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NUCLEAR DATA FOR TRANSACTINIDES IN THE RECOMMENDED EVALUATED NEUTRON DATA LIBRARY (BROND)

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Abstract: The results of the evaluation of the complete system of neutron data for transactinides are discussed. The application of the coupled-channel method even for nuclei with a large spin value (^{235}U), a correct model of level density, a multistep statistical model with taking into account the competition of the fission process as well as the use of new experimental data allow the reliability of neutron data evaluation for transactinides to be increased.

(neutron data evaluation, actinides, optical statistical analysis, excitation functions, level density model)

Introduction

BROND, the USSR computerized neutron evaluated data library, was released in 1987/1988. Upon request it is available on magnetic tape from the IAEA Nuclear Data Section. The characters BROND stand for Biblioteka Rekomendovannykh Ocenennykh Nejtronnykh Dannykh (Library of Recommended Evaluated Neutron Data). BROND contains neutron data for actinides nuclei (isotopes of Th, Np, Pa, Pu, U, Am, Cm), technological and structural materials (Cr, Fe, Ni, Mo, Nb, Zr, Ti, V, Rh, Cd, H, Cl, He, Ca, Li, Be, B, N, Mg, O, F, Na, Al, Si etc.), and fission products (isotopes of Mo, Xe, Tc, Cs, Ru, Sm, Nd,Ce, Pd, Pr, Ag, I, Pm) /1/.

As part of the efforts devoted to the creation of evaluated neutron data library further theoretical and experimental investigations have been carried out, in particular new accurate methods of nuclear data measurements were developed and experimental data were obtained, theoretical models and neutron data evaluation methods were deliberated and used for practical evaluation.

This paper deals with heavy nuclei evaluation problems. For uranium and plutonium isotopes a consistent optical and statistical analysis of mean parameters of neutron resonances, total neutron cross sections, fission cross sections, neutron emission spectra, radiative capture and inelastic cross sections and excitation cross sections for (n,Xn)-reactions was made. Attention was also paid to resonance structure of neutron cross-sections and neutron inelastic excitation functions.

The coupled-channel method for nuclei with large spin values in ground states, more realistic level density models, multi-cascade statistical models with pre-equilibrium decay, different parametrization developments were used for neutron data evaluation quite extensively /2/. New experimental data became available for actinides, for example, fission cross-sections, total cross-sections and the alpha-value measurements, determination of resonance parameters in the resolved resonance energy reason with high energy resolution. The application of realistic theoretical models for analysis of experimental data available and for prediction of data for which no experimental data exist, allowed to raise a level of neutron data evaluation reliability.

Methods Used and Results Obtained

The coupled-channel method was used widely for evaluation and prediction of σ_t, σ_c, σ_n, $\sigma^{dir}_{nn'}$, and T^J_{nlj} for actinides in BROND. Nonspherical optical potential parameters were determined using the SPRT-method /3/. As a result of optimization the following potential parameters for ^{238}U were obtained /2/:

$$V_R = 45{,}87 - 0{,}3E,\ r_R = 1{,}256,\ a_R = 0{,}626$$
$$W_D = \begin{cases} 2{,}95+0{,}4E\ (E \le 10\ \text{MeV}), & r_D = 1{,}260 \\ 6{,}95\ \ (E \ge 10\ \text{MeV}), & a_D = 0{,}555+0{,}0045E \end{cases}$$
$$V_{so} = 7{,}5,\ \beta_2 = 0{,}216,\ \beta_4 = 0{,}080.$$

All the available experimental information on neutron cross-sections for ^{232}Th, ^{235}U, ^{239}Pu, ^{240}Pu, and ^{236}U can be described using the following real and imaginary potential depths:

$$V_R = 49{,}72 - 17\,\frac{N-Z}{A} - 0{,}3\,E$$
$$W_D = 5{,}22 - 10\,\frac{N-Z}{A} + 0{,}4E$$

Geometry potential parameters are the same as in case of ^{238}U and deformation parameters β_2 and β_4 for ^{232}Th, ^{235}U, ^{239}Pu, and ^{236}U are the following: 0,195, 0,078; 0,201, 0,072; 0,217, 0,082; 0,191, 0,094; 0,213, 0,090 /2/.

Comparison of calculated and experimental data for σ_t ^{235}U is given in Fig. 1. The difference between theoretical and experimental total cross sections is not more than 2 % which is within the experimental uncertainties.

In Fig. 2 the experimental /4/,/5/ and calculated /6/ data for the total elastic scattering cross-sections are compared. One can come to the conclusion from Fig. 3 that the old data obtained in the experiments with a not sufficiently high resolution contain the contribution of neutrons inelastically scattered on low-lying levels. This contribution is ~ 10% in the energy region up to 2 MeV and ~ 5% in the energy region 3 - 20 MeV.

