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## THEORETICAL EVALUATION OF NEUTRON AND PROTON INDUCED FISSION CROSS-SECTIONS FOR Pb-Pu TARGETS IN ENERGY RANGE 20-200 MeV

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**Abstract:** . The new MultiConfiguration Fission (MCF) approach is used for calculations of nucleon induced fission cross sections at 20-200 MeV. The cross sections are obtained as a sum of contributions from decay of  $(Z,A,p,h,U)$  configurations, formed after fast stage of fission reaction with weights defined by the population probability  $Y$  of  $(Z,A)$  nucleus in the  $(p,h)$  particle-hole state at the excitation energy  $U$ . The intranuclear cascade model was used to compute the probability  $Y$  and the pre-compound statistical model, - for calculation of fission and de-excitation cross section from state  $(Z,A,p,h,U)$ . Up to 15 fissioning nuclei in 10  $(p,h)$  states at the wide excitation region were taken into account in the calculations.

9 proton induced and 12 neutron induced fission cross sections for nuclei from Pb-204 to Pu-239 have been evaluated in the incident particle energy range between 20 and 200 MeV.

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96/11

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a) *citing the evaluation*

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b) *citing the format*

V. McLane, P.F. Rose, C.L. Dunford (ed.), "Data formats and procedures for the Evaluated Nuclear Data File ENDF-6", report BNL-NCS-44945 (ENDF-102) Rev 2/97 (Brookhaven National Laboratory 1997).

# THEORETICAL EVALUATION OF NEUTRON AND PROTON INDUCED FISSION CROSS-SECTIONS FOR Pb-Pu TARGETS IN ENERGY RANGE 20-200 MeV

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The new code *MCFx* (*M*ulti-*C*onfiguration *F*ission *x*-section) has been recently developed in Khlopin Radium Institute (St.-Petersburg, Russia) under financial support of ISTC (Moscow). The main feature of the approach developed is the step-by-step simulation of all stages of the nucleon-induced nuclear reactions with the heavy target nuclei. The code system includes three main blocks:

- i) first of all, with the coupled channel method the reaction cross-section (probability for nucleon to penetrate in a nuclear volume) is calculated;
- ii) further, using models of the intranuclear cascade and multiple preequilibrium emission the fast stage of reaction is modelled;
- iii) the calculated distributions are used as the entrance data in detailed calculations of particle evaporation from the excited nucleus and multichance fission as it is done in the well-known code STAPRE.

The control calculations have shown that results obtained are defined mainly to such model parameters as fission barriers values, nuclear masses and level density parameters. For these key parameters the separate development were carried out allowing to compute these values at a modern level. It also can be considered as a feature of our approach.

Modelling of a fast stage of reaction has shown, in particular, as far as the structure of experimentally observed neutron or proton induced fission cross-section of given target nucleus energy is complex. The contributions in the experimentally measured fission cross-sections bring a wide spectrum of fissile nuclei with various excitation energies and quantum configurations and weights of these contributions are very different, too. The name of developed program complex **MCFX** emphasizes the importance of this interesting but strongly complicated in practical calculations phenomena (Multi-Configuration Fission cross sections structure).

We used the coupled channel method as it has been done in the code ECIS for the description of the entrance reaction channel. The code ECIS has been modified and the new optical model potential for both neutrons and protons in the whole energy under consideration were developed for the calculations of reaction cross-sections. The comparison of our results for total and reaction cross-sections with the available experimental data shown the rather well agreement of data for all cases. This circumstance allowed us to form the data library on the reaction cross-sections which was further used in the fission cross-section calculations.

At preequilibrium and equilibrium reaction stages the main questions were the proper choice and detailed description of the mutiparticle emission of preequilibrium particles on the base of the exciton model and physically correct model for level density which is one of the main factors defining the competition between particle emission and fission.

Other important components of calculations were fission barriers and nuclear masses. For calculation of experimentally unknown fission barriers the parameterization based on the method of shell corrections has used. Comparison with tables of fission barriers of other authors based on the analysis of experimental data has allowed to fix the parameters of calculations necessary for calculation of fission barriers for nuclei far from of stability valley. For estimation of nuclear masses the modified method of difference Garvi-Kelson relations was applied where the main advantage is

predicted accuracy of an estimation and an opportunity of smooth transition from the experimental and theoretical values for nuclear masses.

The results obtained in the framework of the MCFx code are in a rather good agreement with the experimental data practically without parameter variations.

The data libraries on fission cross-sections for nuclei from Pb to Pu by neutrons and protons of 20-200 MeV energies were formed. The libraries are presented as ENDF-6 format nuclear data files and are available in the electronic form by request.

Main results of the project performance are listed below as follows:

- the modern model code for detailed calculations of main stages of nucleon-induced reactions with heavy target nuclei at 20-200 MeV energy range has been developed;
- the libraries of key model parameters used at calculations are developed that allows to apply the code in the closed kind without attraction of the additional data;
- a large volume of control calculations has been carried out and comparison with known experimental data on fission cross-sections in Pb-Bi area and actinides were performed. Results of comparison show on high enough predictive force of the approach that allows to use a code for generation of the nuclear data;
- the nuclear data files on fission cross-sections in the ENDF-6 format for proton-induced fission of  $^{206,207,208}\text{Pb}$ ,  $^{209}\text{Bi}$ ,  $^{232}\text{Th}$ ,  $^{235,238}\text{U}$ ,  $^{237}\text{Np}$  and  $^{239}\text{Pu}$  were formed;
- the nuclear data files on fission cross-sections in the ENDF-6 format for neutron-induced fission of  $^{204,206,207,208}\text{Pb}$ ,  $^{209}\text{Bi}$ ,  $^{232}\text{Th}$ ,  $^{233,234,235,236,238}\text{U}$ ,  $^{237}\text{Np}$  and  $^{239}\text{Pu}$  were formed;

Detailed description of the approach is given in the following publications:

1. S. Yavshits, G. Boykov, V. Ippolitov, S. Pakhomov, A. Roschin, O. Grudzevich "Multiconfiguration Fission Cross-Sections at Transitional Energy Region 20-200 MeV", *Voprosy Atomnoj Nauki i Tekhniki, seriya Yadernye Konstanty, vypusk 2000-1* (2000) pp. 62 – 70.  
Published also as Report INDC(CCP)-430 (2001) pp. 83 – 94 (see Appendix 1 below)
2. S. Yavshits, S. Pakhomov, O. Gridzevich "Semimicroscopic Treatment of Nuclear Fission Barriers", *Voprosy Atomnoj Nauki i Tekhniki, seriya Yadernye Konstanty, vypusk 2000-1* (2000) pp. 55 – 61.  
Published also as Report INDC(CCP)-430 (2001) pp. 73 – 81 (see Appendix 2 below)
3. Paper presented at the VIII International Seminar on Interaction of Neutrons with Nuclei, Dubna, May 17-20, 2000 (Abstracts) p.87.
4. Proc. of the 9 th International Conference on Nuclear Reaction Mechanisms, ed. by E. Gadioli, Varenna, June 5-9, 2000, pp. 219-226.
5. S. Yavshits et al., in Proc. of the 2000 Symp. on Nuclear Data, eds. N.Yamano, T.Fukahori, JAERI, Tokai, Japan, Nov. 16-17, 2000, pp.277-282.
6. Paper presented at the International Conference on Nuclear Data for Science and Technology, Tsukuba, Japan, Oct. 7-12, 2001, (Abstracts) p.1.2-O-2.

**Appendix 1.** Paper from Voprosy Atomnoj Nauki i Tekhniki, seriya Yadernye Konstanty, vypusk 2000-1 (2000) pp. 62 – 70. Published also in the Report INDC(CCP)-430 (2001) pp. 83 – 94.

## MULTICONFIGURATION FISSION CROSS-SECTIONS AT TRANSITIONAL ENERGY REGION 20-200 MeV

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MULTICONFIGURATION FISSION CROSS-SECTIONS AT TRANSITIONAL ENERGY REGION 20-200 MeV. The new approach to the calculation of nucleon induced fission cross sections at 20-200 MeV is presented. The cross sections of multiconfiguration fission (MCF) is calculated as a mixture of  $(Z,A,p,h,U)$  configurations formed as a result of fast stage of fission reaction with weights defined by the population probability  $Y$  of  $(Z,A)$  nucleus in the  $(p,h)$  particle-hole state at the excitation energy  $U$ . We use the intranuclear cascade model to compute the probability  $Y$  and the precompound-statistical model for calculation of fission and de-excitation cross section from state  $(Z,A,p,h,U)$ . Calculated results are presented for two absorption cross sections obtained with two optical model parameter sets. Up to 15 fissioning nuclei in 10  $(p,h)$  states at the wide excitation region were taken into account in the calculation.

### Introduction

The development of the future nuclear power energetics with application of hybrid technologies using accelerator-based incineration systems requires a knowledge of data library on cross-sections of nuclear reactions in a wide energy range of incident particles (from very slow particles up to beams with energy of a few GeV). The systematic experimental research for such wide energy range is complicated and for many targets in principle impossible. Therefore a key role for the development of the required nuclear data library has a model calculations which should have a sufficient predicting force for the description of fission cross-sections and cross-sections of other competing nuclear reactions with necessary accuracy. At the present time there are a lot of experimental data and rather reliable codes for the calculation of fission cross-sections at incident particle energies up to 20 MeV. At energies of particles about several hundreds MeV (from 200 MeV and higher), rather reliable evaluations can be obtained by means of codes based on the most modern versions of an intranuclear cascade model. At the same time, in an energy range from 20 up to 200 MeV there is a lack of experimental data which are frequently not agreed among themselves and the agreement of results of the best modern model codes is not better than within a factor of 2 or more. Besides, in this energy range, the significant variation of cross section values is observed for both beam energy growth and a transition from nucleus to nucleus.

