ANALYSIS OF THE ²³⁷Np FISSION CROSS-SECTIONS AND THE (n, xn) REACTIONS

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ABSTRACT

Experimental and evaluated cross-section data on fission, (n,3n) and (n2n) reactions leading to the short-lived state of the ^{236}Np nucleus is analyzed using a selfconsistent statistical approach. Integral and differential (n,2n) reaction data are compared. Earlier evaluations appear to be inconsistent with recent experimental data.

In order to evaluate the accumulation of ²³²U in spent fuel from nuclear reactors, it is necessary to have a fairly accurate knowledge of certain nuclear physics constants. Of particular importance is the cross-section for the ²³⁷Np(n,2n) reaction which produces short-lived ²³⁶Np^s and the long-lived ²³⁶Np¹ states of the 236 Np isotope with the half-lives of 22.5 hours and 1.55 x 10⁵ years respectively [1]. The available experimental cross-section data for the $^{237}Np(n,2n)^{236}Np^{3}$ reaction [2-7] do not cover the whole neutron energy range of 6.8-20 MeV which is of interest. For the $^{237}Np(n,2n)^{236}Np^1$ reaction, there are no other data apart from the isomeric ratio, r, of the ²³⁶Np¹ and ²³⁶Np yields for 14 MeV neutrons. Therefore, the existing evaluations of the cross-section \mathbf{g}^{s}_{n2n} are based on model calculations of the \mathbf{g}_{n2n} cross-section, and the σ_{n2n}^{s} cross-section is determined from $\sigma_{n2n}/(1 + r)$ on the assumption that r does not depend on the energy of the incident neutron. As shown in Refs [8 and 9], the latter assumption is not justified. Moreover the model calculations for the cross-section $\boldsymbol{\sigma}_{n2n}$ have the disadvantage that the fission cross-section $\boldsymbol{\sigma}_{nf}$ is used in these calculations only as a parameter.

In view of the above circumstances and in view of the appearance of new experimental fission cross-sections data

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which differ significantly from previous evaluations [10 and 11] it is necessary to establish a consistent analysis of the latest experimental fission cross-sections data , data on the $^{237}Np(n, 2n)^{236}Np^{s}$ reaction and on the isomeric ratio.

The absolute value of σ_{n2n}^{s} can be obtained by normalizing the energy dependence σ_{n2n}^{s} [10 and 12] to the experimentally measured integral cross-section for the fission spectrum $\langle \sigma_{n2n}^{s} \rangle$. Therefore, it is also necessary to analyse the consistency between the differential and integral data on the cross-section for the ²³⁷Np(n,2n)²³⁶Np³ reaction obtained for the ²³⁵U fission spectrum [13 and 14] (which differ by a factor of about 2.5) and for the ²⁵²Cf spontaneous fission neutron spectrum [15].

EXPERIMENTAL DATA

Fission cross-section for ²³⁷Np for neutrons above the (n,nf) reaction threshold. The experimental data in the energy range under consideration can be divided into two groups: absolute measurements [16] and measurements of the ratio of the ²³⁷Np fission cross-section to the $^{235}U(\boldsymbol{\sigma}_{nf}^{7}\boldsymbol{\sigma}_{nf}^{5})$ [15-21] or $^{239}Pu(\boldsymbol{\sigma}_{nf}^{7}/\boldsymbol{\sigma}_{nf}^{9})$ [22] fission cross-sections. The results of the absolute measurements carried out using the time-correlated associated particle method for $E_n = 14.7$ MeV are significantly different from the results of the relative measurements carried out using the "threshold cross-section" method. In the region of the (n,nf) reaction threshold, the experimental data [17-19] are in good agreement with the data in Ref. [16]. The systematically higher values of the data in Ref. [20] as compared with the data in Refs. [18 and 19] is evidently associated with the absolutization of the ratio $\sigma_{nf}^{7}/\sigma_{nf}^{5}$ in Ref. [20], which was based on a comparison of the $\alpha-$ activities of the $^{237}\mathrm{Np}$ and $^{235}\mathrm{U}$ shells which is in itself very unreliable, since the half-life of ²³⁷Np is measured only in one reference. The higher levels in Ref. [22] as compared with Refs [18 and 19] may, to some extent, be attributed to the fact that for the absolutization of the cross-sections for the $\sigma_{nf}^2/\sigma_{nf}^9$ ratio, data on the cross-section $\boldsymbol{g}_{\text{nf}}^{9}$ from Ref. [23] were used.

