8).

Fig. 3b) Dependencies between the masses A of fission fragments and their mean value charges $\langle Z \rangle$ for different m, n values. Curve 1 is constructed for the pre-emission ($m = n = 0$) state of nuclear fragments, 2, respectively, is obtained from formula (8) as for beta-stable isobars.

experimental conditions through the temperature, T , of the ensemble of nuclei-fragments, which can also be related to the cooling time: the longer the time lower the temperature [41]. The many ensemble methods can simulate the real measurement conditions with long exposure times and sample cooling. The data given in table II comparison with the nuclides identified in [30] indicates suitable matches of the calculated and experimental data on the yields of a number of radionuclides ($^{104}_{43}\text{Tc}$, $^{128}_{50}\text{Sn}$, $^{131}_{51}\text{Sb}$, $^{132,134}_{52}\text{Te}$, $^{134,135}_{53}\text{I}$). The absence of some fission fragments of ^{232}Th , given in tables I, II among those identified in [31], it can be explained either they are short-lived

$^{94}_{38}\text{Sr}$, $^{95,99}_{39}\text{Y}$, $^{100,101}_{40}\text{Zr}$, $^{100,101,102}_{41}\text{Nb}$, $^{104}_{42}\text{Mo}$, $^{103,105}_{43}\text{Tc}$, $^{130,132}_{50}\text{Sn}$,
or not gamma active

$^{96,98}_{40}\text{Zr}$, $^{100,102,106}_{42}\text{Mo}$, $^{100,101}_{40}\text{Ru}$, $^{114}_{46}\text{Pd}$, $^{118}_{48}\text{Cd}$, $^{124,132,133}_{51}\text{Sb}$.

And finally, about the discussion of the two- or three- humps topology of MCSFF for actinides, in particular, ^{232}Th . As shown in Fig. 2, the MCSFF is formed as a result of the competition of symmetric and asymmetric fission modes. With increasing the cooling time and the length of the chains of emitted particles, there is the convergence of the asymmetric peaks, which looks like enhanced symmetric fission mode over asymmetric one for fission product yields. This trend is common to all of the actinides. The obtained results can explain the dependence of the MCSFF topology on the conditions of experimental studies. Therefore, the spectra presented in Fig. 2, allow evaluating the

possibilities of experimental methods for MCSFF studies.

IV. CONCLUSION

Thus, the proposed statistical approach and “the many ensembles method” show the possibility to investigate how the post-scission emission of the nuclear particles influences the MCSFF topology, isotopic spectra, and distributions of the most probable charges. The thermodynamic approach used in the paper does not contain time variables. However, the time evolution of the MCSFF topology can be investigated indirectly through many ensembles’ characteristics. Each of these ensembles is obtained by considering the emission of nuclear particles. The length of the emitted beta particle and neutron chains can simulate the time duration for a system of two-fragment clusters after fission. The theory allows us to establish the most probable two fragment clusters after the emission of nuclear particles and investigate their stability parameters. These results are essential for interpreting known experimental studies concerning actinide fission, especially the ^{232}Th isotope. There is a satisfactory matching of the calculated and experimental data obtained for ^{232}Th fission fragments. The explanation of why the experiment does not observe the other most probable fission fragments of this isotope was proposed. These results are also essential to testify the experimental abilities for the MCSFF investigation, particularly by taking into account the “cooling” time in post-fission experiments, presence of gamma-active nuclides.

ACKNOWLEDGEMENTS

The authors are grateful V. Denysov for assistance and discussions with this work.