Fig. 3 compares the differential cross-sections of elastic and inelastic neutron scattering, calculated by the coupled-channel method, with experimental data available. In the calculated cross-sections, the contribution

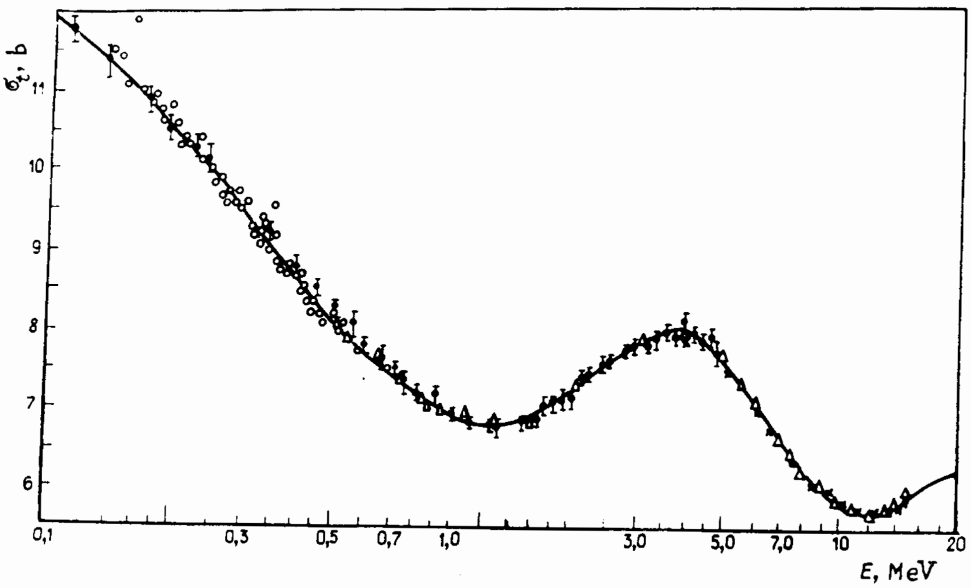

Fig. 1, Comparison of calculated and experimental data for σ_t (^{235}U)

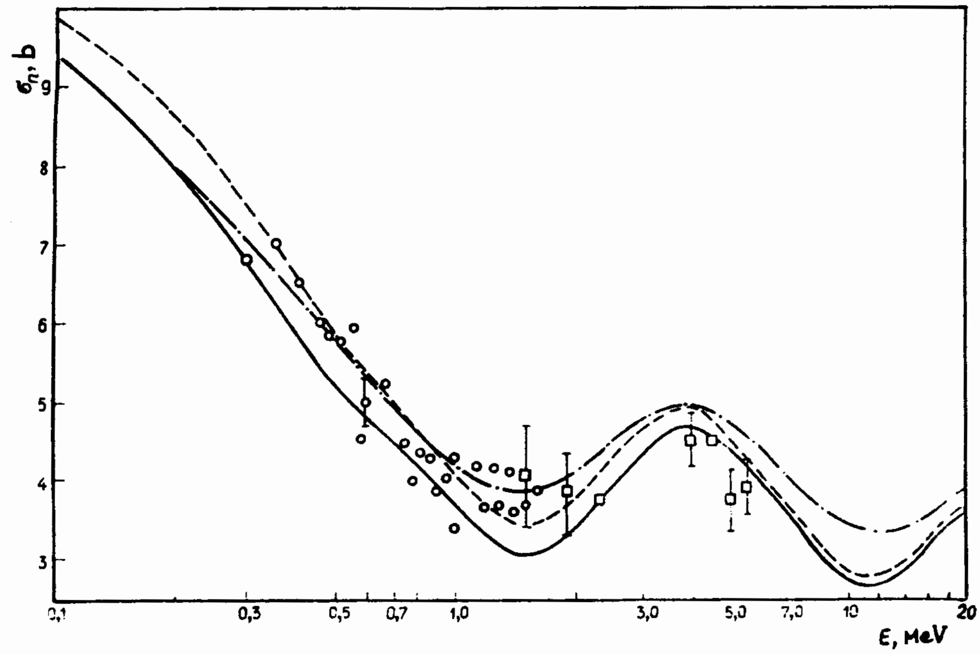

Fig. 2, Comparison of the theoretical /6/ and experimental neutron elastic scattering cross-section for ^{235}U in the energy region 0,1 - 20 MeV (experimental data: □ - /4/, o- /5/). A solid curve - calculation for a ground state ($7/2^-$); a dotted line - calculation for a ground state taking into account the contribution of neutron inelastic scattering for the first five levels (73 eV; 13; 46; 52 and 82 keV); -.- evaluation of ENDF/B - V /7/.

from scattering through a compound nucleus is taken into account which is essential at neutron energies below 4 MeV.