Thus, in the data given in Refs [16-22], there are discrepancies both between the relative measurements of the different authors as well as between the absolute and relative data normalized to the cross-section σ_{nf}^{5} [10]. In the latter case the discrepancy is approximately equal to the value of σ_{n2n}^{5} for $E_n = 14.7$ MeV. The situation is complicated by the fact that the energy dependence of the cross-section σ_{nf}^{7} from the data in Ref. [19], covering the whole energy range of interest to us, shows that the (n,2nf) reaction makes an extremely small contribution to the fission cross-section studied which is not consistent with the isotopic fissibility dependence of neptunium isotopes [24]. These discrepancies may be associated with experimental errors in the measurement of the $\sigma_{nf}^{7}/\sigma_{nf}^{5}$ ratio and with the fact that the evaluation of σ_{nf}^{5} in the ENDF/B-V library [10] is used to obtain the cross-section σ_{nf}^{7} for $E_n > 14$ MeV.

In order to resolve the contradictions between the data of the different authors on the cross-section σ_{nf}^7 for $E_n > 14$ MeV, let us turn to the data in Refs [25-27]. The measurement of the energy dependence of the fission cross-section in the 9-22 MeV range is reported in Ref. [25]. When these results are normalized to the data in Ref. [16] at $E_n = 14.7$ MeV, the data for $E_n < 14$ MeV agree with the data in Ref. [19] and for $E_n > 14$ MeV they show that the (n, 2nf) reaction makes a significant contribution to the observed fission cross-section . In Ref. [26] the fission cross-section is measured in the 5-22 MeV range; however, these data are normalized to the value of σ_{nf}^{7} , equal to 1.62 b ($E_n = 3.4 \text{ MeV}$) . Renormalization of the data in Ref. [26], on which the ENDF/B-V [10] and KEDAK-4 [11] evaluations are based, to the value of $\sigma_{nf}^{7}(E_{n} = 3.4 \text{ MeV})$ equal to 1.56 b [19] does not significantly change the cross-sections in the high neutron energy region, and after normalization of these data to the data at $E_n = 14.7 \text{ MeV} [16]$, they more or less agree with the data for $E_n > 9$ MeV [25]. The data in Ref. [27] are not taken into account since they are twice renormalized in Refs. [25 and 26] in order to improve the fission fragment recording efficiency.

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Thus, as reference values for $E_n \leq 14$ MeV we have selected data from Refs [17-19, 25 and 26] and for $E_n > 14$ MeV, we have selected data from Ref. [16] and renormalized data from Refs [25 and 26] (Fig. 1).

Cross-section for the $\frac{237}{Np}(n,2n)\frac{236}{Np^2}$ reaction. The measurement of this cross-section in the 13.8-15 MeV energy range was reported in Refs [2-6] and in the threshold region at $E_n = 7.10$ MeV in Refs [5 and 7]. All the measurements were made using the activation method by recording the a-activity of the ²³⁶Pu nuclei. They differ only in the methods used to determine the neutron flux. In Refs [3, 4 and 7] the flux was measured relative to the $^{27}Al(n, \alpha)$ reaction and was also monitored [7] with respect to the ²³⁸U(n,f) and ²³⁸U(n,2n) reactions. In Refs [4 and 6], one of the methods used to determine the flux involved evaluating the accumulation of ⁹⁷Zr nuclei during fission of ²³⁷Np. However the use of the fission cross-section, which was 10% higher than the data in Ref. [16], resulted in an overestimation of the $\sigma^{s}_{n,2n}$ cross-section. Therefore, the data obtained in this way in Ref. [4] were not taken into account. The data which resulted from the measurement in which the flux was determined with respect to the $^{238}U(n, f)$ and $^{237}Np(n, f)$ reactions [6] need to be renormalized to the corresponding cross-sections given in Ref. [16]. As a result, the difference in the cross-sections obtained using the two methods of determining the neutron flux [6], was reduced from 5 to 1%. The value of $\sigma_{n,2n}^{s}$ obtained from measurements of the γ -activity of ²³⁶U [6], is 15% lower than the data in Refs [3-6] for the corresponding energy and therefore we will not take it into account.

In the neutron energy region 10-13.8 MeV there are no neutron reaction data available. Information on the $\sigma_{n,zn}^2$ cross-section may be derived from the measurements results of the cross-sections for the ²³⁵U(t,2n)²³⁶Np^s and ²³⁶U(d,2n)²³⁶Np^s [28] reactions. By assuming that the probability of two neutrons being emitted is not dependent upon the way in which the compound nucleus is formed, the reaction cross-section $\sigma^w(n,2n)$ may take the form [28]: $\sigma_{n2n} = [\sigma_{nf}^2/\sigma_{t(d)f}]\sigma_{t(d)2n}^s$ (1) In Ref. [28], data from Ref. [26] were used to determine the σ'_{nf} cross-section; as a result, $\sigma^{s}_{n,2n}$ was over-valued. In this work the calculated σ_{nf} cross-section was used to calculate $\sigma^{s}_{n,2n}$ in accordance with this expression. Below we discuss the agreement of the data obtained in this way with the results in Refs [2-7] and the present work.

