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NATURE AND PROPERTIES OF THE FISSION MODES OF THE NEUTRON DEFICIENT ^{220,224,226}Th NUCLIDES

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Introduction

During the past 10 years, investigations of the mass and the energy distributions in the vicinity of ^{208}Pb using relatively light projectiles have yielded two important conclusions [1]. First of all, it is possible to circumvent the natural obstacle associated with the lack of stable *Po-Fr* target nuclei. Second, overcoming this hurdle makes it possible to study the evolution of the mass asymmetry mode in the fission of lighter nuclei so that one can delineate the physical boundary associated with multimodal fission phenomena.

The parameters of the measured mass-energy distributions of the fragments appear to be in good agreement with the theoretical calculations of V.V.Pashkevich [2]. This suggests that one can probe a new property in fission physics - the valley structure of the barrier. Experimental studies of this property will make it possible to trace out the evolution of the fissioning nucleus between its two critical shapes - the transition state (saddle point) and the scission point. Following this evolution from beginning to end will provide new insights into the mechanism by which the mass and energy distributions are formed. However, this time it is being applied to a significantly expanded region of nuclei. Work by Hulet *et al.* [3] in the Z=100 region has revealed peculiarities in the fission mass and energy spectra which are also suggestive of bimodal structure. These observations can be interpreted as being due to the influence of the valley structure (produced by shell effects) during the descent from saddle to scission.

An obvious question arises in connection with the above results. Is the phenomenon of multimodal fission associated with special circumstances characteristic of the regions around *Pb* and *Fm*, or is it a universal property of fission?

The region of transitional nuclei from *At* to *Th* offers virgin territory for further explorations of multimodal fission phenomena. In this range of nuclei, one can expect dramatic changes in the fission properties as the nucleonic composition and the excitation energy are varied. The transition from mass-symmetric to mass-asymmetric fission and the accompanying changes in the barrier heights and the transition times from saddle to scission for the different fission modes will strongly influence observables such as the fission mass, energy and charge distributions, the fission cross sections, the angular distributions, the pre- and post-fission neutron multiplicities, and the γ-multiplicites. Experimental data on these properties will provide critical testing grounds for theories of both the static and dynamical aspects of the fusion-fission process.

The first experiments on studying the low-energy fission of transition nuclei were started 3 years ago at the FLNR of JINR [4] and then continued in Catania by employing sub-barrier fusion-fission reactions and at GSI by employing fission reactions on secondary beams of radioactive nuclei [5].

The paper presents the results of studying the modal structure of mass and energy distributions of the fission fragments of neutron deficient nuclides ${}^{220}Th$, ${}^{224}Th$ and ${}^{226}Th$ produced in reactions with ${}^{16}O$ and ${}^{18}O$ ions at energies near and below the Coulomb barrier.

It is worth noting that these data do not cover the whole material collected up to date and, first of all, the measurement results concerning the multiplicities of pre- and post-fission neutrons and gamma-quanta in relation to mass fission asymmetry, which are to be presented in future on their analysis being completed.

Experiment

The central problem with studying the low-energy fission (E^{*}< 20-30 MeV) of transition nuclei in reactions with heavy nuclei is that the fusion cross section, σ_{fus}, decreases exponentially, and, along with it, so does the fission cross section, σ_f at ion energies below the Coulomb barrier.

For the reaction ${}^{208}Pb + {}^{16}O \rightarrow {}^{224}Th$ the experiment [6] shows that over the range of excitation energies, near and below the Coulomb barrier, E^{*} = 20-30 MeV (E_i=72-83 MeV), being of interest to us, the fission cross section changes by a factor of five orders of magnitude from 10^2 mbarn to 10^{-3}. This places definite, sufficiently severe constrains on the techniques of the experiment and the energy characteristics of an ion beam. While studying the fission modes over the discussed region of nuclei another important point is reaching a compromise between the necessity of employing the maximal possible intensity of an ion beam and a high requirement to the resolution of a spectrometer measuring the masses and energies of fission fragments, pre- and post-fission neutron emissions and gamma-multiplicity.