As seen from Fig. 3. the former experimental data on angular distributions of "elastic" scattering obtained with a low energy resolution, can be correctly interpreted only with allowance for contribution from inelastic scattering at least for the first two-three excited levels. Therefore, the evaluted data based on the Legendre polynomial expansion of the experimental data, as a rule, underestimate considerably the anisotropy of elastic scattering.

Comparison of calculated /8/ and experimental /9/ - /11/ data on angular distributions of scattered neutrons at 0,7 and 3,4 MeV for ^{235}U given for a group of levels shows that the agreement with experimental data is good. It is the evidence that both direct and compound processes are calculated

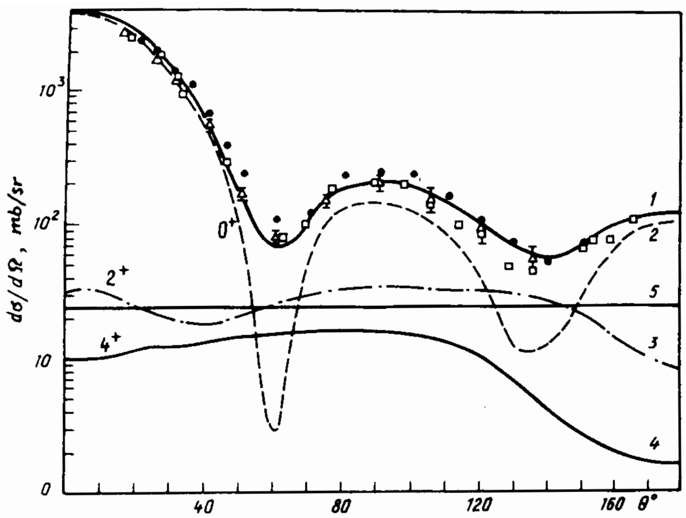

Fig. 3, Comparison of experimental and theoretical data on neutron scattering angular distributions at 2 MeV for ^{238}U (1 - sum of three levels; 2 - elastic scattering on 0+-level; 3, 4 - scattering on levels 2+ and 4+; 5 - sum of compound contributions from three levels)

correctly, because at 0,7 MeV contribution of the compound process is significant (for the level 46 keV, for example, σ_{comp} = 0,168b, σ_{dir} = 0,143b) and at 3,4 MeV it is negligible (< 0,02 %).

Main difficulties in experiments on neutron inelastic scattering cross-sections for ^{235}U are the subtraction of the fission spectrum from a total spectrum and the subtraction of inelastic contributions of low-lying levels from the elastic peak. The neutron inelastic scattering cross-sections for a group of levels of ^{235}U were determined with a not sufficiently high resolution in /12/. In Fig. 4, a comparison of experimental /4, 12, 13/ and calculated data for three groups of levels is given. One can see that theoretical and experimental data agree within experimental uncertainties. More detailed comparison is difficult to make due to a low energy resolution in experiment /12/ (it is not clear, for example, the level 150,5 keV was or was not included in the measured $\sigma_{nn'}$ for a group of levels 100 < Q < 150keV). Of course, it is difficult to make any conclusions about the validity of the theoretical model used for the $\sigma_{nn'}$ (^{235}U) calculation due to a scarcity of experimental data base on ^{235}U inelastic scattering excitation functions.

Note that the calculated $\sigma_{nn'}$ in the region 1,0 - 2,5 MeV are slightly higher than experimental data /4, 12/ obtained with a poor energy resolution (curve 6 in Fig. 4), confirming the assumption made above that the contribution from low-lying levels in these experiments was included into elastic scattering.

The comparison of our evaluated data /6/ and ENDF/B-V /7/ for level excitation functions shows a large difference in the results, which is due to the fact that no contribution from direct processes was allowed for in /7/. The level 170,73 keV is missing in /7/, and the excitation cross-section for the level 172 keV seems to include the excitation cross-sections of two levels (170,73 and 171,36 keV) of evaluation /6/. Besides, there are also differences in the compound contribution to level excitation functions in /6/ and /7/ due to different methods of calculating the fission process and different nuclear level schemes used.