<u>Isomeric ratio in the ²³⁷Np(n,2n) reaction</u>. In Ref. [29] the isomeric ratio r = 0.35 was obtained in a thermonuclear explosion at an average neutron energy of 14 MeV. In Ref. [30] the measurements r = 0.41 in the ²³⁷Np(γ ,n) reaction are given for an excitation energy corresponding to $E_n = 9.6$ MeV. From these data it follows that for $E_n = 9.6-14$ MeV, $r(E_n)$ should decrease as the neutron energy increases. This tendency is confirmed by data from Ref. [31], obtained during the study of the ²³⁸U(d,4n) reaction at $E_d = 21$ MeV, which showed that the states of the ²³⁶Np compound nucleus with spin J = 1 are approximately seven times more frequent than for states with spin J = 6. This means that $r(E_n \approx 19$ MeV) ≈ 0.14 . No direct or indirect information on cross-sections for the interaction of neutrons with the ²³⁷Np nucleus is available.

CALCULATION OF THE CROSS-SECTIONS FOR FISSION AND FOR (N,XN) REACTIONS.

The Hauser-Feshbach statistical theory was used to calculate the cross-sections for the (n,f) and (n,xn) reactions, taking into account conservation of spin and parity for all nuclear reaction cascades [32].

In view of the lack of experimental data which would make it possible to determine the optical potential parameters for ²³⁷Np, the neutron attachment coefficients necessary for the statistical calculations were calculated with the potential [33] for ²³⁸U. This approximation is justified on the basis of the weak isotopic dependence of the neutron absorption cross-section in the energy range studied. The level density in the neutron and fissile channels $\mathbf{p}_{n(f)}(U,J)$ was calculated in the following way. For excitation energies U, which is smaller than energy $U_{n(f)} = (10.7 - \mathbf{m} \Delta_{n(f)}) - 0.028$ A MeV, where m is 0,1,2 for even-even, odd-even and odd-odd nuclei respectively, A is the mass number, $\Delta_{n(f)}$ is the correlation function in the (strongly deformed) ground state, $\mathbf{p}_{n(f)}(U,J)$ is determined using the constant temperature model [34]. In the neutron channel

$$\mathcal{P}_{n} = \frac{1}{T_{n}} \exp\left(\frac{U + m\Delta_{n}}{T_{n}}\right) \frac{2J + 1}{2\sigma^{2}} \exp\left[-\frac{J(J + 1)}{2\sigma^{2}}\right], \qquad (2)$$

where $\Delta_n = 12/\sqrt{A}$ MeV; $T_n = 0.385$ MeV. The parameter for spin dependence of σ^2 at excitation energies U < U_x, where $U_x = 1.2-0.3$ (m + δ_{2m}) MeV, is the boundary of reliable identification of the spin levels and δ_{2m} is the Kronecker δ -symbol determined by the expression $\sigma_n^2 = 0.156A - 26.76$. For $U > U_x$, σ^2 is determined by the linear extrapolation between σ_n and $\sigma^2 F t(U_n)$. Here F_1 is the perpendicular inertia moment and $t(U_n)$ is the thermodynamic temperature at excitation energy U_n .

In the fissile channel the level density is determined from the expression:

$$\rho_f(U,J) = \frac{R}{T_f} \exp\left(\frac{U + m\Delta_f + \delta_f}{T_f}\right) \frac{1}{2\sigma_f^2} \sum_{\kappa=-J}^{J} \exp\left(-\kappa^2/2\kappa_0^2\right) \exp\left[-\frac{J(J+1)}{2\sigma_{\perp f}^2}\right] \cdot$$

Here $K_0^2 = (1/\delta_{\parallel f}^2 - 1/\delta_{\perp f}^2)$, where $\delta_{\parallel f}^2 = F_{\parallel f}t$ ($F_{\parallel f}(F_{11f})$ is the parallel inertia moment). The parameter T_f is determined from the condition

$$T_f = \left\{ \frac{d}{dU} \left[\ln \rho_f(U) \right]_{U=U_f} \right\}^{-1},$$

where $\rho_f = \sigma_{\perp f}^2 \omega_f(U) / \sqrt{2\pi} \sigma_{\parallel f}$. The δ_t parameter is determined from the continuity condition of the level density $\rho_t(U)$ for the excitation energy $U = U_t$ (the corresponding parameter in the neutron channel equals zero): $(1/T_f) exp \left[(U_f + m\Delta_f + \delta_f) / T_f \right]^T = \rho_f(U_f)$. The coefficient R reflects the effect of the saddle configuration asymmetry on the level density. For the internal hump where there is axial and mirror asymmetry, $R = 2\sqrt{2\pi} 6_{\parallel f}$; for the external hump only the mirror symmetry is violated and R = 2 [35]. The density of the internal states $\boldsymbol{\omega}_{n(f)}(U)$ and the spin dependence parameters $\boldsymbol{\sigma}^2_{11}$ and $\boldsymbol{\sigma}^2_{11}$ are determined from the relationships given in Ref. [36], and the correlation function $\Delta_f = \Delta_m + 0.08$ MeV is determined from the description of the fission cross-section energy dependence in the first plateau region. The shell corrections δW_f for the internal and external humps are taken from Ref. [23]. The main level density parameter $a_{f(n)}$ is determined from the relationships in Ref. [36] and its asymptotic value $a_{f(n)}$ from the expression given in Ref. [34] $a_{f(n)} = 0.473A -$ 1.619 x 10⁻³A². The value of the parameters T_f and $\boldsymbol{\delta}_f$ for the internal A and external B humps are: $T_{f}^{A} = 0.38 \text{ Mev}, T_{f}^{B} = 0.39$ Mev, $\delta^{R}_{f} = 0.001$, $\delta^{B}_{f} + 0.24$.