Basing on this consideration, the experiments on measuring the mass and energy distributions of fission fragments which were started on the U-400 accelerator in Dubna and employed the DEMAS fission spectrometer were then continued on the tandem in Catania. For these experiments the new DEMAS-3 spectrometer was developed and produced. It is intended for measuring the mass, energy and angular distributions of fission fragments in coincidence with neutrons and gamma-quanta by a method based on the time-of-flight registration of the correlated reaction products. The diagram of the facilities is shown in figure 1.

Its basis is formed by a two-armed time-of-flight spectrometer of fission fragments. The spectrometer comprises 4 wide-aperture position sensitive avalanche counters, PSACs, utilized as stop detectors and 2 plane-parallel avalanche counters, PPACs, utilized as start detectors in each channel.

The angular and energy characteristics of neutron emission were measured by 8 neutron detectors of the DEMON facility. The detectors were installed 129 cm away from the target at different angles.

Measuring the multiplicity of gamma-quanta was carried out by six 63×63 mm NaJ gamma-detectors being at 14 cm distance away from the target centre.

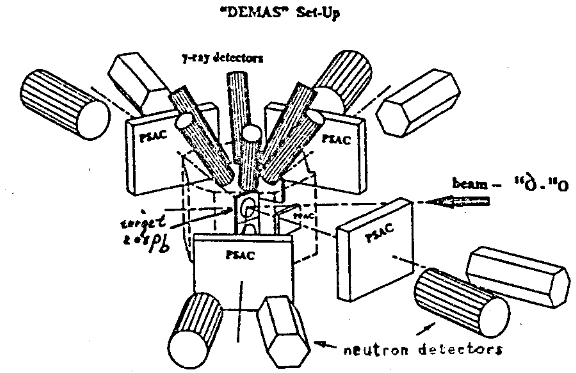

Figure 1

Results and analysis

Presented in Table 1 are the principal characteristics of the investigated reactions and the experimental first moments of the total energy and mass distributions of fission fragments: where E_i is ion energy, E^{*} - the excitation energy of a compound nucleus, E_f - the fission barrier height, E^{*}_{sp} - the excitation energy of a fissioning nucleus at the saddle point, $<E_k>$ - the mean kinetic energy of fission fragments, σ^2_E and σ^2_M - the dispersions of energy mass distributions of fission fragments.

Table 1

Nucleus	E_i, MeV	E^{*}, MeV	E_f, MeV	E^{*}_{sp}, MeV	E_k, MeV	σ^2_E, MeV²	σ^2_M (exp), (amu)²	$\Sigma Y_s/\Sigma Y_a$
${}^{208}Pb+{}^{18}O$	78	26.1	7.0	18.1	165.4±1.1	137±6	280±15	4.1±0.5
	75	23.3		16.3	165.2±2.0	150±11	291±22	2.7±0.8
${}^{208}Pb+{}^{16}O$	108	53.8	7.2	46.6	162.4±1.0	137±9	224±15	-
	85	32.4		25.2	164.0±1.1	114±7	193±13	8.0±1.0
	77	25.0		17.8	163.6±1.2	98±6	187±12	4.8±0.5
	75	23.2		15.9	164.7±1.4	94±6	202±13	3.3±0.4
${}^{204}Pb+{}^{16}O$	108	55.6	8.8	46.8	162.1±1.0	151±10	236±15	-
	85	34.3		25.5	163.1±1.2	118±7	188±12	7.6±0.8
	77	26.9		18.1	163.6±1.2	102±6	166±11	6.0±0.7
	75	25.0		16.2	163.7±1.4	103±6	162±11	6.3±1.0

Shown in figure 2 are the outline plots of *TKE-M* for ^{220}Th, ^{224}Th, ^{226}Th. They show that, at the same excitation energy, the contribution of the asymmetric fission increases significantly with the number of neutrons in a fissioning nucleus.