The energy region of 20-200 MeV is a transitive one from well investigated low-energy nuclear physics to the physics of intermediate energy. It is possible to divide the existing model codes used for calculation of the reaction cross-sections in this transition region into two basic classes:

- Codes for the description of low energy reactions (up to several MeV) using the statistical model based on the Hauser-Feshbach theory and the pre-equilibrium emission model with total angular momentum conservation (codes STAPRE, GNASH and others);

- Codes for the calculation of cross-sections at intermediate energies from several hundreds MeV up to tens of GeV using the various versions of the intranuclear cascade model (cascade-emission model of Ilyanov-Mebel, cascade-exiton model of Toneev-Mashnik and others).

Unfortunately in the transition energy region where the contributions of pre-equilibrium and direct emission, various fission chances, structures of fission barriers and low-energy states are essential, the predicting ability of these model approaches is rather low due to a wide and not always reasonable variation of model parameters for the description of experimental data. The basic problems which need to be solved for this energy region are the description of the entrance channel, the correctness of non-equilibrium process calculations and the systematic of fission barriers including their energy dependence.

The example of fission cross-section description with both, the pre-equilibrium-statistical model (STAPRE code) and the intranuclear cascade-exciton model (code CEM95) with default parameters (i.e. without input parameters fitting) are shown in Figs 1 and 2 for  $^{232}\text{Th}(n,f)$  and  $^{239}\text{Pu}(n,f)$  reactions in comparison with experimental data. The crucial discrepancy between the results of two model calculations and between the calculated and experimental data is clearly observed.

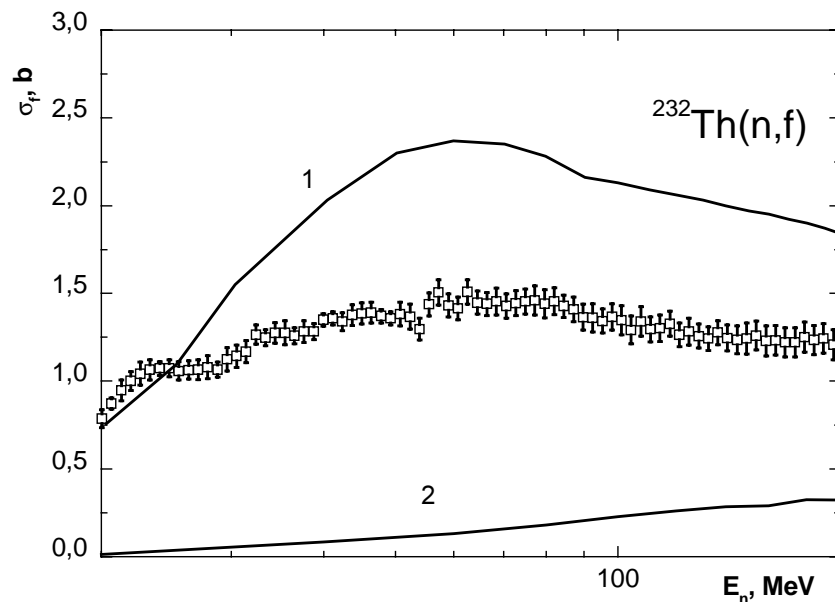


Fig. 1. Comparison of the default versions of CEM95 and STAPRE codes for  $^{232}\text{Th}(n,f)$  reaction. The points are the experimental data. Curve 1 is the STAPRE result, curve 2 is the CEM95 result. See text for details.

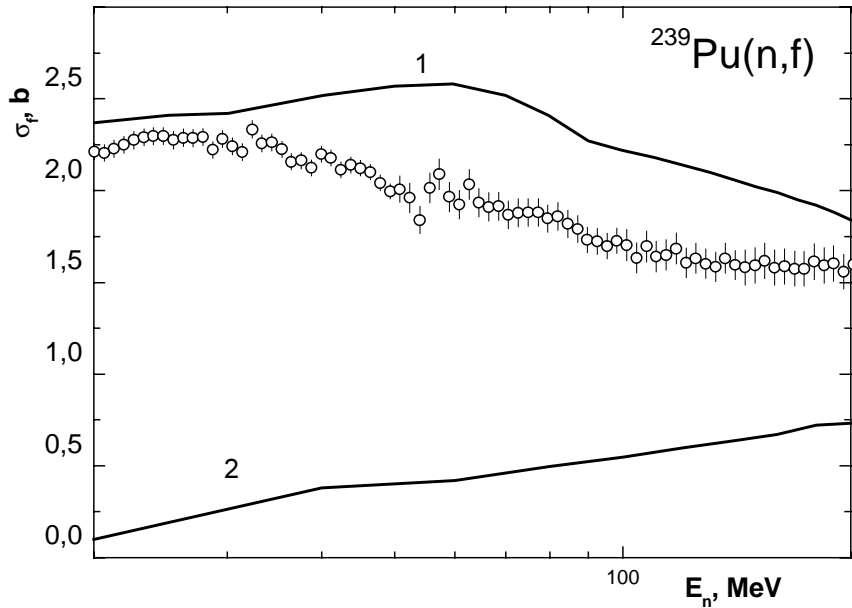


Fig. 2. The same as in Fig. 1, but for  $^{239}\text{Pu}(n,f)$  reaction.

### 1. Optical model calculation of reaction cross-sections for deformed targets

The realistic description of the entrance channel (i.e. calculation of the probability for nucleon to penetrate inside nucleus) is the significant part of any nucleon-induced fission cross-section calculations. The reaction cross-section calculation (absorption) for incident particles at all energies is defined under consideration by nucleon transmission coefficients. Moreover, transmission coefficients define decay widths of intermediate nuclear states at pre-equilibrium and evaporation stage of disintegration of excited nucleus. This is a reason why the reliability of the calculation of the transmission coefficient effects on an accuracy of fission cross-section calculations.

At present the main (and unique) method for this kind of calculations is the optical model of nuclear reactions. For spherical nuclei the calculations can be done with the well-known code SCAT2 [1]. However, as a rule, fissioning nuclei are strongly deformed in their ground states and also have a number of collective low energy excited states. The connection with these states modifies the wave function of the nuclear system rather significantly. The consideration of this modification requires the application of the generalized optical model which has been formulated in the coupled channel method [2] and developed in the works of T. Tamura et al. [3]. On the basis of the Tamura approach, the code ECIS was developed by J. Raynal [4] which allows to calculate the cross-sections of nuclear reactions in the wide energy region [5].

The rotational model level schemes presented in the RIPL library [8] and relativistic kinematics are used in the calculations. The coupling of only three channels are taken into account because the account of more numbers of channels modifies results less than 1%.



On the base of this kind of calculations, the library of proton- and neutron-induced total and reaction cross-sections for 35 nuclei ( $^{208}\text{Pb}$ ,  $^{209}\text{Bi}$ ,  $^{227}\text{Ac}$ ,  $^{228,229,230,232}\text{Th}$ ,  $^{232,233,234,235,236,238,239}\text{U}$ ,  $^{237,238,239}\text{Np}$ ,  $^{238,239,240,242}\text{Pu}$ ,  $^{242,243}\text{Am}$ ,  $^{243,244,245,246,247,248}\text{Cm}$ ,  $^{247}\text{Bk}$ ,  $^{248,249,250,251,252}\text{Cf}$ ) is developed for energies up to 220 MeV.

In Fig. 3 the results of total cross-section calculations are presented for  $^{235}\text{U} + n$  reaction in the case of two optical model parameter sets (OMPS) – by Young [6] and by Konshin [7] in comparison with experimental data (the Young potential has a restricted energy interval of application, so results of calculations are shown up to 100 MeV). In both cases, our results describe the experimental data quite well. But, as it can be seen in Fig. 4, the reaction cross-sections are more sensitive to the choice of the potential: absorption cross-sections of neutron by  $^{238}\text{U}$  are very different (up to 15% around 30 MeV). Due to the lack of the experimental data on reaction cross-sections, the choice between OMPS is difficult and we carried out the calculations for both cases. For the fission cross-section calculation, we used OMPS [7] where the picture of the gross structure in the reaction cross-section is reproduced more reliably in the whole energy region under consideration.

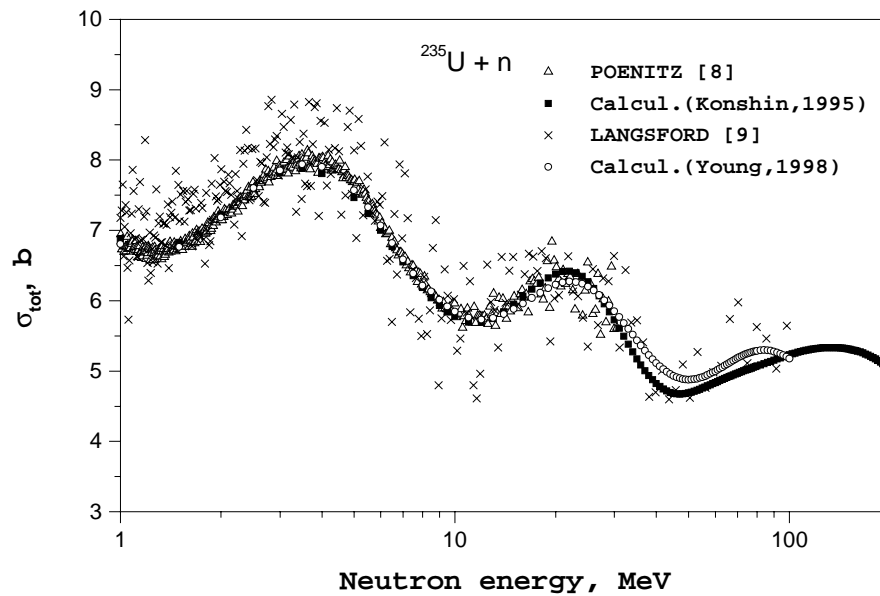


Fig. 3. Total neutron cross section for  $^{235}\text{U}+n$ . Experimental data [8,9], curves are results of OM calculations with OMPS [6] and [7].

