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

V.M. Maslov

ABSTRACT

Experimental and evaluated cross-section data on fission, (n,3n) and (n2n) reactions leading to the short-lived state of the ²³⁶Np 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 ²³⁷Np(n,2n)²³⁶Np^s 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)²³⁶Np¹ reaction, there are no other data apart from the isomeric ratio, r, of the $^{236}\text{Np}^1$ and ^{236}Np yields for 14 MeV neutrons. Therefore, the existing evaluations of the cross-section $\sigma^s{}_{\text{n2n}}$ are based on model calculations of the $\sigma{}_{\text{n2n}}$ cross-section, and the $\sigma^s{}_{\text{n2n}}$ cross-section is determined from σ_{n2n} /(1 + r) on the assumption that r does not depend on th 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 σ_{n2n} have the disadvantage that the fission cross-section σ_{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

 $- 27 -$

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)²³⁶Np^s reaction and on the isomeric ratio.

The absolute value of $\sigma^s_{\texttt{nzn}}$ 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 ${<} \sigma^{s}{}_{n2n}$. Therefore, it is also necessary to analyse the consistency between the differential and integral data on the cross-section for the ²³⁷Np(n,2n)²³⁶Np^s 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 $^{237}_{2}$ Np 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($\sigma_{\text{nf}}^7\sigma_{\text{nf}}^5$) [15-21] or ^{239}Pu ($\sigma_{\text{nf}}^7/\sigma_{\text{nf}}^9$ [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 ${\bm \sigma}_{\rm nf}^{\rm 7}/{\bm \sigma}_{\rm nf}^{\rm 5}$ in Ref. [20], which was based on a comparison of the **α**-activities of the ²³⁷Np and ²³⁵U shell: 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 ${\sigma_{\mathrm{nf}}^\flat}/{\sigma_{\mathrm{nf}}^\mathfrak{s}}$ ratio, data on the cross-section $\sigma_{\text{nf}}^{\text{s}}$ 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 $\sigma_{\rm nf}^{\rm s}$ [10]. In the latter case the discrepancy is approximately equal to the value of $\sigma_{n_{2n}}^s$ for $E_n = 14.7$ MeV. The situation is complicated by the fact that the energy dependence of the cross-section ${\bm \sigma^7}_{\textbf{nf}}$ 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 ${\sigma_{\mathsf{nf}}^\tau}/{\sigma_{\mathsf{nf}}^\mathsf{s}}$ ratio and with the fact that the evaluation of $\sigma_{\rm nf}^{\rm s}$ 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 $\sigma_{\mathsf{nf}}^\prime$, 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 ${\bm \sigma}^7_{\rm nf}$ ($\rm E_n$ = 3.4 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 $E_n = 14.7$ 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.

Thus, as reference values for $E_n < 14$ 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 ²³⁷Np(n,2n)²³⁶Np² 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 ²⁷Al(n, α) reaction and was also monitored [7] with respect to the 238 U(n,f) and 238 U(n,2n) reactions. In Refs [4] and 6], one of the methods used to determine the flux involved evaluating the accumulation of ^{97}Zr nuclei during fission of $^{237}\mathrm{Np}$ However the use of the fission cross-section, which was 10% higher than the data in Ref. [16], resulted in an overestimation of the $\sigma_{n,zn}$ 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 236 U [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 $\sigma^2_{\rm n,\,2n}$ cross-section may be derived from the measurements results of the cross-sections for the 235 U(t,2n) 236 Np^s and 236 U(d,2n) 236 Np 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 $[28]$: $_{\text{nf}}/\sigma_{\text{t(d)}f}$ $\sigma_{\text{t(d)}2n}^{\text{s}}$ (1)

In Ref. [28], data from Ref. [26] were used to determine the $\sigma'_{\rm nf}$ cross-section; as a result, $\sigma^{\mathsf{s}}_{\mathsf{n},\mathsf{2n}}$ was over-valued. In this wor l the calculated $\| \bm{\sigma}_{\sf nf} \|$ cross-section was used to calculate $\bm{\sigma}_{\sf n,2n}^{\sf s}$ 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.