For excitation energies $U > U_{n(f)} \ \mathbf{p}_{n(f)}(U,J)$ is determined from the relationships of the superfluid model [36]. A more detailed model for calculating the level density and the fissile channel permeability, together with the necessary parameters are described in Refs [34, 36, 37], and the method used to calculate the radiation widths is described in Ref. [34].

Let us assume that the main parameter of the pre-equilibrium decay model, the two quasi-particle interaction matrix element $M^2 = 10/A^3$, taken from the description of the spectra for inelastically scattered neutrons for the ²³⁸U nucleus [38], can also be used in the case of ²³⁷Np. This assumption fixes the behavior of the "first chance" fission cross-section. The barrier parameters of the compound nucleus ²³⁷Np, which is fissile in the (n,nf) reaction, are taken from the description of the experimental data for the cross-section σ_{nf} below the (n,2nf) reaction threshold, and the barriers of the ²³⁶Np nucleus are taken from the description of σ_{nf} above the (n,2nf) reaction threshold. Comparison of the experimental and calculated data for the cross-section σ_{nf} is given in Fig. 1, showing the "first

chance" fission cross-section. The energy dependence of the contribution of the "first chance" fission cross-section g_{nf}^1 to the observed fission cross-section σ_{nf} , i.e. $\alpha = \sigma_{nf}^1/\sigma_{nf}$ can be compared with the data for **a** obtained from the analysis of the dependence of the total average energy of the prompt gamma radiation emitted during fission on the average number of prompt fission neutrons [30 and 40]. As can be seen from Fig. 2, these data agree well with our evaluation of **a**. The cross-section $\sigma_{n2n} = \sigma_{n2n}^{1} + \sigma_{n2n}^{s}$ for the ²³⁷Np(n,2n) reaction, calculated at the same time as the fission cross-section, agrees with data from Refs [2-6] (Fig. 3(a)) provided that the isomeric ratio at $E_n = 14-15$ MeV is virtually constant and equal to 0.35 [29]. When $E_n < 13.5$ MeV our cross-section differs significantly from the evaluations in the ENDL [41], KEDAK-4 and ENDF/B-V [30] libraries and when $E_n > 15$ MeV it differs from the evaluations in the ENDL and ENDF/B-V libraries. All the evaluations for the $\sigma_{n^{2n}}$ cross-section in the 14-15 MeV energy range agree (with the exception of the ENDF/B-V evaluation) because they are normalized to the experimental data for the cross-section σ^{s}_{n2n} taking into account the isomeric ratio [29]. In the 9-13 MeV energy range the discrepancy between the ENDL evaluation and the KEDAK-4 evaluation may be linked to the fact that the ENDL evaluation for the fission cross-section is based on the data in Ref. [19], and the KEDAK evaluation is based on data in Ref. [26] (see Fig. 1). The evaluation of the fission cross-section in the ENDF/B-V library is also based on the data in Ref. [26]; however, the evaluation for the cross-section σ_{n2n} is significantly lower than the KEDAK-4 evaluation. This is related to the fact that in the ENDF/B-V library, the cross-section for the ²³⁷Np(n,2n) reaction is determined as $\sigma_{n2n} = 1.35 \sigma_{n2n}^{s}$ and σ_{n2n}^{s} by normalization of the dependence $\sigma_{n2n}^{s}(E_n)$ [12] on the integral data for $\langle \sigma_{n2n}^{s} \rangle_{u}$ [13]. The calculated curve for σ^{s}_{n2n} in Ref. [30] is lower than the data of our work and the data of Ref. [7]. The differences in the cross-sections for the (n,3n) reactions are still more significant. They are caused by differences both in the fission cross-section evaluations and in the cross-sections for formation of the compound nucleus (Fig. 3(b)).