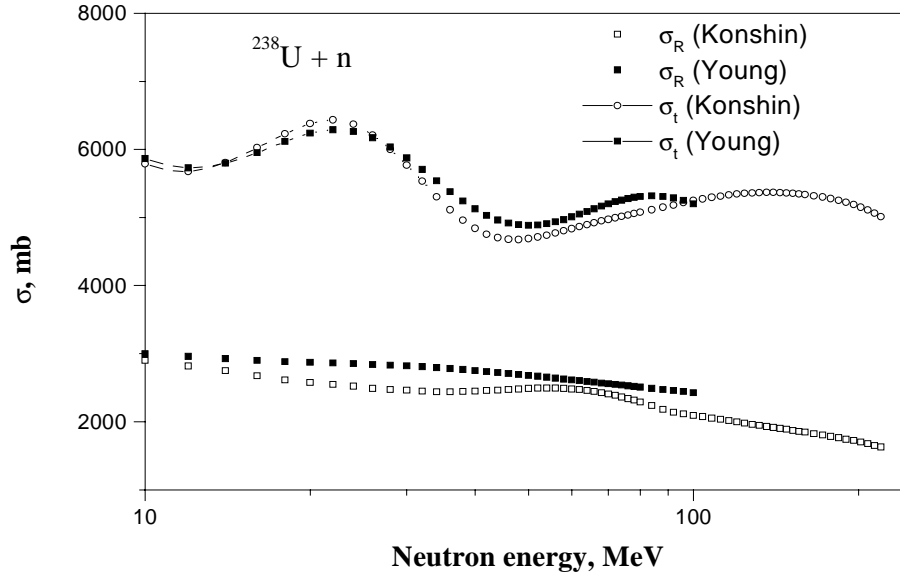


Fig. 4. Calculated cross-sections for  $^{238}\text{U}+n$ :  $\sigma_R$  is the reaction cross-section,  $\sigma_{\text{tot}}$  is the total cross-section.

## 2. Simulation of entrance channel of fission reaction with intranuclear cascade model

We use the intranuclear cascade model for the description of the direct processes on the first stage of fission reaction as it is realized in the CEM code [10] where the so-called “cascade-exciton” model of nuclear reactions has been formulated.

It is assumed in this model that the nuclear reaction goes on three main stages. The first one is the intranuclear cascade where primary particle may undergo a number of re-scattering before its absorption or escape from nuclear volume occurs. The significant point of this scenario is a formation after the cascade stage of excited residual nuclei in different particle-hole configurations which serve as a start for the second pre-equilibrium reaction stage which can be treated in the framework of the standard exciton model. And the third stage is the particle evaporation and/or fission from the equilibrium state of the final nuclear system. The important point of the CEM model is the substantial difference of particle-hole configurations (characterized by exciton number  $n=p+h$ , where  $p$  and  $h$  are the numbers of particles and holes, correspondingly) as well as excitation energy after completion of the first direct reaction stage from the starting configurations of ordinary exciton models. Numerous calculations show that the distributions of residual nuclei after the cascade stage are rather wide on  $n$ ,  $p$ ,  $h$  and energy.

All calculations of intranuclear cascades are performed in the three-dimensional geometry. The nuclear density distribution is described by the Fermi distribution with parameters taken from the experimental data on electron-nucleus scattering. The target nucleus is divided by concentric spheres at 7 zones where the nuclear density is assumed to be a constant. The diffusiveness of nuclear density and potential edge is taken into account. For intranuclear collisions of nucleons, the Pauli principle forbids the collision with energy of secondary particles less than Fermi energy.

The main condition of the intranuclear cascade model applicability is the smallness of the length of de Broglie waves for all interacted particles; the wave length must be less than the average distance between intranuclear nucleons  $\sim 1$  fm. In this case, the picture of the interaction is approximately the semiclassical one and it is possible to say about the particle trajectories and two-particle collisions inside nucleus. This condition restricts the energy region of incident particles above 50 MeV. Such a limitation is a significant point of the model. However, in order to improve it, the way out of the frameworks of intranuclear cascade is necessary with the description of

reaction mechanism in the terms of rather strict quantum-statistical approach. Practically the low limit of applicability of intranuclear cascade can be established in the analysis of calculation results for the given case.

The results of our calculations for main characteristics of residual nuclei formed after the cascade stage are presented in Figs 5-8 for  $^{232}\text{Th}$  in the transitive energy region 20-200 MeV. The yields of residual nuclei are presented in Figs 5-6 for protons and neutrons in the entrance channel. The presented results show that at low energies almost all incident particles are absorbed by target nucleus with formation of compound nucleus. For higher energies the yield of compound nucleus falls down significantly. The maximal yield in this case corresponds to the escape of one neutron or proton from target nucleus.

The yields in the direct reactions  $^{232}\text{Th}(n,2n)^{231}\text{Th}$  and  $^{232}\text{Th}(n,n')^{232}\text{Th}$  with formation of  $^{231}\text{Th}$  and  $^{232}\text{Th}$  in two particle-hole configurations are shown in Figs 7-9 as a function of excitation energy for the given residual nucleus. As it can be seen, the configurations with the low exciton number ( $p+h$ ) have significantly higher yields as compared with the configurations characterized by large exciton numbers. Nevertheless, these configurations have a similar contribution to the fission cross-sections due to high excitation energies of high-exciton configurations (Fig. 5).

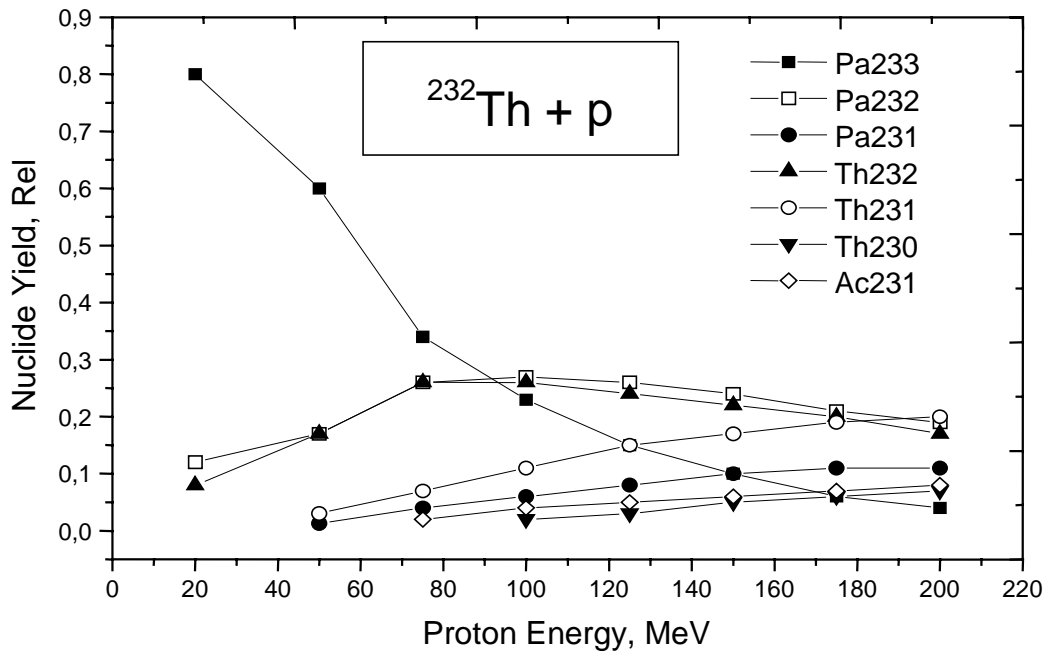


Fig. 5. The yield of residual nuclei after the cascade stage for  $^{232}\text{Th}+p$  reaction.

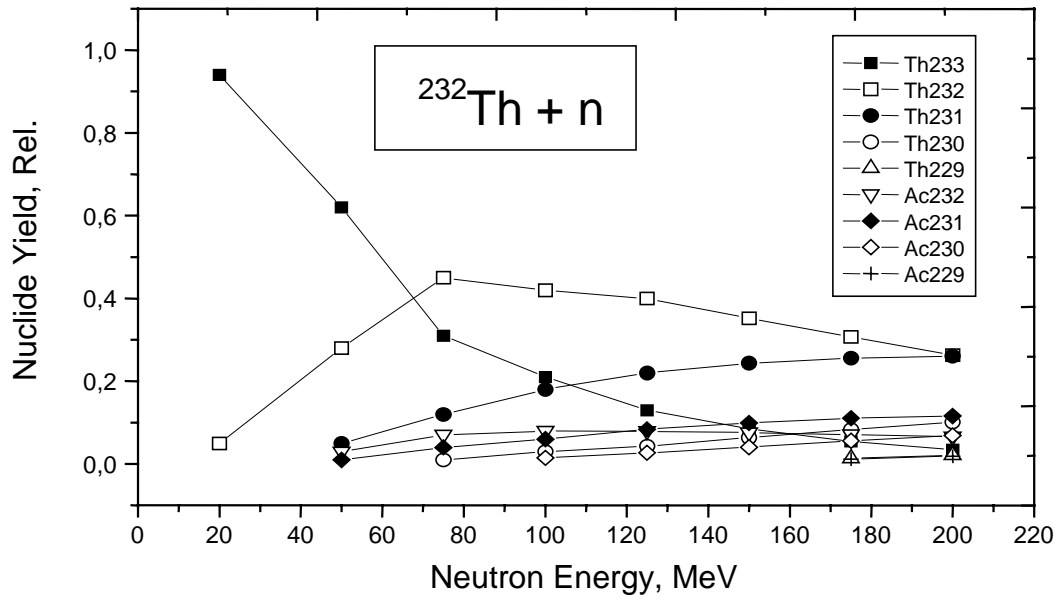


Fig. 6. The same as in Fig. 5 but for  $^{232}\text{Th}+n$  reaction.