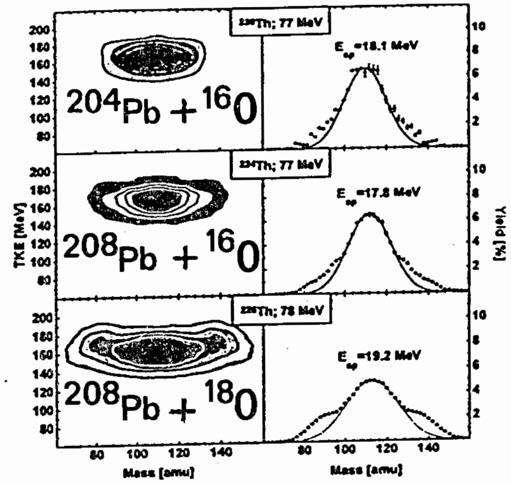

Figure 2

An interesting peculiarity about the obtained dependencies, *Y(M)*, *TKE(M)* and σ^2_E(M) for ^{224}Th and ^{226}Th, is the presence of a structure in the region of *M*>130 related to two asymmetric fission modes appearing: a_0 and a_1 of the mean masses M_{a0}=132 and M_{a1}=140. To estimate the contributions of each mode (the symmetric mode and two asymmetric modes) of the integral mass distributions, the three-component analysis was carried out assuming the partial distributions, $Y_i(M)$, being of the Gaussian form.

The results of the three-component analysis of the ^{220}Th, ^{224}Th and ^{226}Th fragment mass distributions are presented in Table 1 and in figure 3.

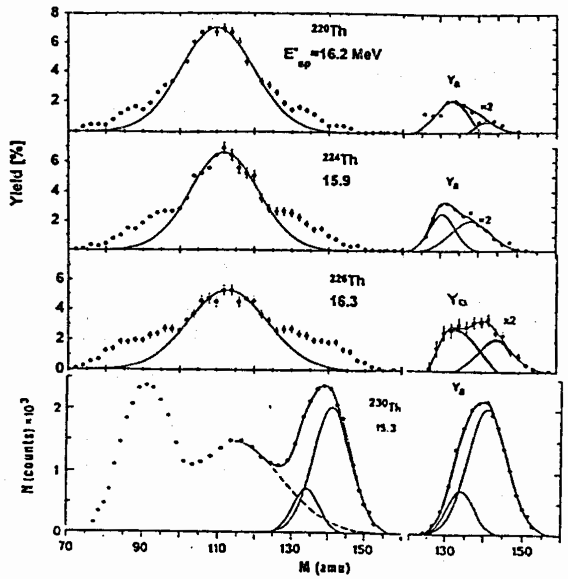

Figure 3

The dependence of ratio $Y^{\prime}_a/Y^{\prime}_s$ on the excitation energy of the fissioning nuclei at the saddle point E^{*}_{sp} is shown in figure 4. Together with our experimental data the analysis

results of the mass distributions for heavier *Th*-isotopes studied in the reactions with neutrons, γ-quanta and ^4He-ions are also shown. The values $Y^{\prime}_a/Y^{\prime}_s$ for all studied nuclei at the same excitation energy $E^{*}_{sp}\approx$16 MeV are shown on insert in figure 4.