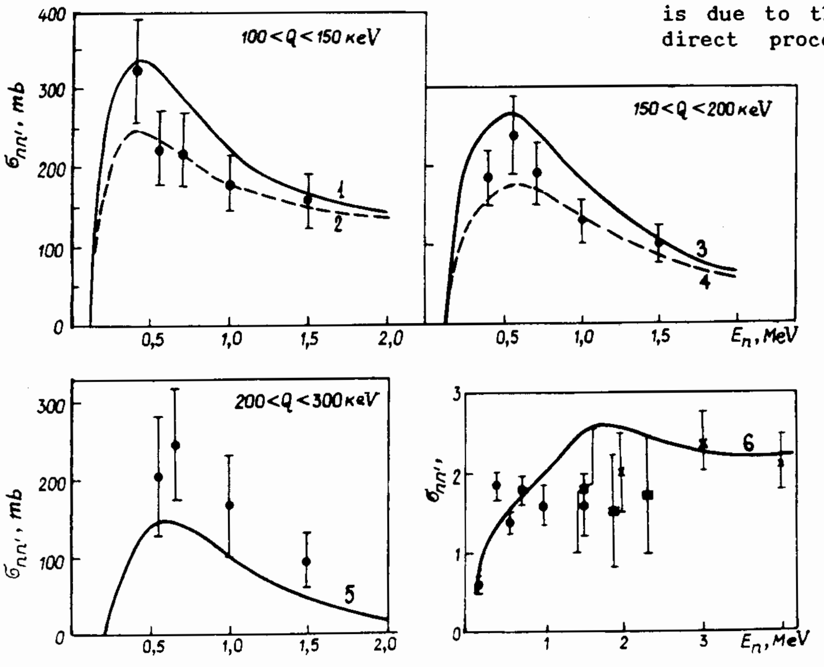

Fig. 4, Comparison of theoretical and experimental data for excitation of group levels of ^{235}U for neutron inelastic scattering. Curve 1 - sum of levels 103; 129,3 and 150,3 keV; 2 - sum of levels 103 and 129,3 keV; 3 - sum of levels 170,7; 171,4; 197,1 and 150,5 keV; 4 - sum of levels 170,7 and 197,1 keV; 5 - sum of levels 225,4; 291,1; 294,7 keV; 6 - the total inelastic cross-section. Experimental data: ɪ - /12/, ▪ - /4/, ✕ - /13/.

Naturally, the differences in the level excitation functions led to the differences in the total inelastic cross-sections as much as 1,5 - 2,0 times (Fig. 5). The major difference in the region from 0,1 - 2,0 MeV is due to the neglection of direct processes for low-lying states of ^{235}U in ENDF/B-V. In the region above 7 MeV, a larger $\sigma_{nn'}$ obtained in /6/ is attributed to the contribution of pre-equilibrium processes. Both these effects lead to a more rigid neutron emission spectrum. ^{239}Pu inelastic scattering cross-section for excitation of levels above the experimental threshold set obtained by Smith /14/ is compared in Fig. 6. Our evaluated data /15/ are in good agreement with the evaluation of Arthur /16/.

It is seen from the results given above that a generalized optical model should be used as a method for reliable evaluation of neutron cross sections of heavy deformed nuclei. The application of the spherical optical model is possible in cases when an accuracy required is not high (about 20 - 30 %).

The generalized optical model allows to calculate direct inelastic scattering cross-sections and its use leads to changing neutron transmission coefficients for different partial waves. The compound formation cross-section is firstly affected by neutron transmission coefficients and hence inelastic scattering cross-sections, total and on levels, will depend on the reliability of neutron transmission calculations. An example of using neutron transmission coefficients calculated with spherical and nonspherical models for ^{239}Pu level excitation functions /2/ is given in Fig. 7.

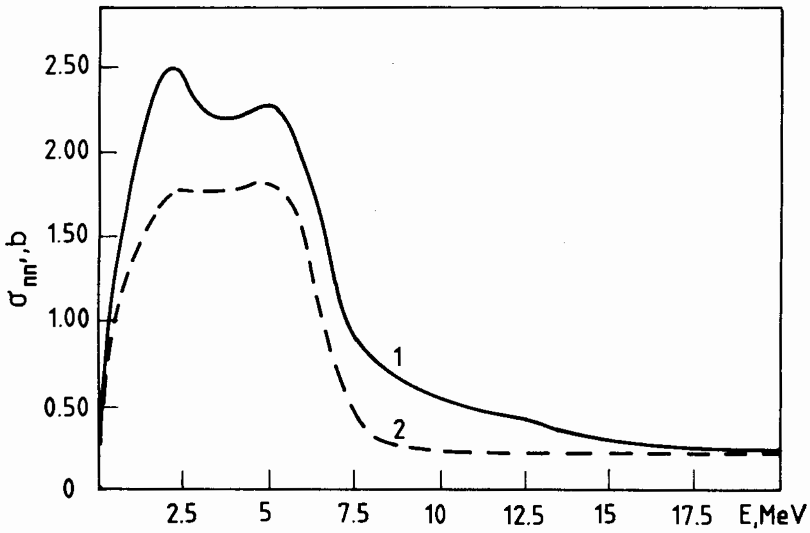

Fig. 5, The neutron inelastic scattering cross-section for ^{235}U from the threshold up to 20 MeV. Solid curve - calculations /6/, dotted curve - ENDF/B-V /7/.