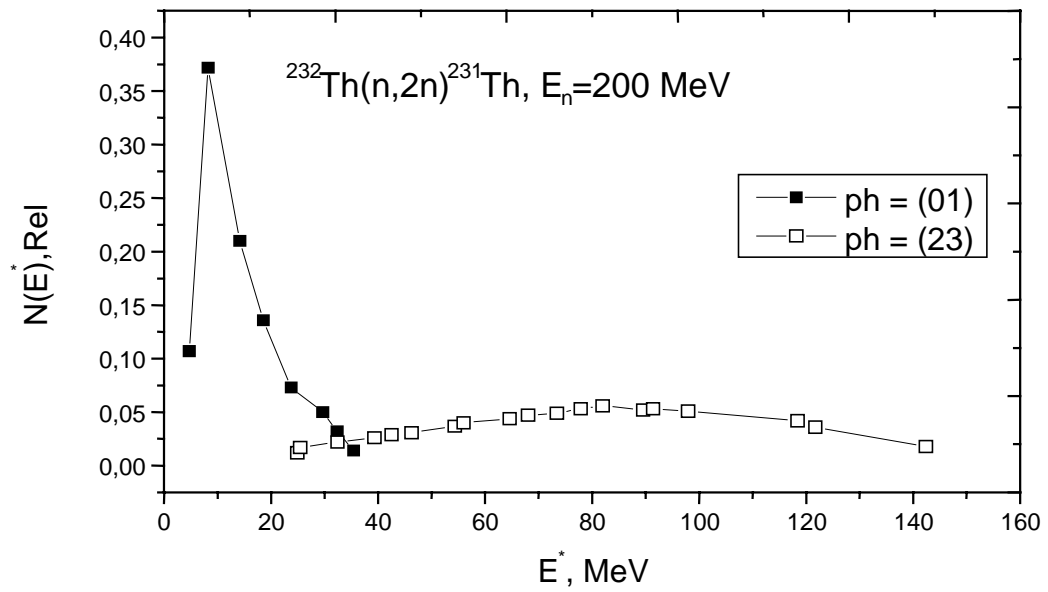


Fig. 7. The configuration yields in the direct reaction  $^{232}\text{Th}(n,2n)$  as a function excitation energy.

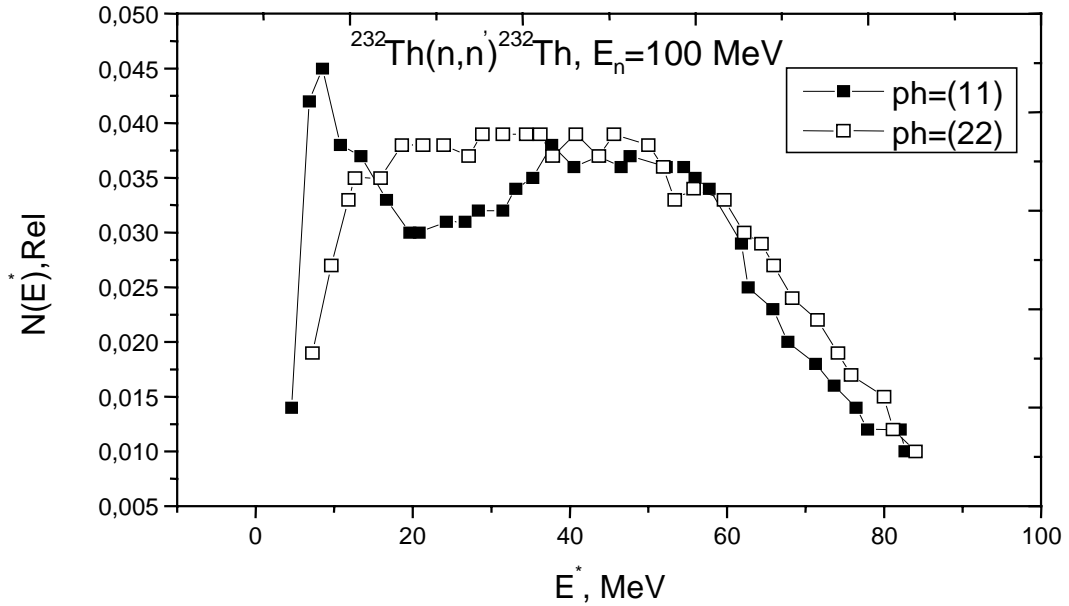


Fig. 8. The same as in Fig. 7 but for the direct reaction  $^{232}\text{Th}(n,n')$ .

The modification of the CEM95 code has been necessary for these kind of calculations. The block of intranuclear cascade has been separated from the code and the new module has been written down for the data acquisition in the output file. The necessary accuracy is achieved when 500 000 histories are taken into account. The configurations with yields more than 1% are only taken into consideration.

This kind of results serves as an input data for the calculations of pre-equilibrium and equilibrium decay of excited nuclei (code STAPRE). For each configuration, it is necessary to carry out the calculations of particle emission and fission cross-sections which further are summed with corresponding weights.

### 3. Calculation of multiconfiguration fission cross sections

The STAPRE code [11] was developed for the calculation of cross sections of reactions caused by particles with a competition of several particles,  $\gamma$ -quanta and fission in the assumption of sequential evaporation.

Each step of evaporation is described by the statistical model of nuclear reactions in a Hauser-Feshbach-Moldauer formalism [12,13] which take into account the conservation laws of the total angular momentum and parity. For the first particle the opportunity of pre-equilibrium decay is taken into account.

The code provides that in entrance and in exit channels of reaction, there can be neutrons, protons, alpha-particles and deuterons. There is an opportunity to set any particle or nucleus in the entrance channel. The way of definition of nuclei which fission is in question is thus realized.

During the 25 years of the code usage, a lot of changes and improvements were included. Some special efforts were done for calculations of fission cross sections in the 20-200 MeV region. For the calculation of the particles emission and fission with particle energies up to 200 MeV, the adaptation of the block of the statistical model of the STAPRE code was executed:

- Additional (up to 30) cascades of the particle emission to cover the whole energy range are included. The appropriate files are extended;
- The energy bin of integration is decreased from 2.5 up to 0.5 MeV to get necessary accuracy of calculations for the near threshold energy. In the current version, the whole energy region is divided into 380 bins;
- The doubled precision (REAL\*8) for the maintenance of the protection from machine overflows is added.

The usage of the statistical model of nuclear reactions in the STAPRE code leads to the necessity to provide the large number of the input data: level density of the excited nuclei, characteristic of discrete levels (energy, spin and parity), fission barriers, particle binding energies, etc. It should be stressed that the large work on the preparation of the input data for calculations of nuclear reaction cross sections with theoretical models is done by the experts under the IAEA coordinated research program [14]. Unfortunately, in a part of level density the attention is only given to nuclei close to the stability valley, and there are mistakes in files of discrete levels.

The following work has been done in order to satisfy the input data needs for the calculations of fission cross sections for nuclei far from a beta-stability valley:

- i) compilation of the schemes of discrete levels of nuclei with  $A=1-260$ ,
- ii) the analysis of the schemes for the level missing,
- iii) the analysis of the spins and parities of levels.

The information on the excited levels of nuclei is logically divided into two parts: discrete levels described by energy, spin and parity; and continuum, where level density for the given excitation energy is used. For the STAPRE code a LDP file containing the level density parameters within the framework of the generalized model of a superfluid nucleus and SEDL file containing the characteristics of discrete levels, 2821 nuclei were formed.

The preparation of the initial data is organized according to an output file of the intranuclear cascade code, which contains the yields  $Y(Z,A,p,h,U)$  of nucleus  $(Z,A)$  in the certain particle-hole configurations  $p-h$  distributed on the excitation energies  $U$ . Hence, all nuclei, all configurations and all excitation energies have to be included in the entrance channel for the STAPRE code. Additional feature of the initial files formation is needed to take into account all steps of the particle emission for the fission cross section calculation (the chance structure of cross sections).

The PRESTAPR code is created and tested. It uses the data files as follows: LDP - parameters of level densities, SEDL - discrete levels characteristics, BARRIER - doubled humped fission barriers, GRD - parameters giant dipole resonances, Bn\_MOL - energy of separation for the neutrons, protons and alpha-particles.

The PRESTAPR code prepares initial files for the calculations of the fission cross-sections by the STAPRE code using the list of nuclei, their particle-hole configurations and excitation energies (Z,A,p,h,U) calculated by the intranuclear cascade code and using the prepared LDP, SEDL, GDR, BARRIER, Bn\_MOL files of initial parameters.

The result of the STAPRE code is the file of fission cross sections for the nuclei at the given configuration with the given excitation energies for the n-th chance, i.e.:

$$\sigma_f^n(Z, A, p, h, U).$$

The processing of this file consists in calculation of fission cross sections as a multiconfigurational mixture, that is as a sum on all residual nuclei (Z,A,p,h,U) with weight Y(Z,A,p,h,U):

$$\sigma_f(E) = \sum_n \sum_{Z,A,p,h} \int \sigma_f^n(Z, A, p, h, U) \cdot Y(Z, A, p, h, U) dU$$

The procedure of the summation according this formulae is realized in the service program PROSTAPR.

#### 4. The results of calculations

The preliminary calculations were performed for the case of  $^{232}\text{Th}$  and  $^{239}\text{Pu}$  neutron-induced fission in the energy region 20-200 MeV. The analysis of the role of different residual nuclei configurations in the fission cross-section formation were carried out. Up to 11 fission chances have been taken into account.