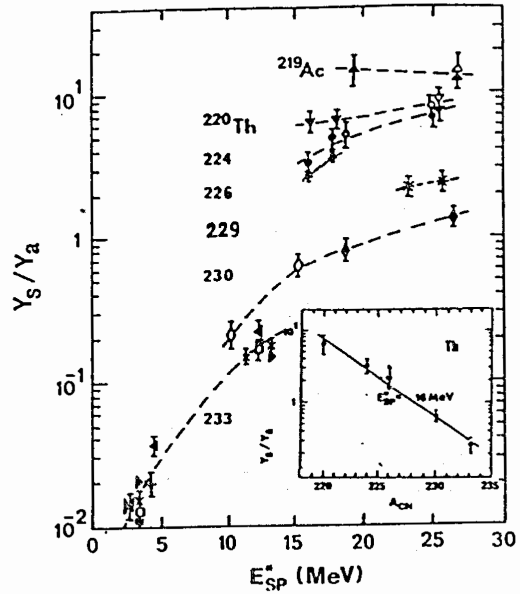

Figure 4

At the changing of free energy of the fissioning nucleus *Q-TKE*, i.e. at selection fission events for different intervals of *TKE* very important properties of fragment mass distributions are manifested. Figure 5 shows the differential mass distribution of ^{224}Th and ^{226}Th at the excitation energy E^{*}=26 MeV. It is easily observed that in the case of ^{226}Th at changing total kinetic energy *TKE* from 146 MeV up to 194 MeV a strong transformation of mass distributions from a symmetric shape, through the three-hampered one, into the asymmetric one with relation peak-valley >3 takes place. On the other hand, for ^{224}Th the symmetric fission mode dominates up to most high *TKE* values. So, if the nucleon composition of a fissioning nucleus is changed by two neutrons, the transition is observed from symmetric to asymmetric fission at free energy *Q-TKE* striving for zero. Obviously, that observed effect is of the same scale as that of the spontaneous fission of ^{256}Fm and ^{258}Fm nuclei (ΔN=2). It means that in spite of the excitation energy being high enough (E^{*}=26 MeV), peculiarities of potential energy deformation surface for each of these *Th*-isotopes are to very strongly affect the formation of fragment mass and energy distributions on the whole and fission modes in particular.

In order to understand the nature and the properties of fission modes the analysis of the differential spectra of total kinetic energy as function of mass asymmetry parameter is very important. For this purpose we have analyzed the experimental spectra *N(TKE)* of ^{226}Th (E_i=78 MeV) which are shown in figure 6 for three regions of mass split *M*=*A*/2=112-114, *M*=132(134) and *M*=142. It is seen, the spectra are differed by their parameters, namely, by the average total kinetic energy *TKE* and its dispersion δ_{TKE}. When one of the fission modes is dominant in the total spectrum, for instance, symmetric (*M*=112) or asymmetric a_0 (*M*=142), the gaussian shape and essentially smaller dispersion δ_{TKE} are present in comparison to the M=132 or 134 a.u.m. spectrum where the contributions of all fission models (s, a_0 and a_1) are comparable.

The analysis of the former spectra (M=134) has been performed and its results are shown in figure 6 and given in Table 2. As seen from Table 2 the components are essentially differed in the mean total kinetic energies and its dispersions and, consequently, by their configurations at the scission point. This observation is a strong proof of the phenomenon of the independent fission modes.

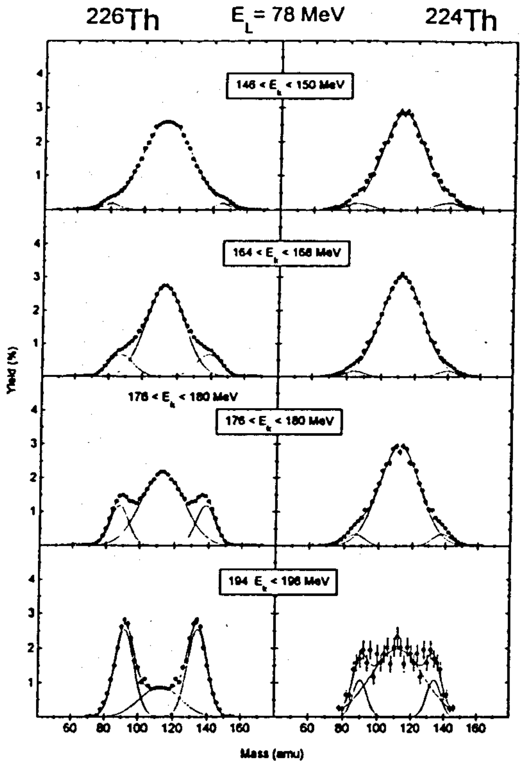

Figure 5

Fission modes differ not only in the shape of partial mass distributions $Y^i(M)$ and the value of the average total kinetic energy of fragments TKE_i, but also they must have different saddle points [2,7] in the corresponding valleys of potential energy surface which are responsible for the origin of this phenomenon. It means that mass-symmetric and mass-asymmetric fission modes, their corresponding yields Y_i^t, contributions to the total fission cross section T_f or fission probability P_f and their energy dependences will be defined by the fission barrier with different parameters, first of all the height of barrier E_f^i. For some transitional nuclei near β-stability (Ra, Ac, At) the difference between E_f^s and E_f^a has been observed experimentally [1,8].