Isomeric ratio in the ²³⁷Np(n,2n) reaction. 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 $^{237}\mathrm{Np}$ ($\pmb{\gamma},$ 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 236 Np compound nucleus with spin $J = 1$ are approximately seven times more frequent than for states with spin $J = 6$. This means that $r(E_n \approx 19 \text{ MeV}) \approx 0.14$. 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 U, J) was calculated in the following way. For excitation energies U, which is smaller than energy $U_{n(f)} = (10.7 - m\Delta_{n(f)})$ - 0.028A 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, $p_{n(f)}(U,J)$ is determined using the constant temperature model [34]. In the neutron channel

$$
\rho_n = \frac{1}{T_n} \exp\left(\frac{U + m\Delta_n}{T_n}\right) \frac{2J + 1}{2\sigma^2} \exp\left[-\frac{J(J+1)}{2\sigma^2}\right],\tag{2}
$$

where $\Delta_n = 12/\sqrt{A}$ MeV; $T_n = 0.385$ MeV. The parameter for spin dependence of σ^2 at excitation energies $U \leq U_{x}$, 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 σ_{n}^{2} = 0.156A - 26.76. For $U > U_x$, σ^2 is determined by the linear extrapolation between σ_n and σ^2 F t(U_n). Here F₁ 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:

$$
\rho_f(U, J) = \frac{R}{T_f} \exp\left(\frac{U + m\Delta_f + \delta_f}{T_f}\right) \frac{1}{2\sigma_f^2} \sum_{K=-J}^{J} \exp\left(-K^2/2K_0^2\right) \exp\left[-\frac{J(J+1)}{2\sigma_{Lf}^2}\right].
$$

Here K²₀ = (1/6²₁₄ - 1/6²₁₄), where 6^{2}_{ff} - F_{ff} t (F_{ff} (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 $\rho_f = \frac{\sigma^2}{\sigma^2} \omega_f(U) \sqrt{2\pi} \sigma_{\theta}$. The δ_t parameter is determined from the continuity condition of the level density $p_f(U)$ for the excitation energy $U = U_f$ (the corresponding parameter in the neutron channel equals zero): $(1/T_f e^{\frac{m}{2}})exp[(U_f + m \Delta_f + \delta_f)/T_f] = \rho_f(U_f)$.

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, $R = 2\sqrt{2\pi} \cdot \delta_{\parallel f}$; for the external hump only the mirror symmetry is violated and $R = 2$ [35]. The density of the internal states $\omega_{n(r)}(U)$ and the spin dependence parameters $\sigma^2{}_{11}$ and σ^2 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 δ_f for the internal A and external B humps are: T_{f}^{A} = 0.38 Mev, T_{f}^{B} = 0.39 Mev, $\delta^A_f = 0.001$, $\delta^B_f + 0.24$.

For excitation energies $U > U_{n(f)}$ $\rho_{n(f)}(U, J)$ 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/A³, 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 σ_{nf}^1 to the observed fission cross-section σ_{nf} , i.e. $\alpha = \sigma_{\text{nf}}^1/\sigma_{\text{nf}}$ can be compared with the data for α 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 α . The cross-section $\sigma_{\rm n2n}$ = $\sigma^{\rm n}_{\rm n2n}$ + $\sigma^{\rm s}_{\rm n2n}$ 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 σ_{n2n} 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 $\sigma_{\texttt{n2n}}^{s}$ 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 σ_{n2n} =1.35 $\sigma_{\text{n2n}}^{\text{s}}$ and $\sigma_{\text{n2n}}^{\text{s}}$ by normalization of the dependence $\sigma_{\text{n2n}}^{\text{s}}(E_{\text{n}})$ [12] on the integral data for $\langle \sigma_{\text{n2n}}^{\text{s}} \rangle_y$ [13]. The calculated curve for $\sigma_{\text{n2n}}^{\text{s}}$ 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 the calculated dependence $\sigma_{n2n}(E_n)$ as $6_{2n}^S(E_n) = 6_{n2n}(E_n)/[1 + \tau(E_n)].$ In order to determine $r(E_n)$ the results of the calculations in Ref. [9] were used where the isomeric ratio is obtained bysimulating 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 $\sigma_{\texttt{n2n}}^{\texttt{s}}(\mathrm{E}_{\texttt{n}})$ 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 $\sigma_{\texttt{nzn}}^{\texttt{s}}$ by interpolation of the values given in Ref. [7] when E_n equals 7.09 and 7.47 Mev. The evaluation for $\sigma_{_{n2n}}^{s}$ 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 $^{237}\mathrm{Np}$ (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:

20 .20 6,8 ⁷ 0.

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 $\langle \sigma_{\text{max}}^s \rangle_u$. The difference between these values is significantly 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 α activities and the cross-section ${<\sigma^s}_{\rm n2n}{>_{\rm u}}$ was determined by solving the kinetic equations. In Ref. [14] the value of $<\sigma^{s}{}_{\mathsf{n2n}}>_{\mathsf{u}},$ 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 $\langle \sigma_{n2n}^3 \rangle_v$ [14] increases to 2.67 mb. The curves for $\sigma_{\texttt{n2n}}^s(\text{E})$ _n [10] and [42] and the corresponding data for $<$ σ ³_{n2n} $>$ _v [13] and [14] are shown in Fig. 5. The result of averaging the dependence for $\sigma_{n_{2n}}^s(E_n)$ [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 $<$ σ^s _{n2n} $>$ _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 ${<}{{\sigma^s}_\mathsf{n2n}} >^\mathtt{U}$ by 20%. This tendency corresponds to the dependence $\sigma_{\texttt{n2n}}^{\texttt{s}}(\texttt{E}_\texttt{n})$ 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 $\sigma^s_{n2n}(E_n)$ dependence with the measurements which used the ²⁵²Cf spontaneous fission neutron spectrum. Using the recommendations in Ref. [45] for the cross-section $<\!\mathfrak{g}^{\mathsf{s}}_{\text{n2n}}\!\!>_{\mathsf{c}\mathsf{f}}$ in the ratio $\pmb{\chi}\,(\mathtt{E}_{\mathsf{n}})$ for $^{252}\!\mathbb{C}\,\mathsf{f}$ we obtain the values 3.23 and 3.47 MeV, taking into account the modification in $\sigma_{n2n}^s(E_n)$ for $E_n \le 7.5$ MeV i.e. values less than $<$ σ_{n2n} $>$ $_{cf}$ = 4.66 \pm 0.47 mb from Ref. [15].

Thus we can conclude that our data on the $\sigma^s_{n2n}(E_n)$ 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 $\sigma^s_{\rm n2n}(\rm E_n)$ 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, $\langle \sigma^s_{n2n} \rangle_{\upsilon}$ lies in the range of 1.97-2.43 mb. Discrepancies between measurements of the cross-section ${<} \sigma^s_{\sf n2n} >_{\sf ct}$ [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 ²³⁷Np (n, 2n)²³⁶Np^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 $\langle \sigma_{\text{n2n}}^{\text{s}} \rangle_{\text{cf}}$ [15] on the one hand, and the data in Ref. [14] for ${<} \sigma^{s}{}_{n2n}$ $>$ _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: Δ - [15], Δ - [17], \triangledown - [18], \bigoplus - [19], $x - [20]$, \Box - [21], \star - [22], \triangledown - [25], \lozenge - [26].