The example of this kind of analysis is shown in Fig. 9 where fission cross-sections for different residual nuclei formed in the reaction  $^{232}\text{Th} + n$  are presented in the units of absorption cross-section. The results show that if for low energy fission the main part of total fission cross-section is due to fission of the compound nucleus, then for higher energies the role of first residual nuclei from (n,xn) reactions increases. Such a result is directly connected with the formation probability and excitation energy distribution of residual nuclei which have been obtained in the intranuclear cascade calculation.

The total fission cross-section is presented in Fig. 10 and Fig. 11. The cross-sections have been calculated for two variants of absorption cross-section calculations, that is for Konshin and Young potentials. It is seen that even these preliminary results are able to reproduce the observed fission cross-section behavior. The Konshin potential for the absorption cross-section gives a better description of the experiment.

The results obtained are only preliminary ones because the performance of detailed calculations in a wide region of nuclei requires more accurate values of main model parameters such as optical potentials, fission barriers and their dependence on excitation energy, level density at the saddle point and so on. The additional analysis of the role of different configurations and fission chances is necessary, too, as well as the optimization of main code routines in order to reduce computer time especially for the calculations of high energy fission.

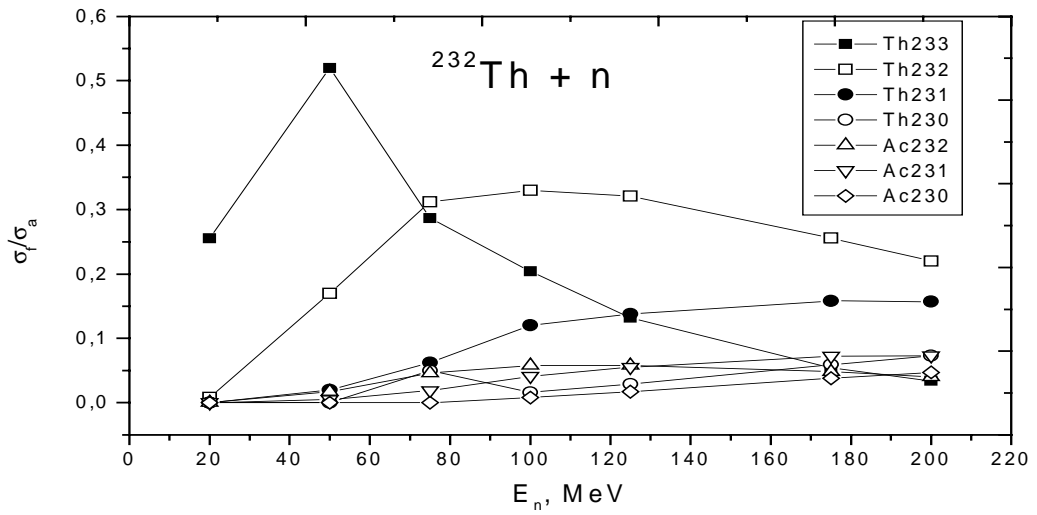


Fig. 9. Relative fission cross-sections of residual nuclei in reaction  $^{232}\text{Th}+n$ .

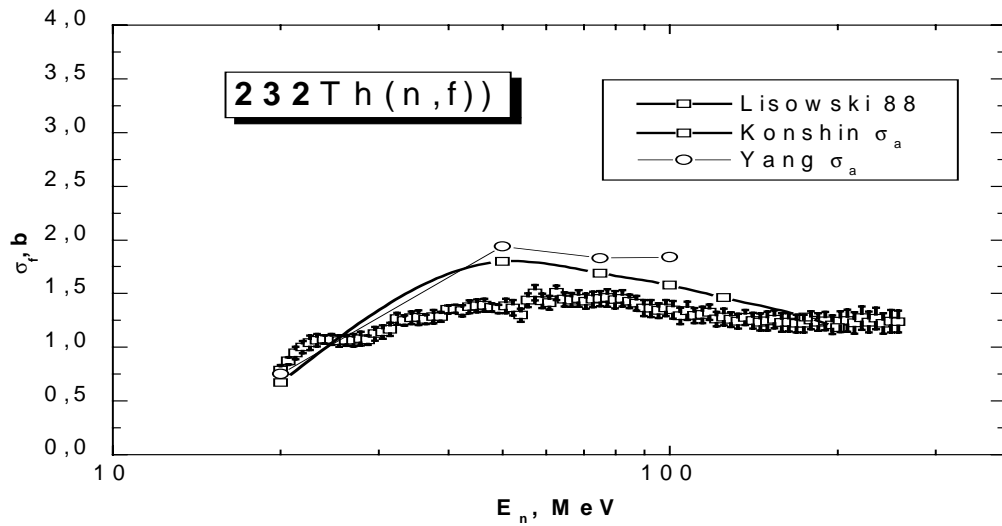


Fig. 10. Total fission cross-section for two optical potentials in comparison with the experimental data.



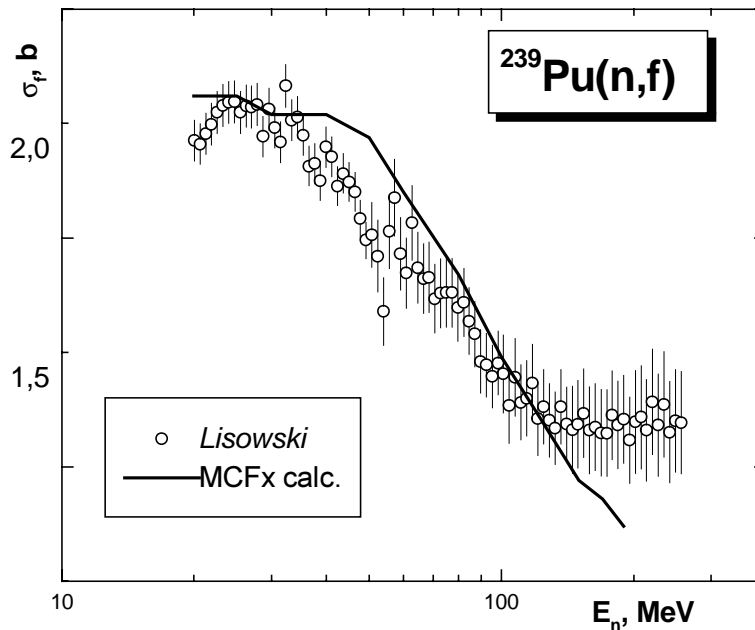


Fig. 11. The same but for  $^{239}\text{Pu}+n$ .

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## SEMIMICROSCOPIC TREATMENT OF NUCLEAR FISSION BARRIERS

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SEMIMICROSCOPIC TREATMENT OF NUCLEAR FISSION BARRIERS. The model is proposed to calculate the fission barriers of heavy nuclei with account of their possible dependence on excitation energy. The model is based on the shell correction method for calculation of potential energy of deformation with nuclear shape parameterization in the lemniscate coordinates.

The development of reliable fission barrier systematics is one of the main points in the development of fission cross-section systematics itself especially in the fields where calculated results cannot be compared with experiments directly. Besides, the well-known semiempirical systematics of fission barriers [1] includes only range of traditional nuclei at low excitation energy while the development of new nuclear accelerator-driven systems (ADS) requires to expand the systematics in order to include neutron deficient nuclei with sufficiently high excitations.

The Strutinsky method [2] of shell correction was used as a base for the new systematics. The necessary single-particle level schemes were calculated with the DIANA code [3]. The calculation of smooth “liquid drop” part of deformation energy is based on the improved Yukawa potential (Yukawa-plus-exponential model [4]).

We used Cassini ovaloids as a basis for the description of nuclear shape defined in the lemniscate coordinates [5]. The deviations of nuclear shapes from the basic figures which define high deformation modes are described by coefficients at corresponding Legendre polynomials. So, the configuration of the fissioning system is defined by the following parameter set:

$$\{\alpha_f\} = (A, Z, \varepsilon, [\alpha_m], T), \quad (1)$$

where  $A$ ,  $Z$  are mass and charge numbers,  $\varepsilon$  is the deformation of the basic figure,  $[\alpha_m]$  are coefficients at polynomials and  $T$  is the temperature of the fissioning nucleus.

The transformation of the  $(R, x)$  point in the lemniscate coordinate system to the cylindrical coordinates is as follows:

$$\begin{aligned} r &= 2^{-1/2} [R^4 + 2 S R^2 (2x^2 - 1) + S^2]^{1/2} - R^2(2x^2 - 1) - S]^{1/2}, \\ z &= 2^{-1/2} \text{sign}(x) [R^4 + 2 S R^2 (2x^2 - 1) + S^2]^{1/2} + R^2(2x^2 - 1) + S]^{1/2}, \end{aligned} \quad (2)$$

$$S = \varepsilon R_0^2$$

where  $S$  and  $R$  are

$$R(x) = R_0 \left( 1 + \sum_{m=1}^N \alpha_m P_m(x) \right) \quad (3)$$

and  $P_m$  are Legendre polynomials.

In this model the parameter  $\varepsilon$  can be considered as a fission coordinate. At  $\varepsilon=0$  (and  $[\alpha_m]=0$ ) the nucleus has the spherical shape. At  $0<\varepsilon<0.4$  the shape is close to an ellipsoidal one. At  $0.5<\varepsilon<1$  the neck arises and develops and at  $\varepsilon=1$  the neck radius is equal 0, i.e. nucleus is divided in two fragments:

$$S = \varepsilon R_0^2, R_0 = r_0 A^{1/3}. \quad (4)$$

It is commonly used to restrict the set of deformation parameters by degree  $N=4$ . Then the coefficient  $\alpha_1$  defines fragment mass asymmetry,  $\alpha_2$  is correlated with  $\varepsilon$  and can be omitted,  $\alpha_3$  defines octupole deformation and  $\alpha_4$  defines hexadecapole deformations.