Now let us examine the process for obtaining $\sigma_{n2n}^{s}(E_n)$ from $\mathcal{G}_{n2n}^{S}(\mathcal{E}_{n}) = \mathcal{G}_{n2n}(\mathcal{E}_{n}) / [1 + z(\mathcal{E}_{n})].$ the calculated dependence $\boldsymbol{\sigma}_{n2n}(E_n)$ as In order to determine $r(E_n)$ the results of the calculations in Ref. [9] were used where the isomeric ratio is obtained by simulating the low-lying level structure of the ²³⁶Np nucleus. The results of Ref. [9] agree well with the data from Ref [8], obtained using a method which is very different from that used in Ref. [9], but differ significantly from the evaluation of $r(E_n)$ in Ref. [30], based essentially on the data from Ref. [5] which are 30% too low and the consequent tendency in Refs [29-31] for $r(E_n)$ to decrease as the energy increases. The evaluation of $\sigma^{s}_{n2n}(E_{n})$ in the present work agrees well with the data from Refs [2-7] whereas the experimental data in Ref. [28], even after the renormalization described above, do not agree well with the data in Refs [2-6] and the present evaluation. When $E_n < 7.5$ MeV the calculated curve is lower than the experimental data in Ref. [7], however, as can be seen from Refs. [9] the excitation of the residual of ²³⁶Np nucleus is here so small that statistical modelling of the gamma transitions becomes scarcely justified, therefore, in this energy range we will determine σ_{n2n}^{s} by interpolation of the values given in Ref. [7] when E_n equals 7.09 and 7.47 Mev. The evaluation for σ_{n2n}^{s} in KEDAK-4 obtained with the assumption of the independence of the isomeric ratio on energy i.e. $r(E_n) = 0.38$ [3] is higher than the data in Refs. [5] and 7] and the use of $r(E_n)$ [9] only intensifies the differences. The evaluation in Ref. [30] is significantly lower than the experimental data [5 and 7] and this fact is associated with the evaluation of $r(E_n)$ (Fig. 4).

COMPARISON OF THE INTEGRAL AND DIFFERENTIAL DATA FOR THE CROSS-SECTION OF THE REACTION ²³⁷Np (n, 2n)²³⁶Np^s

The integral cross-section for the reaction ²³⁷Np (n,2n)²³⁶Np^s averaged over the fission neutron spectrum is directly involved in calculations of the ²³²U accumulation in reactor fuel. This can be represented in the form:

$$< \mathcal{O}_{n2n}^{s} > = \int_{6,8}^{20} \mathcal{O}_{n2n}^{s}(E_{n}) \chi(E_{n}) dE / \int_{0}^{20} \chi(E_{n}) dE_{n} ,$$

where $\chi(E_n)$ is the fission neutron spectrum. In Refs [13 and 14] the values 1.05 and 2.4 mb were obtained respectively for The difference between these values is significantly $< \sigma_{n2n}^{s} >_{u}$. greater than the errors ascribed to them by the authors. Essentially in Ref. [13] the ratio of ²³⁶Pu and ²³⁸Pu concentrations in fuel was measured by comparing the **a** activities and the cross-section $\langle g_{n2n}^{s} \rangle_{u}$ was determined by solving the kinetic equations. In Ref. [14] the value of $\langle \sigma_{n_{2n}} \rangle_{u}$, obtained by averaging the cross-section $\sigma_{n2n}^{s}(E_n)$ [42] over the spectrum [43], was used to evaluate the dependence of accumulation of ²³⁶Pu in the fuel on a result of burnup. If these data are averaged over the fission spectrum [44] which was used to simulate the reactor neutron spectrum in Ref. [13], the cross-section $\langle \sigma_{n2n}^{s} \rangle_{u}$ [14] increases to 2.67 mb. The curves for $\sigma_{n2n}^{s}(E)_{n}$ [10] and [42] and the corresponding data for $\langle \sigma_{n_{2n}} \rangle_{u}$ [13] and [14] are shown in Fig. 5. The result of averaging the dependence for $\sigma_{n2n}^{s}(E_n)$ [30] virtually coincides with the data in Ref. [13]. As has already been pointed out, the curves in Refs [10 and 42] do not agree with the data in Ref. [7] for $\sigma_{n2n}^{s}(E_n)$, however, in Ref. [14], it is shown that $\langle \mathbf{g}^{\mathbf{s}}_{n2n} \rangle_{u} = 2.43$ mb which gives a higher evaluation of the dependence of accumulation of ²³⁶Pu on burnup, i.e. there is a possibility of reducing the value $\langle \sigma_{n_{2n}}^{s} \rangle^{v}$ by 20%. This tendency corresponds to the dependence $\sigma_{n2n}^{s}(E_{n})$ obtained in the present paper, its averaging over the spectra [43 and 44] gives 2.02 and 1.82 mb respectively and taking into account the modification of the calculated dependence $\sigma_{n2n}^{s}(E_n)$ for $n \leq 7.5$ MeV, it gives 2.17 and 1.97 mb.