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

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

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

Fig. 5. ²³⁷Np(n,2n) reaction cross-section. Calculation: this work;-x-x- KEDAK-4 [42]; $ENDL [41]; \ldots \ldots [30]; \ldots \ldots \ldots \text{ENDF/B-V}.$ Experimental data: \square - $\{2\}$; \blacktriangle - $\{3\}$; ∇ - $\{4\}$; \blacktriangle - [5]; \blacklozenge - renormalized data from Ref. [30]; \blacksquare - [6]; \spadesuit - [7]; \sf{O} - [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].
- [2] PERKIN, J.L., COLEMAN, R.F., Cross-sections for the (n,2n) reactions of ²³²Th, ²³⁸U, ²³⁷Np with 14 MeV neutrons J. Nucl. Energ. A/B 14 (1961) 69.
- [3] LANDRUM, J.H., NAGLE, R,J., LINDNER, M., (n,2n) Crosssections for ²³⁸U and ²³⁷Np in the region of 14 MeV, Phys Rev. C8 (1970) 1938.
- [4] LINDEKE, K., SPECHT, S., BORN, H.J., Determination of the ²³⁷Np(n,2n)²³⁶Np cross-section at 15 MeV neutron energy, Phys. Rev. C8 (1975) 1507.
- [5] NISHI, T., FUJIWARA, I., IMANISHI, N., Report NEANDC(J)- 42L (1975).
- [6] GROMOVA, E.A., KOVALENKO, S.S., NEMILOV, Yu.A., et al., Measurement of the cross-section for the ²³⁷Np(n, 2n) reaction at neutron energies of 14.8 MeV, At. Ehnerg. 54 (1983) 108 [in Russian] .
- [7] KORNILOV, N.V., BARYBA, V.Ya., BALITSKIJ, A.V., et al., Measurement of the cross-sections for the ²³⁷Np(n,2n)²³⁶Np (22.5h) reactions in the 7-10 MeV neutron energy range, At. Ehnerg. 58 (1985) 117 [in Russian].
- 8] 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.
- [9] IGNATYUK, A.V., KORNILOV, N.V., MASLOV, V.M., PASHCHENKO, A.B., Isomer ratio and ²³⁷Np(n, 2n) reaction cross-section, in Proc. of the 15th Intern. Symp. on Physics of Fission, Gaussig, 1985).
- [10] KINSEY, R., Report ENDF-201, Brookhaven (1979).
[11] KEDAK-4, The German Nuclear Data Library (1983)
- KEDAK-4, The German Nuclear Data Library (1983).
- [12] PEARLSTEIN, S., Analysis of (n,2n) cross-sections for 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.
- [14] WIESE, H.W., FISHER, U., GOEL, B., Analysis of neutron cross-sections for the formation of 236Pu and 58,60Co in both thermal and fast reactors, (in Proc. Nucl. Data for Sci. and Techn., Antwerp, 1982), Holland (1983) 202.
- [15] 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. Ehnerg. 60 (1986) 68 [in Russian].
- [16] DUSHIN, V.N., FOMICHEV, A.V., KOVALENKO, S.S., et al., Statistical analysis of fission cross-section measurements on ^{233,235,238}U, ²³⁷Np, ^{239,242}Pu at neutro 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, 237 Np, 239,240,241 Pu relative to that of 235 U 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 237 Np/²³⁹Pu and ²⁴¹Am/²³⁹Pu in the 0.13-7.0 MeV neutron energy range, At. Ehnerg. 45 (1978) 440 [in Russian].
- [23] ANTSIPOV, 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].
- [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 Ail-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 ratio²³⁶Np(1)/²³⁶Np(s) in the reaction²³⁷Np(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 1'application aux calculus de 236Pu, see Ref. [14] p. 673.
- HYUZENGA, D.R., VANDENBOSH, R., "Nuclear Fission" in $[31]$ Yadernye Reaktsii, Moscow, Atomizdat 2 (1964) 51 [in Russian].
- [32] [33] UHL, M., STROHMAYER, B., Report IRK 76/01, Vienna (1976). 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, 0., 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 Ail-Union Conference on Neutron Physics", Kiev 2-6 October 1983), Moscow, TSNII Atominform 2 (1983) 318 [in Russian].

Submitted for publication 17 July 1986