The different fission stages for  $^{240}\text{Pu}$  are presented in Fig. 1 vs. parameter  $\varepsilon$  where  $\varepsilon$  varies in the diapason 0-1 with step 0.1 and  $\alpha_1=0.06$ ,  $\alpha_3=0$  and  $\alpha_4=0.016$ . This set of parameters corresponds to the formation of pair fragments  $^{100}\text{Y}$  and  $^{140}\text{Cs}$ .

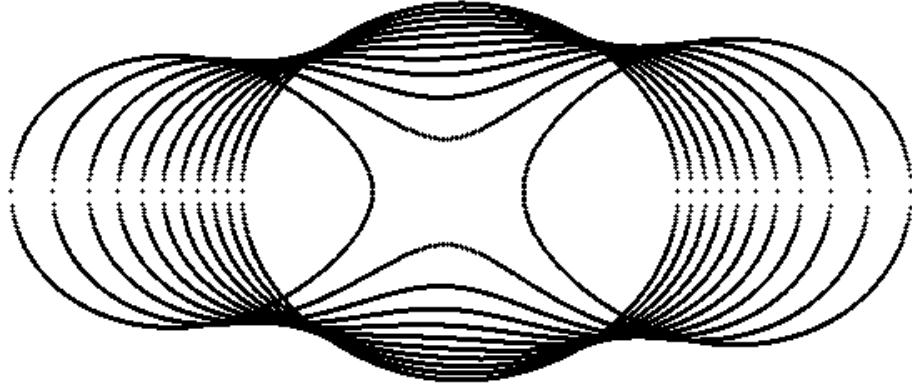


Fig. 1. Different  $^{240}\text{Pu}$  fission stages.

The potential energy of deformation at the ground state in the shell correction method has the following form:

$$E=E_{\text{coul}}+E_{\text{nuc}}+\delta U_n + \delta U_p + E_{p_n} + E_{p_p}, \quad (5)$$

where  $E_{\text{coul}}$  is the coulomb energy of the charged liquid drop,  $E_{\text{nuc}}$  is the nuclear part (surface energy),  $\delta U_n$ ,  $\delta U_p$  and  $E_{p_n}$ ,  $E_{p_p}$  are shell corrections and pairing energies for neutrons and protons correspondingly.

The shell corrections as well as the pairing energies have been calculated on the base of single-particle spectra obtained by means of the DIANA code in the deformed Woods-Saxon well  $V(\{\alpha_f\})$ .

According [4] the value of coulomb energy for axially symmetrical and uniformly charged the liquid drop can be presented by the integral:

$$E_{\text{coul}} = \pi \rho^2 \int_{z_1}^{z_2} dz \int_0^{r(z)} dr \int_{z_1}^{z_2} dz' \{ [r^2(z') - r^2(z) - (z - z')^2 + \frac{dr^2(z')}{dz'}(z - z')] F(a, b) + E(a, b) \}, \quad (6)$$

where  $\rho$  is the charge density,  $F(a,b)$  and  $E(a,b)$  are the complete elliptical integrals of the first and second kinds, correspondingly, and the value of the parameters  $a$  and  $b$  are defined by equations:

$$a(r, z) = \sqrt{(r+r')^2 + (z'-z)^2}, \quad b(r, z) = \sqrt{(r'-r)^2 + (z'-z)^2}, \quad (7)$$

The nuclear part of energy in the Yukawa+exponential model [4] has the following form:

$$E_{nuc} = c_s / (4\pi r_0^2) \int_{z_1}^{z_2} dz \int_{z_1}^{z_2} dz' \int_0^{2\pi} d\varphi \{ 2 - [(\sigma/a)^2 + 2\sigma/a + 2] e^{-\sigma/a} \} r(z) [r(z) - r(z') \cos(\varphi) - r'(z)(z-z')r(z') - r(z) \cos(\varphi) - \frac{dr(z')}{dz'}(z'-z)] / \sigma^4, \quad (8)$$

where  $\sigma = [r^2(z) + r^2(z') - 2r(z)r(z')\cos(\varphi) + z^2 + z'^2 - 2zz']^{1/2}$ ;  
 $c_s$  is the effective constant of surface energy,  $c_s = a_s [1 - k_s (N-Z)^2 / (N+Z)^2]$ ;  
 $a_s$  is the constant of surface energy ( $a_s = 21.7$  MeV);  
 $k_s$  is the constant of isospin asymmetry ( $k_s = 3.0$ );  
 $a$  is the radius of improved Yukawa potential ( $a = 0.65$  fm);  
 $r_0$  is the nuclear radius constant ( $r_0 = 1.18$  fm).

The example of deformation energy calculation is shown in Fig. 2 where the curves of potential energy and its components for fission of  $^{240}\text{Pu}$  are presented as follows: liquid drop part  $E_{lq}$  ( $E_{lq} = E_{coul} + E_{nuc}$ ), shell correction  $E_{sc}$  ( $E_{sc} = \delta U_n + \delta U_p$ ) and pairing energy  $E_p$  ( $E_p = E_{p_n}$  and  $E_{p_p}$ ) as a function of parameter  $\epsilon$ . The heights of fission barriers are shown in the same Figure.

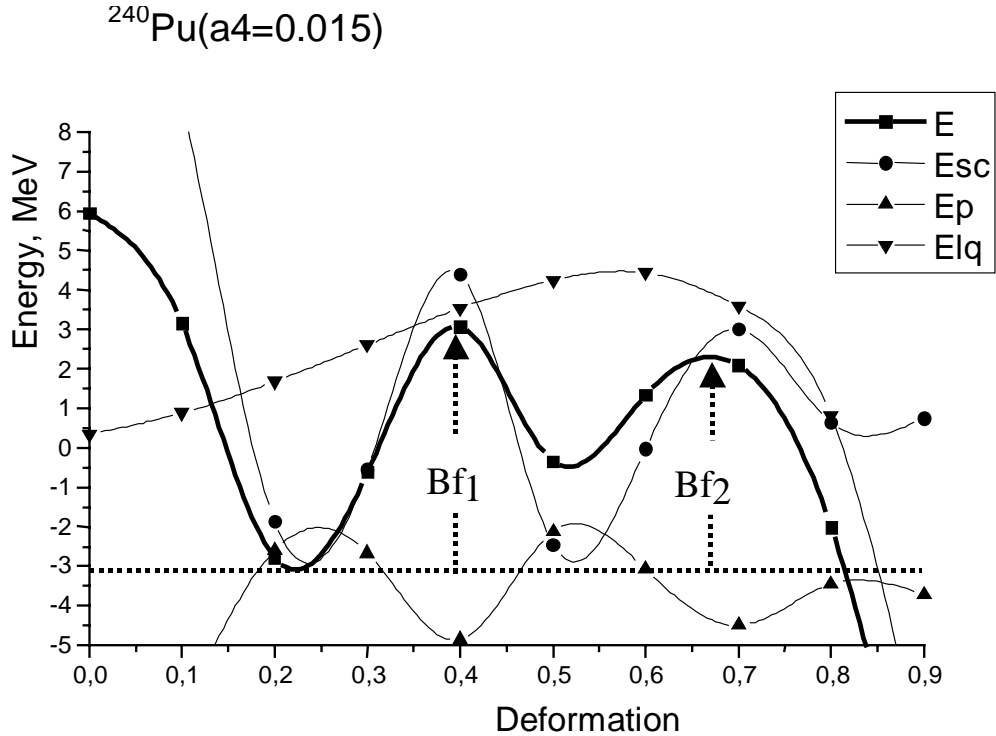


Fig. 2. Deformation energy  $E$  and its components  $E_{lq}$ ,  $E_{sc}$  and  $E_p$  for  $^{240}\text{Pu}$  fission vs. deformation and fission barriers  $Bf_1$  and  $Bf_2$ .

The parabolic approximation near maximums was used for the evaluation of fission barriers  $Bf_1$  and  $Bf_2$  as well as to estimate barriers curvatures as a second derivative of deformation energy  $E$  at the barrier top (C) which is necessary for the calculation of frequencies  $\omega$ :

$$\omega = k \sqrt{C/B} , \quad (9)$$

$$B = 3m/(4\pi) r_0^2 A^{5/3} /2, \quad (10)$$

where  $k$  is the fitting coefficient,  $B$  is the hydrodynamic mass parameter and  $m$  is the nucleon mass.

The potential energy  $E$  at each stage of fission has to be minimal on all deformation parameters  $[\alpha_m]$ . For the definition of the optimal set of these parameters, the effective routine of the multidimensional optimization has been developed which is based on the complex algorithm [6]. However, in practice, there is no possibility to achieve the satisfactory agreement between calculated fission barriers with known semiempirical systematics [1]. The possible reason of this disadvantage may be due to the used shape parameterization which is the main problem in the description of the fission evolution.

Such a circumstance forces to use some parameterization of deformation parameters in the ground state of fissioning nucleus  $\{\alpha_m\}$  by some smooth functions which depend in their turn on the mass and the charge numbers of nucleus or fissility parameter  $Z^2/A$ . The example of barriers behavior as functions of  $\alpha_1$ ,  $\alpha_4$  fixed along fission path is presented in Fig. 3 (A,B) for the case of  $^{240}\text{Pu}$ .

The coefficient  $\alpha_1$  is connected with the formation of the fragment mass distribution. In the actinide region the peak of the mass distribution of the heavy fragments is situated around  $A_H \approx 140$  a.e.m. and the asymmetry parameter can be fixed (from 0.12 up to 0.05 for transition from Th to Cf). It is seen in Fig. 3 that the variation of  $\alpha_4$  coefficient from  $-0.05$  up to  $0.1$  leads to the monotonous increase of the heights for both barriers  $Bf_1$  and  $Bf_2$ .

Other important values which define barriers heights are surface energy parameter  $C_s$  and coulomb radius  $R_0$  (Fig. 3 C,D).