It is interesting to compare our data on the $\sigma_{n2n}^{s}(E_n)$ dependence with the measurements which used the ²⁵²Cf spontaneous fission neutron spectrum. Using the recommendations in Ref. [45] for the cross-section $\langle \sigma_{n2n}^{s} \rangle_{cf}$ in the ratio $\chi(E_n)$ for ²⁵²Cf we obtain the values 3.23 and 3.47 MeV, taking into account the modification in $\sigma_{n2n}^{s}(E_n)$ for $E_n \leq 7.5$ MeV i.e. values less than $\langle \sigma_{n2n}^{s} \rangle_{cf} = 4.66 \pm 0.47$ mb from Ref. [15]. Thus we can conclude that our data on the $\sigma_{n2n}^{s}(E_n)$ cross-section agree with the recommendations in Ref. [14] but do not agree well with the data in Ref. [15]. In order to obtain agreement with them, the value of $\sigma_{n2n}^{s}(E_n)$ close to the threshold would have to be significantly increased, at least to the level of the curve in Ref. [42], averaging of which over the ²⁵²Cf spontaneous fission neutron spectrum gives 4.24 mb.

Thus, $\langle \sigma_{n_{2n}} \rangle_{u}$ lies in the range of 1.97-2.43 mb. Discrepancies between measurements of the cross-section $\langle \sigma_{n_{2n}} \rangle_{cf}$ [15] and the evaluation for the dependence $\sigma_{n_{2n}}^{s}(E_n)$ in the present work may be caused by measurement errors and inaccurate approximations of the ²⁵²Cf spontaneous fission neutron spectrum [45].

An analysis of the experimental data on the cross-sections and the ^{237}Np (n, 2n) $^{236}Np^{s}$ reactions makes it possible to evaluate the energy dependence of the fission cross-section above the threshold of the (n,nf) reaction. Within the framework of the consistent optical-statistical approach, cross-sections were also obtained for the reactions (n,2n) and (n,3n).

The differences found between the measurements for $\langle \sigma_{n2n}^{\circ} \rangle_{cf}$ [15] on the one hand, and the data in Ref. [14] for $\langle \sigma_{n2n}^{\circ} \rangle_{u}$ and the evaluation in this work on the other hand, leaves the problem of the consistency of integral and differential data on cross-sections for the reaction ²³⁷Np (n,2n) unresolved.

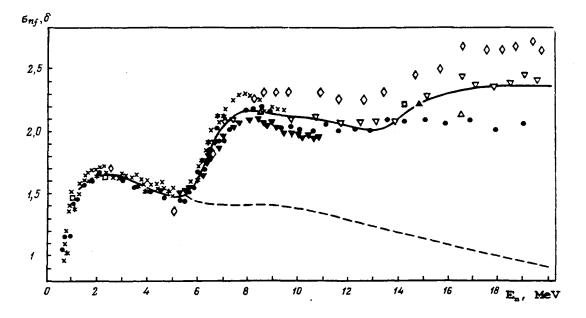


Fig. 1. Neutron fission cross-section for ²³⁷Np. Continuous curve - calculation; broken curve -"first chance" fission cross-section. Experimental data from the following references: $\blacktriangle - [15], \blacktriangle - [17], \blacktriangledown - [18], \blacklozenge - [19], \varkappa - [20],$ $\square - [21], \bigstar - [22], \blacktriangledown - [25], \diamondsuit - [26].$

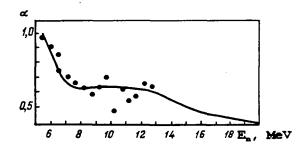


Fig. 2. Dependence of "first chance" contribution on the total fission cross-section for ^{237}Np . Continuous curve - calculation; \bullet - experimental data [39 and 40].

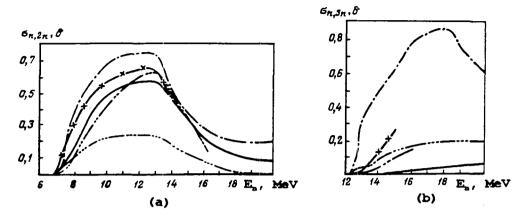


Fig. 3. Cross-section for the reactions: (a) $-\frac{237}{\text{Np}(n,2n)}$, (b) $-\frac{237}{\text{Np}(n,3n)}$. Calculation: this work; x-x-x KEDAK-4 [42];..... ENDL [41]; [30]; ENDF/B-V.

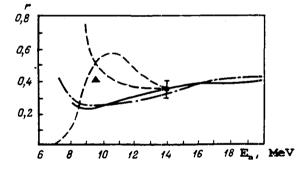


Fig. 4. Energy dependence of the isomeric ratio in the ²³⁷Np(n,2n) reaction. Calculation: [9]; _.__.[8];- - - [30]; Experimental data: ● - [29]; ▲ - value taken from reference [30].

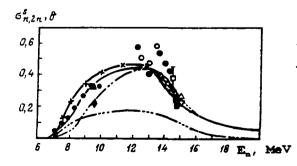


Fig. 5. ²³⁷Np(n,2n) reaction cross-section. Calculation: this work;-x-x- KEDAK-4 [42];ENDL [41];....[30];....ENDF/B-V. Experimental data: □ - [2]; ▲ - [3]; ▼ - [4]; ▲ - [5]; ♦ - renormalized data from Ref. [30]; ■ - [6]; ● - [7]; ○ - [28].