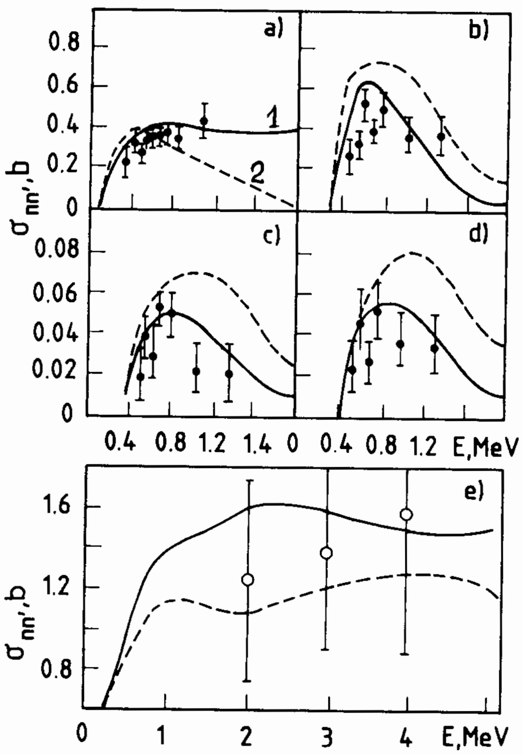

Fig. 7, Comparison of experimental and evaluated data for level excitation functions for ^{239}Pu. 1 - the coupled channel method used for neutron trans-mission calculations; 2 - spherical optical model. a - sum of levels 57 and 76keV; b - level 285 keV, c - 330 keV; d - sum of levels 387 and 392keV; e - total inelastic cross-section.

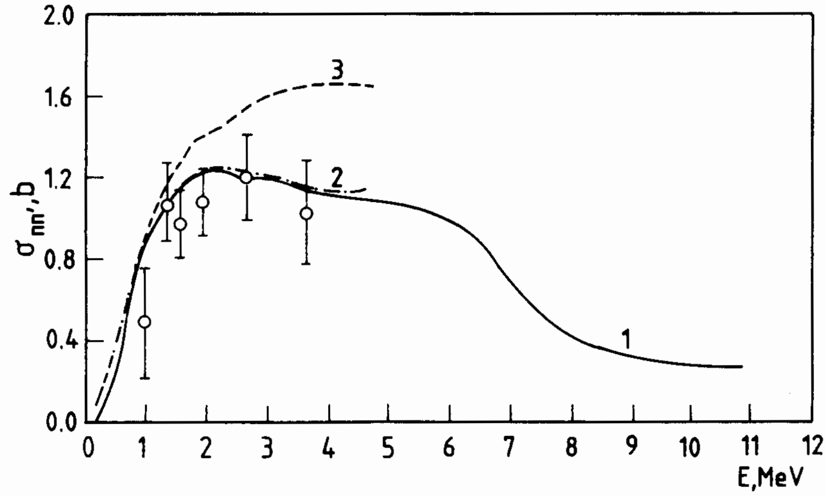

Fig. 6, Comparison of inelastic scattering data for ^{239} Pu obtained by Smith /14/ with evaluations /15/ (curve 1), /16/ (curve 2) and ENDF/B-V (curve 3).

It is seen that usage of transmission coefficients from the coupled channel method and taking into account the direct excitation of low levels allow to get a better agreement with experimental data not only for lower levels but also for that levels excitation of which determined fully by the compound nucleus decay. One can note that experimental data on the total inelastic scattering cross-section are too low because they do not contain the contribution of the first excited level with energy 8 keV.

The nuclear level density is of great importance for all practical applications of the statistical theory of nuclear reactions. The Fermi-gas model is mostly used due to its simplicity. But inconsistency of this model with the conclusions of the microscopic theory and some experimental data /17/ led to the revision of applicability of this model for the neutron cross-section analysis. The microscopic approach based on combinatorial methods of calculations of energies of highly excited multiparticle states was developed /18/. But these methods are too time-consuming to be applied for nuclear data evaluation. The statistical method of description of average characteristics of excited nuclei developed by Ignatjuk et al. /19/ is rather simple and convenient for practical calculations and at the same time includes the main results of the microscopic theory. The method takes into account a shell structure in a single-particle spectrum, superfluid correlation effects and coherent collective effects.

In Fig. 8 the results of ^{239}Pu excitation level function calculations made for different level density models are given.