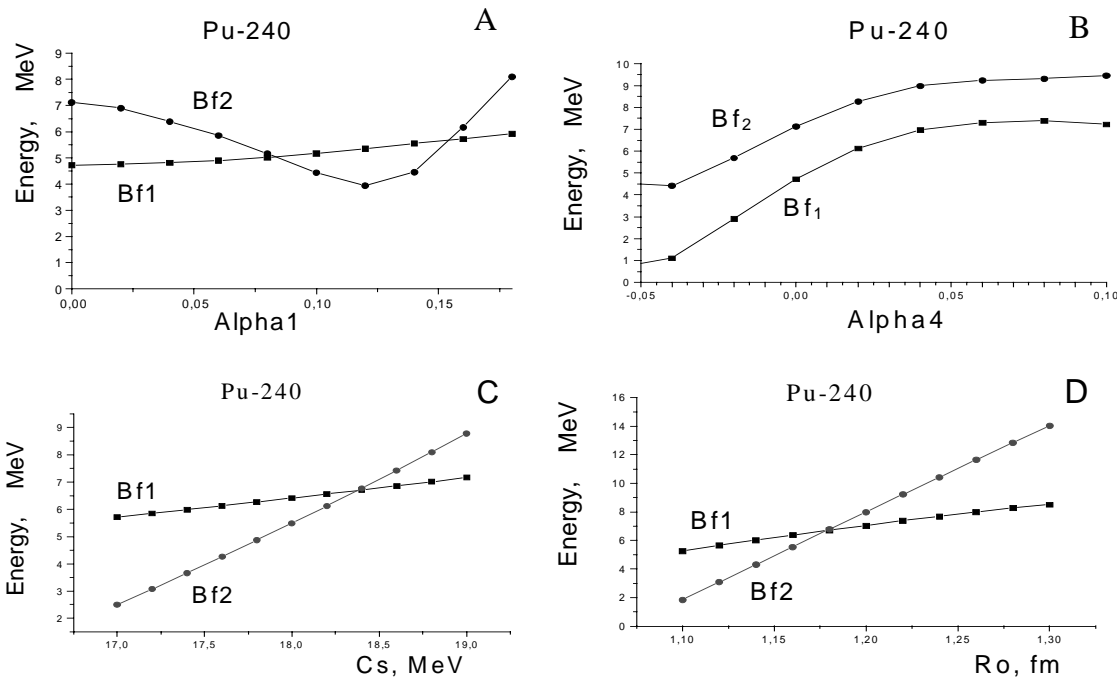


Fig. 3. The fission barriers heights vs. deformation parameters  $\alpha_1$ ,  $\alpha_2$ ,  $\alpha_3$ ,  $\alpha_4$  and parameters  $C_s$ ,  $R_0$ .

We used the parameters  $C_s$  and  $\alpha_4$  as fitting parameters to construct the systematics. In order to check the possible odd-even effects, the actinide region has been divided in three groups: even group  $^{232}\text{Th}$ ,  $^{234}\text{Th}$ ,  $^{236}\text{U}$ ,  $^{240}\text{U}$ ,  $^{240}\text{Pu}$ ,  $^{244}\text{Pu}$ ,  $^{250}\text{Cm}$ ; odd-even group  $^{229}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{239}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{243}\text{Pu}$ ,  $^{245}\text{Cm}$ ,  $^{249}\text{Cm}$ ,  $^{253}\text{Cf}$ ; even-odd group  $^{232}\text{Pa}$ ,  $^{234}\text{Np}$ ,  $^{236}\text{Np}$ ,  $^{238}\text{Np}$ ,  $^{240}\text{Am}$ ,  $^{242}\text{Am}$ ,  $^{244}\text{Am}$ ,  $^{250}\text{Bk}$ ; and odd-odd group of nuclides  $^{231}\text{Pa}$ ,  $^{235}\text{Np}$ ,  $^{239}\text{Np}$ ,  $^{243}\text{Am}$ ,  $^{247}\text{Am}$ ,  $^{249}\text{Bk}$ . These nuclides uniformly cover the tables of the known fission barrier systematics.

We defined the optimal set of parameters  $C_s$  and  $\alpha_4$  from the condition of the best concurrence with Back systematics [1]. The analysis of the results has not revealed any odd-even staggering in the behavior of parameters. Such a circumstance allow to use the same functions  $C_s(N,Z)$  and  $\alpha_4(N,Z)$  for the whole table. The values of parameters obtained from fitting as well as their parameterization are presented in Table 1. The quality of these parameterizations is sufficiently high – the deviations from data obtained with fitting on barrier systematic are less than 2%.

Table 1

Values of  $C_s$  and  $\alpha_4$  and their (N,Z) dependencies

Nuclides	$C_s$	$\alpha_4$	Nuclides	$C_s$	$\alpha_4$
Th232	17.65	0.055	Cf253	18.9	-0.065
Th234	17.3	0.08	Pa232	18.01	0.046
U236	17.9	0.04	Np234	18.41	0.029
U240	17.55	0.035	Np236	18.15	0.029
Pu240	18.25	0.02	Np238	18.15	0.024
Pu244	17.85	0	Am240	18.24	0.022
Cm250	18	-0.035	Am242	18	0.015
Th229	17.85	0.06	Am244	17.91	0.005
Th233	17.4	0.065	Bk250	18.49	-0.035
U235	17.85	0.048	Pa231	18.07	0.05
U239	17.35	0.07	Np235	18.29	0.033
Pu239	18.05	0.04	Np239	17.9	0.024
Pu243	17.9	0.01	Am243	18.03	0.008
Cm245	18	0.005	Am247	17.97	-0.006
Cm249	17.95	-0.026	Bk249	18.29	-0.024
$C_s = 20.54 (1 - 2.58 (N-Z)^2/A^2) - 6.72 + 0.185 Z^2/A$					
$\alpha_4 = 1.162 - 0.0036 Z - 0.0033 A$					

The obtained function  $C_s(N,Z)$  now contains two terms: the first one is close to the function used in [4] ( $21.7-3(N-Z)^2/A^2$ ); the appearance of the coulomb parameter  $Z^2/A$  in the second term is probably connected to the necessity to improve the coulomb part of the Nix and Sierk model [4]. The hexadecopole deformation falls as mass number of nuclide growths and values of  $\alpha_4$  are close to the commonly used ones (see, for example [6]).

The comparison of the calculated barriers with values given by Back values [1] is presented in Table 2 and Fig. 4. The quite satisfactory results have been obtained as it can be seen in the presented data.

Table 2

The heights of  $Bf_1$  and  $Bf_2$  and the values of  $h\omega_1$ ,  $h\omega_2$  (MeV)

	$Bf_1$ (calc.)	$Bf_1$ (Back)	$h\omega_1$ (calc.)	$h\omega_1$ (Back)	$Bf_2$ (calc.)	$Bf_2$ (Back)	$h\omega_2$ (calc.)	$h\omega_2$ (Back)
Th229	6.82	(6.02)	0.89	(0.9)	7.21	6.30±0.20	0.49	-
Th230	6.33	-	0.88	-	6.7	6.5±0.3	0.52	-
Th231	6.37	(6.02)	0.9	(0.9)	6.76	6.22±0.20	0.55	0.52±0.10
Th232	5.89	<5.50	0.88	-	6.34	6.15±0.20	0.58	0.50±0.10
Th233	5.91	(6.02)	0.91	(0.9)	6.38	6.28±0.20	0.63	0.45±0.10
Th234	5.51	6.15±0.20	0.88	1.00±0.10	6.06	6.52±0.20	0.66	0.75±0.10
Pa231	6.64	5.75±0.30	0.9	(0.8)	6.64	5.85±0.30	0.56	(0.45)
Pa232	6.67	5.75±0.30	0.91	(0.6)	6.76	6.10±0.30	0.6	(0.45)
Pa233	6.22	5.85±0.30	0.9	(0.8)	6.34	6.00±0.30	0.63	(0.4)
U232	6.2	5.54±0.20	0.86	0.80±0.10	5.99	5.45±0.20	0.58	0.55±0.10
U234	5.97	6.20±0.25	0.88	1.00±0.10	5.81	5.95±0.25	0.64	0.65±0.10
U235	6.17	6.10±0.30	0.93	(0.85)	5.91	5.65±0.30	0.68	(0.5)
U236	5.81	5.70±0.20	0.9	0.90±0.10	5.69	5.68±0.20	0.7	0.50±0.10
U237	6.07	6.35±0.30	0.93	(0.85)	5.86	5.95±0.30	0.74	(0.55)
U238	5.7	5.90±0.20	0.9	1.00±0.10	5.6	6.12±0.20	0.74	0.62±0.10
U239	5.93	6.55±0.30	0.93	(0.9)	5.76	6.30±0.30	0.77	(0.65)
U240	5.52	5.75±0.20	0.89	1.00±0.10	5.41	5.95±0.20	0.76	0.70±0.10
Np234	6.57	5.35±0.30	0.91	(0.6)	6.07	5.00±0.30	0.65	(0.42)
Np235	6.26	5.60±0.30	0.91	(0.8)	5.79	5.20±0.30	0.68	(0.55)
Np236	6.49	5.70±0.30	0.96	(0.6)	5.91	5.20±0.30	0.72	(0.42)
Np237	6.13	5.70±0.30	0.92	(0.8)	5.74	5.50±0.30	0.73	(0.55)
Np238	6.36	6.00±0.30	0.95	(0.6)	5.89	6.00±0.30	0.77	(0.42)
Np239	6.01	5.85±0.30	0.92	(0.8)	5.67	5.50±0.30	0.77	(0.55)
Pu238	5.83	5.90±0.20	0.9	0.80±0.10	5.19	5.20±0.30	0.72	0.55±0.10
Pu239	6.13	6.43±0.20	0.93	1.00±0.10	5.41	(5.5)	0.75	(0.55)
Pu240	5.88	5.80±0.20	0.91	0.82±0.10	5.23	5.45±0.20	0.74	0.60±0.10
Pu241	6.16	6.25±0.20	0.93	1.10±0.10	5.46	(5.5)	0.76	(0.55)
Pu242	5.86	5.60±0.20	0.89	0.82±0.10	5.22	5.63±0.20	0.74	0.59±0.10
Pu243	6.1	6.05±0.20	0.91	0.80±0.10	5.38	(5.6)	0.74	(0.55)
Pu244	5.81	<5.6	0.87	-	5.12	5.35±0.20	0.72	0.57±0.10
Pu245	6.06	5.72±0.20	0.88	0.90±0.10	5.33	(5.45)	0.72	(0.55)
Am240	6.32	6.35±0.20	0.94	0.70±0.10	5.36	(4.8)	0.77	(0.42)
Am242	6.33	6.38±0.20	0.93	0.50±0.10	5.37	(4.8)	0.76	(0.42)
Am243	6.1	5.98±0.20	0.91	0.75±0.10	5.14	(4.8)	0.73	(0.55)
Am244	6.31	6.18±0.20	0.92	0.50±0.10	5.3	(4.8)	0.73	(0.42)
Am245	6.02	5.88±0.20	0.88	0.85±0.10	5.02	(4.8)	0.7	(0.55)
Am247	5.83	5.60±0.20	0.85	0.90±0.10	4.73	(4.8)	0.65	(0.55)
Cm244	5.98	6.12±0.20	0.9	0.90±0.10	4.65	<4.9	0.67	-
Cm245	5.4	6.38±0.20	0.8	0.65±0.10	4.84	(4.2)	0.67	(0.55)
Cm247	6.41	6.20±0.20	0.89	0.70±0.10	4.93	(4.2)	0.65	(0.55)
Cm248	5.97	6.15±0.20	0.85	0.90±0.10	4.47	<4.6	0.62	-
Cm249	6.13	5.80±0.20	0.85	0.75±0.10	5.78	(4.2)	0.7	(0.55)
Cm250	5.79	5.15±0.20	0.82	0.72±0.10	3.84	3.90±0.30	0.37	0.69±0.10
Bk249	6.28	6.05±0.20	0.87	0.80±0.10	4.23	(4.2)	0.61	(0.55)
Cf253	6.7	5.60±0.30	0.85	1.10±0.10	3.46	(4.2)	0.42	(0.55)