REFERENCES

- [1] MATVEEV, L.V., TSENTER, Eh.M., Uranium-232 and its influence on the radiation situation in the nuclear fuel cycle, Moscow, Ehnergoatomizdat (1985) [in Russian].
- PERKIN, J.L., COLEMAN, R.F., Cross-sections for the (n,2n) reactions of ²³²Th, ²³⁸U, ²³⁷Np with 14 MeV neutrons, [2] J. Nucl. Energ. A/B 14 (1961) 69.
- LANDRUM, J.H., NAGLE, R,J., LINDNER, M., (n,2n) Cross-sections for ²³⁸U and ²³⁷Np in the region of 14 MeV, Phys. [3] Rev. C8 (1970) 1938.
- LINDEKE, K., SPECHT, S., BORN, H.J., Determination of the ²³⁷Np(n,2n)²³⁶Np cross-section at 15 MeV neutron energy, [4] Phys. Rev. C8 (1975) 1507.
- NISHI, T., FUJIWARA, I., IMANISHI, N., Report NEANDC(J)-[5] 42L (1975).
- GROMOVA, E.A., KOVALENKO, S.S., NEMILOV, Yu.A., et al., [6] Measurement of the cross-section for the $^{237}Np(n,2n)$ reaction at neutron energies of 14.8 MeV, At. Ehnerg. 54 (1983) 108 [in Russian].
- KORNILOV, N.V., BARYBA, V.Ya., BALITSKIJ, A.V., et al., [7] Measurement of the cross-sections for the $^{237}Np(n,2n)^{236}Np$ (22.5h) reactions in the 7-10 MeV neutron energy range, At. Ehnerg. 58 (1985) 117 [in Russian].
- 81 GARDNER, D.G., GARDNER, M.A., HOFF, R.W., The necessity of discrete level modelling in isomer ratio calculations for neutron induced reactions on deformed nuclei, Report,
- UCAR-10062-83/1 (1981) 51. IGNATYUK, A.V., KORNILOV, N.V., MASLOV, V.M., PASHCHENKO, A.B., Isomer ratio and ²³⁷Np(n,2n) reaction cross-section, [9] in Proc. of the 15th Intern. Symp. on Physics of Fission, Gaussig, 1985).
- KINSEY, R., Report ENDF-201, Brookhaven (1979). [10]
- [11] KEDAK-4, The German Nuclear Data Library (1983).
- PEARLSTEIN, S., Analysis of (n,2n) cross-sections for [12]medium and heavy mass nuclei, Nucl. Sci. Eng. 23 (1965) 238.
- [13] PAULSEN, C.K., HENNELY, E.J., Cross-section measurement of plutonium-236 formation in plutonium-238 by ²³⁷Np(n,2n) reaction, Nucl. Sci. Eng. 55 (1974) 24.
- WIESE, H.W., FISHER, U., GOEL, B., Analysis of neutron [14]cross-sections for the formation of ²³⁶Pu and ^{58,60}Co in
- both thermal and fast reactors, (in Proc. Nucl. Data for Sci. and Techn., Antwerp, 1982), Holland (1983) 202. GROMOVA, E.A., KOVALENKO, S.S., NEMILOV, Yu.A., et al., Cross-section for the ²³⁷Np(n,2n)²³⁶Np (22.5h) reaction caused by neutrons from spontaneous fission of ²⁵²Cf, At. [15] Ehnerg. 60 (1986) 68 [in Russian].
- DUSHIN, V.N., FOMICHEV, A.V., KOVALENKO, S.S., et al., [16] Statistical analysis of fission cross-section measurements on ^{233,235,238}U, ²³⁷Np, ^{239,242}Pu at neutron energies of 2.6, 8.5 and 14.7 MeV, (in Proc. IAEA Cons. Meeting on the ²³⁵U Fast Neutron Fission Cross-section and the ²⁵²Cf Fission Neutron Spectrum, Smolenice 1983), Report INDC(NDS)-146/L (1983) 53.