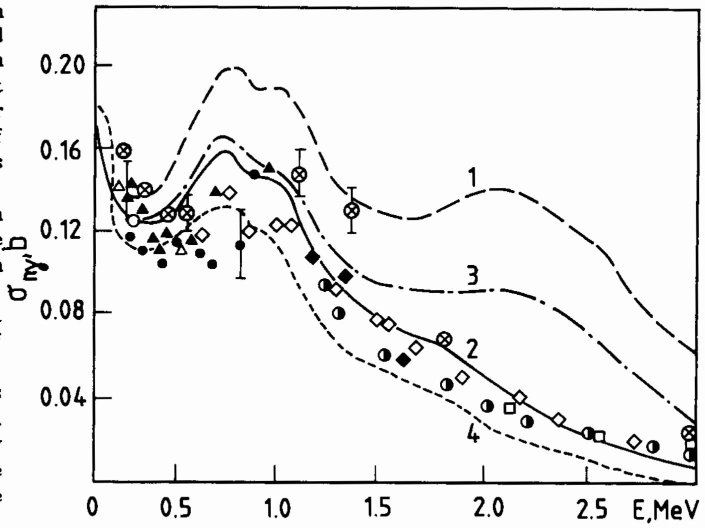

Fig. 9, Comparison of experimental and theoretical data for $\sigma_{n\gamma}(^{238}U)$ obtained with different level density models. Curves: 1 – the Fermi-gas model, $\Gamma\gamma$-energy dependence – Lorenz type; 2 – the Fermi-gas model involving collective effects, $\Gamma\gamma$-Lorenz energy dependence, 3 – superfluid nucleus model involving collective effects, $\Gamma\gamma$-Lorenz energy dependence. 4 – same as 2, but for $\Gamma\gamma$-Weiskopf energy dependence used ($\langle D\rangle_{obs}$ = 24,8 eV, $\langle\Gamma\gamma\rangle_{obs}$ = 23,5 meV, T_n calculated by the coupled channel method)

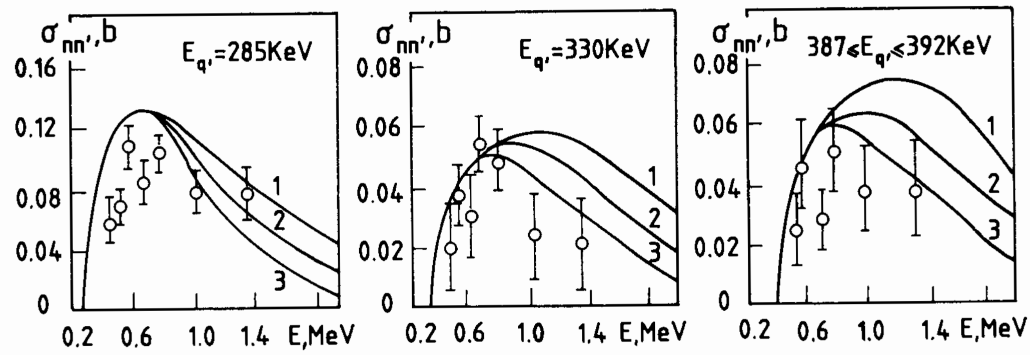

Fig. 8, Level excitation cross-sections for ^{239}Pu for different level density models (1 - Fermi-gas; 2 - superfluid nucleus; 3 - Fermi-gas involving collective effects according to /19/).

It is seen from Fig. 8 that the best description of experimental data is reached by using the Fermi-gas model involving collective effects. The superfluid nucleus model gives a somewhat worse description in a higher energy region.

The calculated values of radiative capture cross-sections $\sigma_{n\gamma}$ depend strongly on the level density model used which affects radiative transmission coefficients and, hence, $\sigma_{n\gamma}$. In Fig. 9 a comparison of calculated and experimental data for $\sigma_{n\gamma}$ (^{238}U) is given.

The best agreement with experimental data on $\sigma_{n\gamma}$ (^{238}U) can be obtained by using the Fermi-gas level density model involving collective effects and Lorenz $\Gamma\gamma$ energy dependence. Similar results were obtained /2/ for ^{239}Pu and ^{240}Pu.

The attempt to apply the superfluid model or the Fermi-gas model involving collective effects for description of cumulative level sums N(E) for transactinides, for which an experimental information on low-lying levels is available, leads to unsatisfactory results. A satisfactory description of N(E) can be obtained by using a constant temperature model for low-lying levels in the region up to 1,0 - 1,5 MeV. For transactinides $\bar{T}$ fluctuates weakly around its average value $\bar{T}$ = = 0,385 MeV for even-even nuclei. For odd nuclei the fluctuation of the parameter $\bar{T}$ is more significant, but $\bar{T}$ is practically the same as for even-even nuclei.

For nuclei with a little experimental information for low-lying states one can use the average parameters $\bar{T}$ = 0,385 MeV and $E_o = 0$, $E_o = -\Delta_o$ and $E_o = -2\Delta_o$ for even-even, odd, and odd-odd nuclei, respectively.

Using these parameters one can obtain a good description of cumulative level sums for nuclei, in first turn for even-even nuclei, spectra of which is studied in details, but often the data on $\langle D\rangle_{exp}$ are absent.