-
- [1] V. Denisov and V. Plyuyko, *Problems of nuclear physics and nuclear reactions*, 41, Vol. 45 (Kyiv University Publishing and Printing Center, Kyiv, Ukraine, 2013) p. 430 pages, https://inis.iaea.org/collection/NCLCollectionStore/_Public/45/091/45091761.pdf (visited 2020-09-04).
- [2] A. Andreyev, K. Nishio, and K.-H. Schmidt, Reports on Progress in Physics **81**, 016301 (2018).
- [3] M. Bender, R. Bernard, G. Bertsch, *et al.*, “Future of nuclear fission theory,” (2020), arXiv:2005.10216 [nucl-th].
- [4] A. Aprahamian, K. Langanke, and M. Wiescher, Progress in Particle and Nuclear Physics **54**, 535 (2005).
- [5] B. Tathagata, S. Nath, and S. Pal, Phys. Rev. C **99**, 024610 (2019).
- [6] N. Carjan, F. Ivanyuk, and Y. Oganessian, Phys. Rev. C **99**, 064606 (2019).
- [7] *Fission Product Yield Data for the Transmutation of Minor Actinide Nuclear Waste*, Non-serial Publications (INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna, 2008).
- [8] Y. Gangrsky, B. Dahsuren, and B. Markov, *Fragments of nuclear fission* (Energoatomizdat, Moscow, 1986) 172 pages.
- [9] A. Chatillon, J. Taïeb, J.-F. Martin, E. Pellereau, *et al.*, EPJ Web of Conferences **111**, 08001 (2016).
- [10] K.-H. Schmidt and B. Jurado, Reports on Progress in Physics **81**, 106301 (2018).
- [11] P. Gysbers, G. Hagen, J. D. Holt, G. R. Jansen, T. D. Morris, P. Navrátil, T. Papenbrock, S. Quaglioni, A. Schwenk, S. R. Stroberg, and K. A. Wendt, Nature Physics **15**, 428 (2019).
- [12] F. A. Ivanyuk, C. Ishizuka, M. D. Usang, and S. Chiba, Phys. Rev. C **97**, 054331 (2018).
- [13] Vardaci, E., Di Nitto, A., Nadtochy, P.N., La Rana, G., Cinausero, M., Prete, G., Gelli, N., Kozulin, E.M., Knyazheva, G.N., Itkis, I.M., Ashaduzzaman, M., Davide, F., Pulcini, A., Quero, D., and Rath, P.K., EPJ Web Conf. **193**, 01002 (2018).
- [14] Tamagno, Pierre, Bouland, Olivier, Serot, Olivier, and Moller, Peter, EPJ Web Conf. **146**, 04013 (2017).
- [15] P. Möller, J. Nix, and W. Swiatecki, Nuclear Physics A **469**, 1 (1987).
- [16] J. Sadhukhan, W. Nazarewicz, and N. Schunck, Phys. Rev. C **93**, 011304 (2016).
- [17] D. Regnier, N. Dubray, and N. Schunck, Phys. Rev. C **99**, 024611 (2019).
- [18] A. Bulgac, S. Jin, K. J. Roche, N. Schunck, and I. Stetcu, Phys. Rev. C **100**, 034615 (2019).
- [19] G. Scamps, C. Simenel, and D. Lacroix, Phys. Rev. C **92**, 011602 (2015).
- [20] S. T. Holloway, T. Kawano, and P. Moller, J.Korean Phys.Soc. **59**, 875 (2011).
- [21] M. Lammer, “Fission product yields: Minor actinides up to 150 mev,” in *Fission Product Yield Data for the Transmutation of Minor Actinide Nuclear Waste* (INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 2008) pp. 1–6, http://www-pub.iaea.org/MTCD/Publications/PDF/Pub1286_web.pdf.
- [22] H. Zhang, J. Dong, G. Royer, W. Zuo, and J. Li, Phys. Rev. C **80**, 037307 (2009).
- [23] V. Maslyuk, O. Parlag, O. Lendyel, T. Marynets, and M. Romanyuk, Journal of Modern Physics **4**, 1555 (2013).
- [24] E. Bogomolova, A. Grashin, A. Efimenko, and I. Lukasevich, Nuclear Constants , 054607 (1995).
- [25] K.-H. Schmidt and B. Jurado, Phys. Rev. C **83**, 014607 (2011).
- [26] V. Maslyuk, O. Parlag, O. Lendyel, T. Marynets, M. Romanyuk, O. Shevchenko, J. Ranyuk, and A. Dovbnya, Nuclear Physics A **955**, 79 (2016).
- [27] V. T. Maslyuk and A. V. Smolyanyuk, EPL (Europhysics Letters) **112**, 52001 (2015).
- [28] J.-F. Lemaître, S. Goriely, S. Hilaire, and J.-L. Sida, Phys. Rev. C **99**, 034612 (2019).
- [29] V. Maslyuk, O. Parlag, M. I. Romanyuk, O. I. Lendyel, T. Marinetc, O. G. Laver, and I. F. Povkhan, Phys. Rev. C **98**, 064608 (2018).
- [30] H. Naik, A. Goswami, G. N. Kim, M. Lee, K. Kim, S. Suryanarayana, E. Kim, S. Shin, and M.-H. Cho, Phys. Rev. C **86**, 054607 (2012).
- [31] H. Naik, G. N. Kim, K. S. Kim, and M. Zaman, Phys. Rev. C **100**, 014606 (2019).
- [32] L. Zhirifalko, *Statistical Physics of Materials* (John Wiley & Sons, New York, 1973) 382 pages.
- [33] N. Wang and M. Liu, Phys. Rev. C **84**, 051303 (2011).
- [34] M. Wang, G. Audi, A. Wapstra, F. Kondev, M. MacCormick, X. Xu, and B. Pfeiffer, Chinese Physics C **36**, 1603 (2012).
- [35] G. Audi, F. Kondev, M. Wang, W. Huang, and S. Naimi, Chinese Physics C **41**, 030001 (2017).
- [36] G. Audi, M. Wang, A. Wapstra, F. Kondev, M. MacCormick, X. Xu, and B. Pfeiffer, Chinese Physics C **36**, 1287 (2012).
- [37] O. G. Sitenko and V. Tartakovsky, *Theory of nucleus, Nuclear Structure and Nuclear Interaction*, Vol. 84 (Springer Netherlands, Dordrecht, 1997) 615 pages.
- [38] Mazurek, K., Nadtochy, P. N., Ryabov, E. G., and Adeev, G. D., Eur. Phys. J. A **53**, 79 (2017).
- [39] D. Kaji, S. Goto, M. Fujita, T. Shinozuka, M. Fujioka, and H. Kudo, Journal of Nuclear and Radiochemical Sciences **3**, 7 (2002).
- [40] A. Wahl, *Fission Product Yield Data for the Transmutation of Minor Actinide Nuclear Waste*, Systematics of fission product yields (International Atomic Energy Agency, Vienna, 2008) pp. 117–148.
- [41] The authors are grateful to U.R. for the proposed idea.