- [17] GOVERDOVSKIJ, A.A., GORDYUSHIN, A.K., KUZ'MINOV, B.D. et al., Measurement of fission cross-section ratios for ²³⁷Np and ²³⁵U by the isotope impurity method, Voprosy atomnoj nauki i tekhniki. Ser. Yadernye Konstanty 3 57 (1985) 13 [in Russian].
- [18] GOVERDOVSKIJ, A.A., GORDYUSHIN, A.K., KUS'MINOV, B.D. et al., Measurement of the fission cross-section ratio for ²³⁷Np and ²³⁵U by neutrons in the 4-11 MeV energy range, At. Ehnerg. 58 (1985) 137 [in Russian].
- [19] BEHRENS, J.W., BROWNE, J.C., WALDEN, J.C., Measurement of the neutron-induced fission cross-section of ²³⁷Np relative to ²³⁵U from 20 keV to 30 MeV, Nucl. Sci. Eng. 80 (1982) 393.
- [20] MEADOWS, J.W., The fission cross-section of ²³⁷Np relative to ²³⁵U from 0.1 to 9.4 MeV, Nucl. Sci. Eng. 85 (1983) 271.
- [21] WHITE, P.H., WARNER, G.P., The fission cross-sections of ^{233,234,236,238}U, ²³⁷Np, ^{239,240,241}Pu relative to that of ²³⁵U for neutrons in the energy range 1-14 MeV, J. Nucl. Energy 21 (1967) 671.
- [22] KUPRIYANOV, V.M., FURSOV, B.I., IVANOV, V.I., SMIRENKIN, G.N., Measurement of the fission cross-section ratios ²³⁷Np/²³⁹Pu and ²⁴¹Am/²³⁹Pu in the 0.13-7.0 MeV neutron energy range, At. Ehnerg. 45 (1978) 440 [in Russian].
- [23] ANTŠIPOV, G.V., KOH'SHIN, V.A., SUKHOVITSKIJ, E.Sh., "Nuclear Constants for Plutonium Isotopes", Minsk, Nauka i Tekhnika (1982) [in Russian].
- [24] KUPRIYANOV, V.M., SMIRENKIN, G.I., FURSOV, B.I., Systematics of neutron cross-sections and other characteristics of the fission probability of
- transuranium nuclei, Yad. Fiz 39 (1984) 281 [in Russian].
 [25] PANKRATOV, V.M., VLASOV, N.A., RYBAKOV, B.V., Fission
 cross-sections for ²³⁵Th, ²³⁵U and ²³⁸U by neutrons with
 energies of 10-22 MeV, At. Ehnerg. 9 (1960) 399 [in
 Russian].
- [26] PANKRATOV, V.M., Fission cross-section for ²³²Th, ³³³U, ²³⁵U, ²³⁷Np and ²³⁸U by neutrons in the 5-37 MeV energy range, At, Ehnerg, 14 (1963) 177 [in Russian].
- At. Ehnerg. 14 (1963) 177 [in Russian]. [27] KALININ, S.P., PANKRATOV, V.M., (in Proc. 2nd Int. Conf. on the Peaceful Uses of Atomic Energy, Geneva 1958), Moscow, Atomizdat 1 (1959) 387.
- [28] ANDREEV, M.F., SEROV, V.I., Evaluation of the cross-section for the (n,2n) reaction for heavy nuclei based on the results of studies with charged particles, (In "Nejtronnaya fizika: Proc. 5th All-Union Conf. on Neutron Physics", Kiev 15-19 September 1980), Moscow, TSNII atominform 3 (1980) 301 [in Russian].
- [29] MEYERS, W.A., LINDNER, M., NEWBURY, R.S., The isomer ratio ²³⁶Np(1)/²³⁶Np(s) in the reaction ²³⁷Np(n,2n)²³⁶Np from neutrons produced in thermonuclear devices, J. Inorg. Nucl. Chem. 37 (1975) 637.
- [30] FORT, E., DERRIEN, H., DOAT, J.P., Evaluation des sections efficaces neutroniques de ²³⁷Np entre 5 MeV et 16 MeV. Etude particuliere pour l'application aux calculus de ²³⁶Pu, see Ref. [14] p. 673.

- [31] HYUZENGA, D.R., VANDENBOSH, R., "Nuclear Fission" in Yadernye Reaktsii, Moscow, Atomizdat 2 (1964) 51 [in Russian].
- [32] UHL, M., STROHMAYER, B., Report IRK 76/01, Vienna (1976).
 [33] LAGRANGE, Ch., Results of coupled channels calculations for neutron cross-sections of a set of actinide nuclei, Report INDC(FR)-56/L (1982).
- [34] ANTSIPOV, G.V., KON'SHIN, V.A., MASLOV, V.M., Level density and radiation widths of transactinides, Voprosy atomnoj nauki i tekhnika. Ser. Yadernye Konstanty 3 (1985) 25 [in Russian].
- [35] BOR, O., MOTTEL'SON, B., "Structure of the Nucleus", Moscow, Mir 2 (1977) [in Russian].
- [36] IGNATYUK, A.V., ISTEKOV, K.K., SMIRENKIN, G.N., Role of collective effects in level density systematics, Yad. Fiz. 29 (1979) 875.
- [37] IGNATYUK, A.V., KLEPATSKIJ, A.B., MASLOV, V.M., et al., Analysis of fission cross-sections of uranium and plutonium isotopes by neutrons in the first plateau region", Yad. Fiz. 42 (1985) 569.
- [38] GRUDZEVICH, O.T., IGNATYUK, A.V., MASLOV, V.M. et al., Consistent description of the (n,f) and (n,xn) for transactinides, (in "Nejtronnaya Fizika", Proceedings of the Sixth All-Union Conference on Neutron Physics", Kiev 2-6 October 1983), Moscow, TSNII Atominform 2 (1983) 318 [in Russian].

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