In our case the method of the transition state (Bohr-Wheeler formula) can be used for description of the ratio $Y_i^t(E^*)$ or, which is the same, of the ratio of the

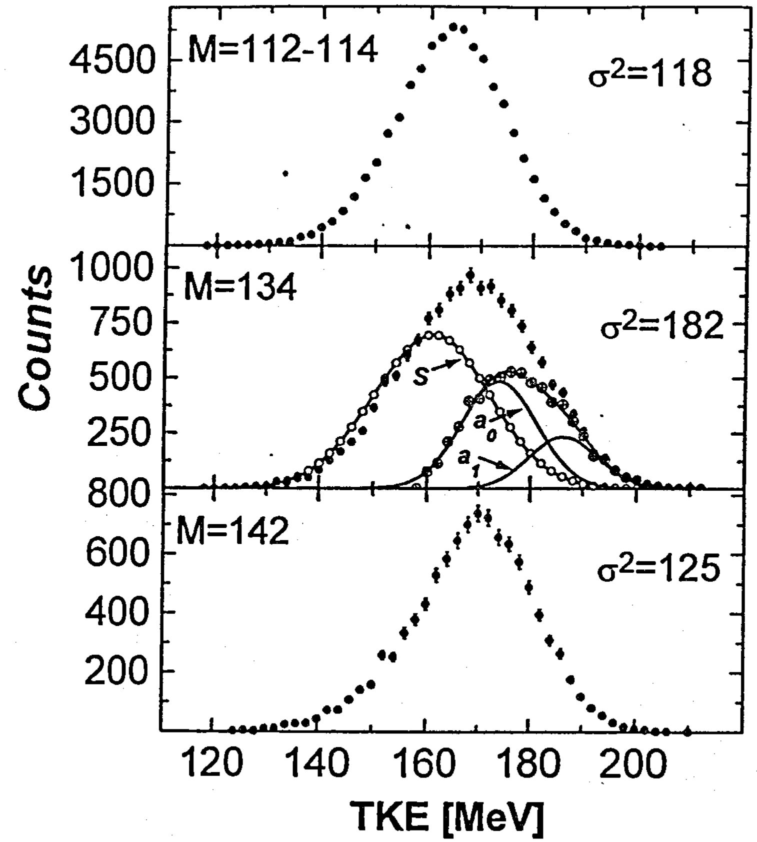

Figure 6

Table 2

	M=132			M=134		
	s	a_0	a_1	s	a_0	a_1
TKE, MeV	161	173.9	188.5	160	173.8	186.2
σ_{TKE}, MeV	26.0	16.5	15.3	26.0	14.6	13.1
Y_s^t, %	0.67	0.26	0.7	0.6	0.28	0.12

average fission widths for distinct fission modes $\Gamma_f^i(E^*)$ and for definition of the value $E_f^s - E_f^a$

$$Y_a^i(E^*)/Y_s^i(E^*) = \frac{\Gamma_f^a(E^*)}{\Gamma_f^s(E^*)} = \frac{\int_0^{E^*-E_f^a}\rho_a(u)du}{\int_0^{E^*-E_f^s}\rho_s(u)du} \quad (1)$$

For $E^* = const$ we have the following approximation

$$Y_a^i / Y_s^i \cong \frac{K_{rot}^a}{K_{rot}^s}\exp\left(\frac{E_f^s - E_f^a}{T}\right) \quad (2)$$

where K_{rot} is the rotating coefficients of the collective enhancement of level density, $\rho(u)$ - internal level density of a nucleus for which in (1) the constant temperature is used, T - nuclear temperature.