Systematics of parameters T and E_o can be used for calculations of the main level density parameter a. The dependence of the parameter a (Bn) on atomic mass A can be represented by /20/:

$$a(Bn) = 1,619.10^{-3}A^2 + 0,4730A$$

Theoretical models which have been tested against experimental data available for ^{238}U, ^{235}U, ^{239}Pu were used for ^{236}U neutron data evaluation and theoretical prediction /21/. The up-to-date analysis led to more reliable data for inelastic scattering excitation functions, (n, 2n), (n, 3n) cross-sections, angular distributions of elastically and inelastically scattered neutrons, total, inelastic and elastic cross-sections for which experimental data are not available.

In Fig. 10 a comparison of σ_t, σ_n, $\sigma_{nn'}$ obtained in our evaluation /21/ and /22/ is given. For σ_t in the energy region 0,1 - 1,0 MeV the ENDF/B-V evaluation is 10% higher than /21/, in the region 2 - 20 MeV both evaluations are in agreement whithin 3%.

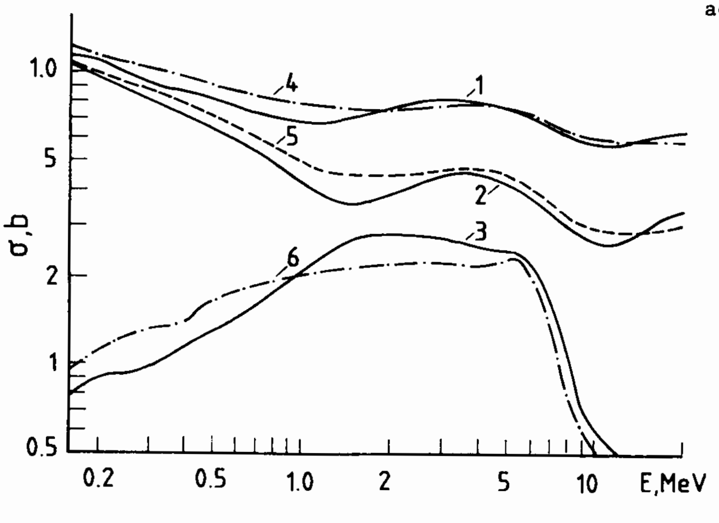

Fig. 10, The total cross-section (curve 1), elastic scattering (curve 2) and inelastic scattering (3) for ^{236}U evaluated in /21/ and the comparison with ENDF/B-V /22/ (curves 4, 5, and 6 - for σ_t, σ_n and $\sigma_{nn'}$ respectively).

When comparing two evaluations on σ_n and $\sigma_{nn'}$, for ^{236}U the same tendency is observed as in case of ^{235}U, namely, σ_n in /22/ is higher than in /21/ in the energy region 1 - 2 MeV by 20%, but $\sigma_{nn'}$ in this region lower by 20 %. It can be explained by the fact that as in case of ^{235}U in the ENDF/B-V /22/ the inelastic scattering contribution of low-lying levels was included into the elastic channel. Higher values of $\sigma_{nn'}$ in the region 1 - 5 MeV obtained in /21/ are the results of the contribution of direct processes on low-lying levels. In the higher energy region the contribution of pre-equilibrium effects taken into account in /21/ leads to larger $\sigma_{nn'}$.

In Fig. 11 the evaluated level excitation functions for the first two levels of ^{236}U are given and comparison is made with the ENDF/B-V. It is seen that in the ENDF/B-V a direct excitation process of the levels 45,24 keV and 149,48 keV is not taken into account.

A satisfactory description of the energy dependence of fission cross-sections was obtained by using the Fermi-gas model with shell and collective effects /19/, taking into account the energy dependence of shell effects, increasing the correlation function Δ_f on 0,07 MeV of the fissile nucleus in a transition state, and by obtaining the correct parametrization of the constant temperature model near the fission thresholds /21/. The fission and (n, 2n), (n, 3n) - cross-sections calculated within the framework of this model are shown in Fig. 12 and 13.

Based on the abovementioned, one can come to the conclusion that the complete evaluated neutron data files for actinides in the energy region from 10^{-5} eV to 20 MeV which are a part of the BROND library, are created by using up-to-date theoretical models and the all experimental data base available. Comparison with the evaluated data of ENDF/B-V for several actinides shows some deficiencies of these data.