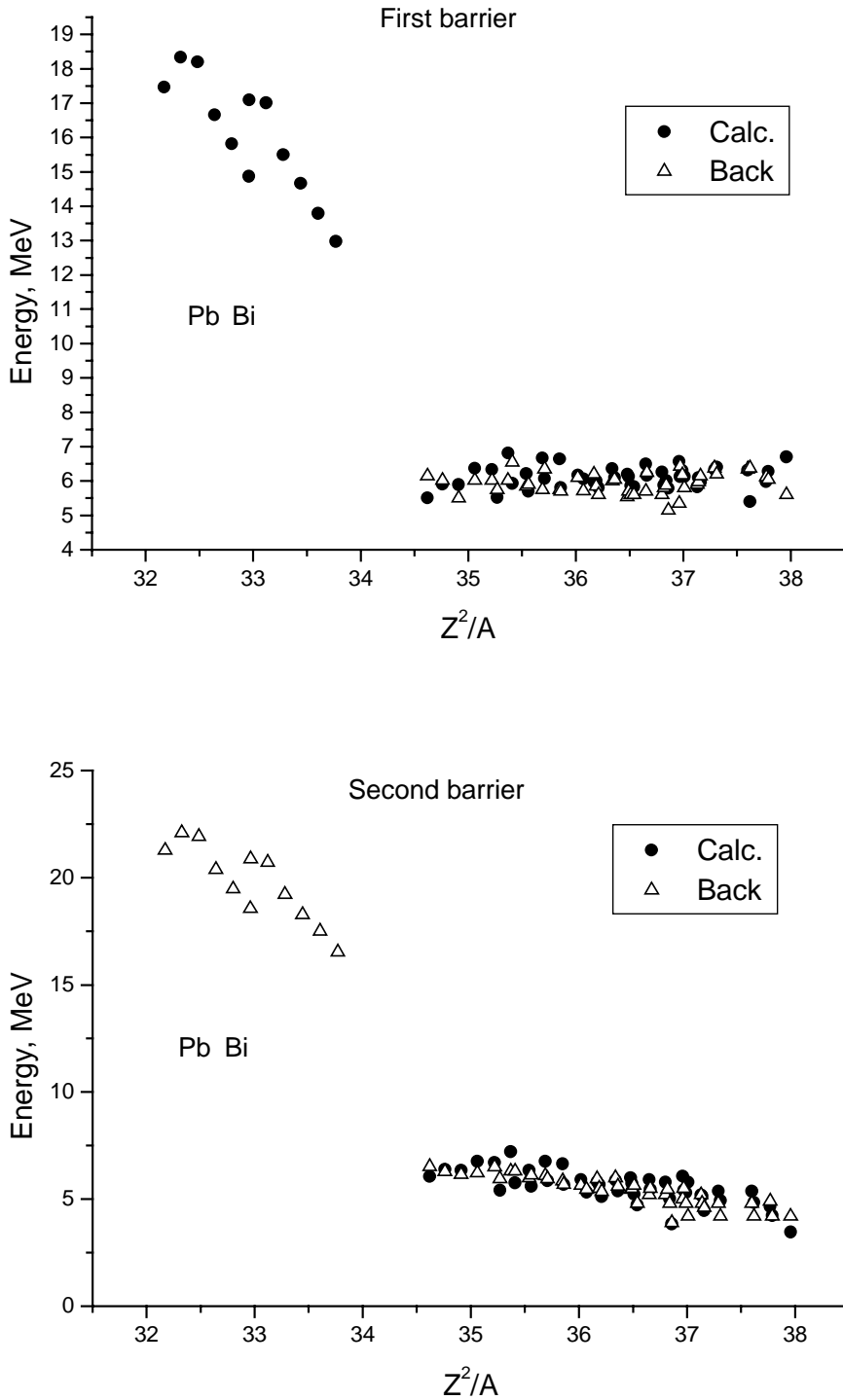


Fig. 4. The comparison of our results with data of Back [1].

The left part of the figures ( $Z^2/A < 34$ ) presents the data obtained for nuclides which are very important for ADS technologies, i.e. isotopes of Pb and Bi ( $^{204,205,206,207,208,209}\text{Pb}$  and  $^{204,205,206,207,208,209}\text{Bi}$ ) where fission barriers are poorly investigated. The barriers in these cases were calculated for  $\alpha_1=0$  (symmetric fragment mass distribution). It is seen from the data that the barrier heights decrease for more neutron-deficient isotopes.

The interaction of high-energy particles (up to 1 GeV for ADS) with target nuclei leads to the formation of high and very high excited nuclei which can undergo the fission. It is necessary for these reactions to include the dependence of fission barriers on excitation energy (or nuclear temperature  $T$ ) into consideration. We use a simple model to demonstrate this kind of dependence,

$E = E_{lq} + (E_{sc} + E_p) \cdot \exp(-T/T_{cr})$ , where  $T_{cr}$  is the parameter. The dependence of deformation energy for the case of  $^{240}\text{Pu}$  on the temperature for  $T_{cr} = 0.75$  MeV is shown in Fig. 5 along the fission axes. The degeneration of structure effects with temperature growth is clearly seen in the figure. At  $T \geq 2$  MeV the barrier is only defined by liquid drop nuclear properties.

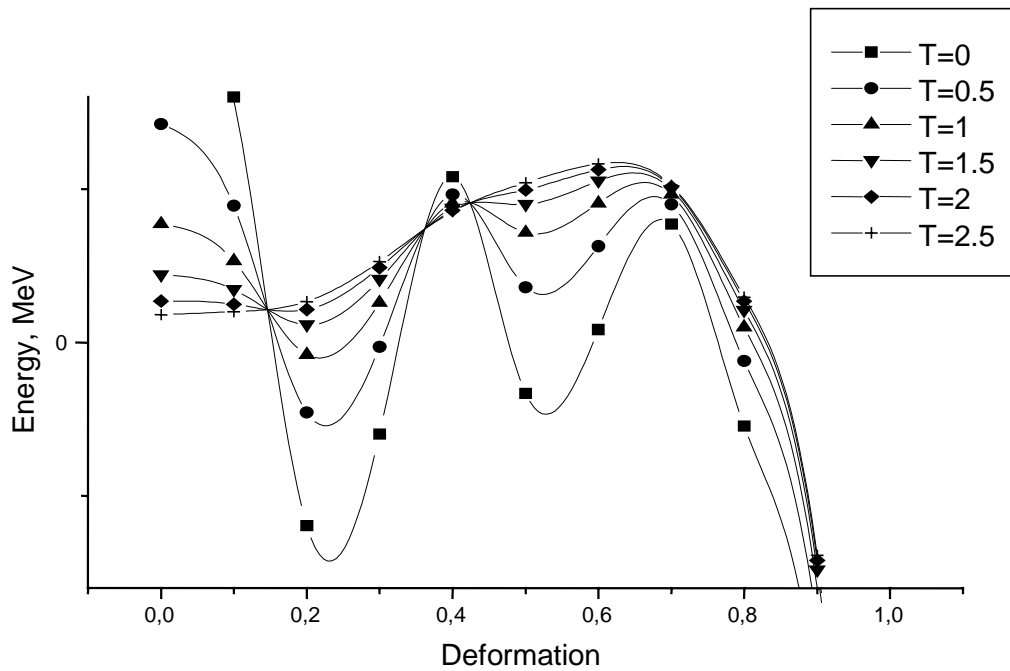


Fig. 5. The temperature dependence of deformation energy.

So, the semimicroscopic model is proposed in this work which allows to calculate the fission barriers of heavy nuclei with account of their possible dependence on excitation energy.

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