The values of $E_f^s - E_f^a$ deduced from relation (2) for different coefficients $C_I = K_{rot}^a / K_{rot}^s$ [9] are given in Table 3. It can be concluded that the quantity (difference of the symmetric and asymmetric fission barriers) for $C_I = 0.7$ (axial and mirror symmetries are contained) and $C_I = -3.3$ (not rotating symmetry) are not in agreement with the experiment because in the first case symmetric fission is predicted to dominate up to ^{230}Th which is not correct, in the second case asymmetric fission is predicted to dominate for all nuclei which is not also correct. So, we conclude that only values $E_f^s - E_f^a$, deduced with parameter $C_I = -1.9$, corresponding to the pear shapes of *Th* nuclei in the ground state, are in agreement with the experiment.

Table 3

Compound nucleus	$(E_f^s - E_f^a)^{exp.}$, MeV			$(E_f^s - E_f^a)^{theor.}$, MeV
	$C_I = -1.9$	$C_I = 0.7$	$C_I = -3.3$	
^{220}Th	-0.3	-2.57	1.3	-1
^{224}Th	+0.8	-2.2	2.4	+0.9
^{226}Th	1.15	-1.95	2.75	+1.2
^{230}Th	2.6	-0.4	4.2	+2.9
^{232}Th	3.2	0.6	4.8	+3.1
^{233}Th	3.7	1.1	5.3	

Discussion of the results

As is mentioned above, the key concept underlying the understanding of fission modes is a potential energy of deformation as function of the nuclear shape. To describe qualitatively the results of the symmetric and asymmetric yields of the mass distributions of the thorium isotopes fission fragments the potential energy surfaces were considered in the space of deformation parameters which specify the nuclear shape in the coordinate system associated with Cassinian ovals [2]. In this parametrization the liquid-drop model shapes along the fission path are described as a fast convergent series.

In the calculation taking into account, the shell correction the higher harmonics of up to the 7th order were used.

The barrier structures in ^{220}Th and ^{226}Th can be seen in figure 7, where the potential energy surface as a function of the elongation parameter α and the asymmetry parameter α_3 is depicted. At each point of the surface the minimization with respect to the parameters α_4 - α_7 was carried out.

Neither the ground state and the second minimum, nor the barrier between them which is usually called the barrier *A* are shown in these figures. The fission valley ascends from the mirror symmetrical second minimum to the barrier *B* which becomes visible at considerable asymmetric shapes in both parts of figure 7 in the depth of the figures and then passes through a very shallow third minimum which has been experimentally observed in the heavier thorium isotopes. The surface is completely symmetrical with respect to a changing sign of α_3 so that only one of the two valleys corresponding to asymmetric shapes of different space orientation is explicitly discussed here. Details of the landscape in the vicinity of barrier *C* are not seen from the position chosen if figure 7 and are different for various isotopes. During the descent a shift of the valley to the smaller asymmetry is seen for ^{220}Th and even the existence of the two valleys with a very small ridge between them is seen for ^{226}Th.

A symmetrical-shape valley is seen to begin at $\alpha_2 = 0.7$. It is running almost parallel to the asymmetric-shape valley. Still another shallow minimum exists in its course to the separated fragments for ^{226}Th. The valley may be populated only from the asymmetric valley over a pass on a very thin ridge that separates the two valleys. That ridge is apparently due to the shell effects and should disappear soon with the heating resulting in a dramatic increase in the symmetrical yield as it is known for the heavier nuclei.