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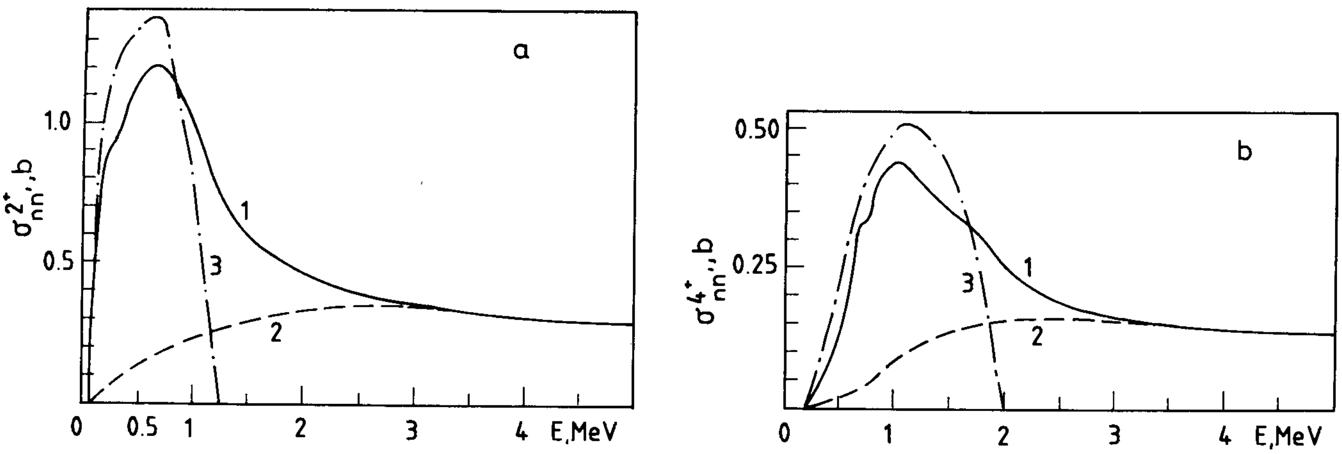

Fig.11, Excitation cross-sections for ^{236}U levels: a - 2+ (45,24keV; b - 4+ (149,48keV); (1 - total, 2 - direct; 1,2 - evaluation /21/, 3 - evaluation ENDF/B-V /22/).

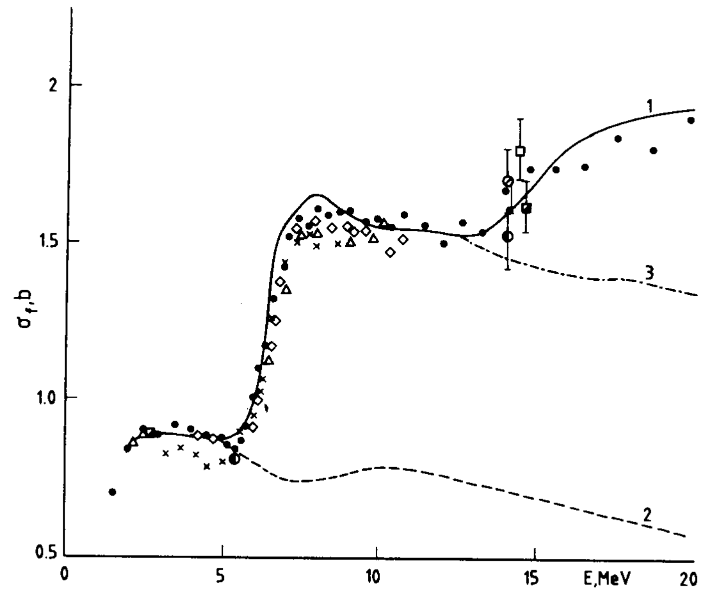

Fig. 12, Comparison of experimental and calculated /21/ σ_f (^{236}U) (1 - σ_{nF}; 2 - σ_{nf}; 3 - σ_{nf} + $\sigma_{nn'f}$).

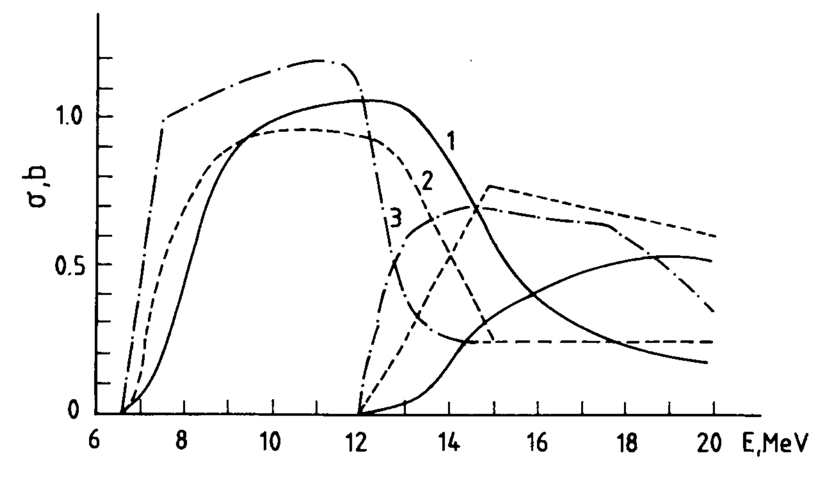

Fig. 13, (n, 2n) and (n, 3n) - reaction cross-sections for ^{236}U (1 - /21/, 2 - /22/, 3 - /23/).

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