The same type of the potential energy surface was analyzed for all thorium isotopes beginning from ^{220}Th to ^{232}Th. All of them have analogous barrier structures, the difference being only in the details of the landscape on the bottoms of the asymmetric and symmetrical valleys and in the relative height of the third

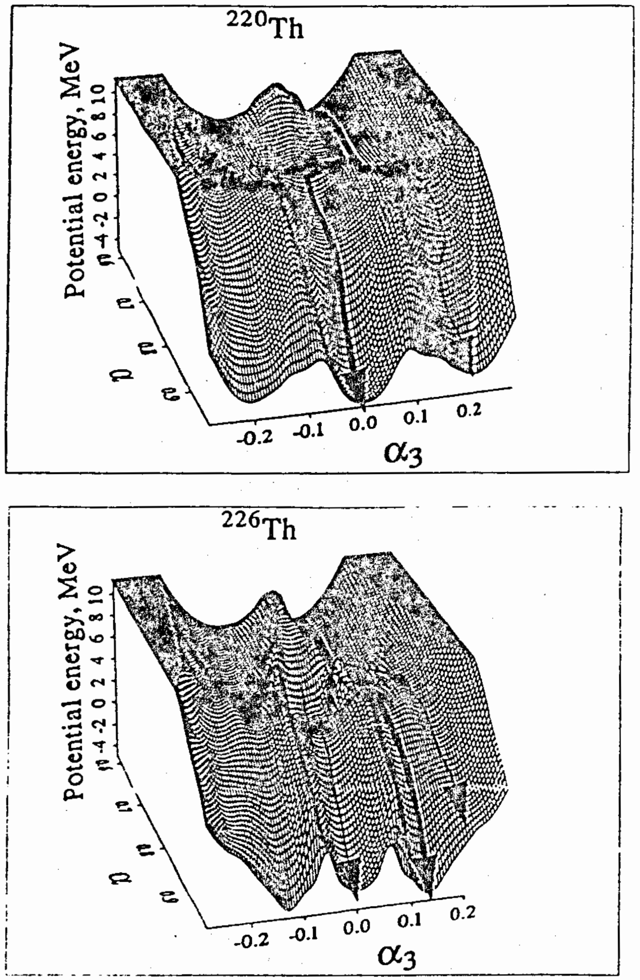

Figure 7

fission barrier *Ec* beyond the asymmetric valley and the pass *Es* from asymmetric to symmetrical valley. That quantity is crucial for the fission fragments mass distribution and determines the ratio of the symmetrical yield to the asymmetric one. The height of the fission barrier *Ec* with respect to the pass from the asymmetric to symmetrical valley *Es* is shown in figure 8. It is seen to be positive for the light isotopes and negative for the heavier ones which qualitatively and quantitatively (see also Table 3) explains the transition from the symmetrical to

asymmetric fission fragment mass distribution observed for these isotopes and described in the previous section. The exact position of the transition point between the predominantly symmetrical to predominantly asymmetric fission is expected to be sensitive to the excitation energy of the compound nucleus, as is mentioned earlier.

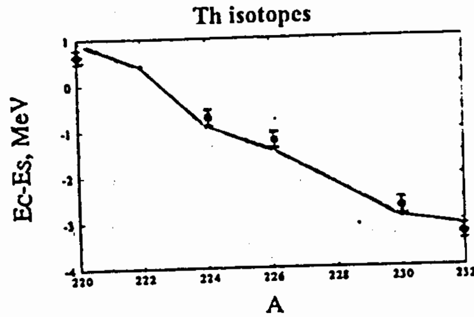

Figure 8.

Conclusion

The experimental and theoretical research of the characteristics of low-energy fission of the neutron deficient isotopes ^{220}Th, ^{224}Th, ^{226}Th, the results of which are given in the present paper, was the first successful attempt to employ subbarrier fusion reactions of heavy ions and nuclei for this purpose.

The analysis of the experimental data obtained by these investigations allowed one to reveal essential characteristics of the mass and energy distributions of fragments in relation to the excitation energy and nucleonic composition of fissioning nuclei. It was established that each fission mode (the symmetric mode *s* and the two asymmetric modes, a_0 and a_1) has its own definite set of characteristics: Y_i, TKE_i, σ^i_{TKE}, E^i_f. These results give a sufficiently rigorous physical proof of fission modes being independent and genetically related to the corresponding valleys on the potential energy deformation surface of the nuclei under study.

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