Evaluation of the $^{93}\text{Nb}(n,\gamma)$ Reaction Cross-Section

Konstantin Zolotarev$^1$ and Sergei Badikov$^2$

$^1$ Institute of Physics and Power Engineering, 249033 Obninsk, Russia
$^2$ National Research Nuclear University “MePHI”, 115409 Moscow, Russia

Abstract. New evaluation of the $^{93}\text{Nb}(n,\gamma)^{94}\text{Nb}$ reaction cross-section important for retrospective reactor dosimetry was carried out. At neutron energies below 7.5 keV the evaluation is based on the experimental data. The results of measurements extracted from the EXFOR library were corrected (when necessary) to new recommended values of monitor reaction cross-sections and decay data. The resonance analysis of the $^{93}\text{Nb}(n,\gamma)$ and the $^{93}\text{Nb}(n,\text{tot})$ reaction cross-sections was carried out within the Reich-Moore formalism. 48 new resonances were identified as compared to the Mughabghab systematics. In the energy range from 7.5 keV to 20 MeV the evaluation is based on the experimental data and theoretical model calculations. The recommended cross-sections and their covariances in this energy range were evaluated by an approximation of the experimental and calculated values with the PADE2 code. The new evaluation provides, essentially, better agreement with the experimental data and the recommended value of the resonance integral compared to other evaluations.

1. Introduction

Operation time of a nuclear power plant with Pressurized Water Reactor (PWR) depends mainly on residual lifetime of a reactor pressure vessel. The residual lifetime is caused by the degree of the embrittlement of the constructed materials that depends on the neutron fluence on the wall of the reactor.

The direct experimental assessment of the neutron fluence for the entire period of the reactor operation can be based only on the activity measurements of the samples extracted from the inner wall of the reactor vessel. The samples must contain nuclides with long half-lives to fix dependence of the neutron fluence on the time. From this point of view, some nuclides of chromium, iron, nickel, cobalt and niobium are of interest.

Reliable activation cross-section evaluations are necessary for getting accurate estimates of the neutron fluence on the wall of reactor. For this reason the $^{93}\text{Nb}(n,\gamma)^{94}\text{Nb}$ reaction is of special interest since the niobium is a construction material and the cross-section covers all the energy range important for reactor applications. Recently it was shown that a very small amount of Nb-94 can be detected in the chips of the RPV wall [1].

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2. Status of the $^{93}$Nb(n,$\gamma$) Reaction Cross-section Evaluations

At present there are 3 cross-section evaluations [2–4] and the recommended data by Mughabghab [5]. At the thermal point, the evaluated cross-sections agree with each other and the recommended data (1.15 ± 0.5 b) within declared uncertainties. In the resolved resonance region the evaluated data is somewhat contradictory. The total numbers of the resonances identified in the ENDF/B-VII.1, JENDL-4.0, ROSFOND evaluations and Mughabghab’s systematics are similar (194, 201, 199 and 202 resonances, correspondingly, see Table 1). At the same time, there exists a considerable disagreement between the numbers of the s- and p-resonances. In particular, authors of the ENDF/B-VII.1, JENDL-4.0, ROSFOND evaluations and Mughabghab presented the parameters of 148, 139, 99, 66 s-resonances and 46, 62, 100, 136 p-resonances, respectively (the difference is more than 2 times). As follows from the ENDF/B-VII.1 and JENDL-4.0 evaluations, the density of the p-resonances decreases with increasing the neutron energy $E$. This fact looks suspicious because the average neutron width of the p-resonances is proportional to $E^{3/2}$. In crude approximation, the density of levels $\rho$ is proportional to $2^{J+1}$. For the ENDF/B-VII.1 evaluated data, this is approximately true for all the spin systems whereas for the JENDL-4.0 and ROSFOND evaluations this is not the case.

The IRDFF-v.1.02 evaluation [6] of the $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction cross-section was taken from the ENDF/B-VII.1 library. It is based on the results of measurements made before 1986. Since that time 9 new experiments were carried out. In the resolved resonance region the IRDFF-v.1.02 evaluated cross-section is parameterized with the single level Breit-Wigner formulae – least accurate approximation of the R-matrix theory. Finally, the IRDFF-v.1.02 evaluation has a background cross-section given in File 3. These facts justify a renewal of the IRDFF-v.1.02 evaluation.

3. The Experimental Data Base

Natural niobium is a monoisotope element. The residual nucleus of the $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction has a half-life 20300 ± 1600 years. $^{94}$Nb decays by $\beta^-$ – emission (branching ratio 100%) to the ground and excited states of $^{94}$Mo, which are depopulated by gamma transitions and internal conversion. Recommended decay data was taken from [7].

The experimental data base for the evaluation of the $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction cross-section was compiled on the basis of information extracted from the EXFOR library [8]. A deadline for a retrieval of the data was May 30, 2013. The results of 33 experiments [9–41] have been analysed. A part of the experimental data ([13–15], [17, 18], [21], [23], [26], [28–30], [35–38]) was corrected to new recommended monitor reaction cross-sections and decay data. For some data, special corrections were introduced. In particular, Gibbons et al. [17] have used the $^{nat}$In(n,$\gamma$) reaction as a monitor in relative measurements. The new recommended monitor cross sections for this experiment was calculated as a sum of the ENDF/B-VII.1 evaluations for the $^{113}$In(n,$\gamma$) and $^{115}$In(n,$\gamma$) reaction cross-sections. Results of the original relative measurements of Stavisskij and Shapar’ [18] have been normalized by the authors to the measured cross section value of 65 mb at 0.4 MeV. For this reason these experimental data were renormalized to the preliminary evaluated cross section value (55.9 ± 2.5 mb) at a neutron energy of 0.4 MeV. Authors have not provided any information on the resolution broadening in the experiment by Drindak et al. [41]. So, these experimental data were not included into the database prepared for the analysis.

There is only one measurement of the $^{93}$Nb radiative capture cross section above 3 MeV [25]. Therefore the experimental data included in the data base were supplemented by the results of theoretical model calculations in the energy range 3–20 MeV.

The $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction cross-section has an evident resonance structure at low energies. So, for a proper description of the $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction cross-section, a joint analysis of the experimental data for the $^{93}$Nb(n,tot), $^{93}$Nb(n,$\gamma$)$^{94}$Nb and $^{93}$Nb(n,el) cross-sections must be carried out. The results
of the total neutron cross-section measurements on $^{93}$Nb were extracted from publications [41–49]. The high resolution measurements were described in [42, 44]. Neutron elastic cross-sections in the resonance region were measured only for the one experiment cited in reference [50].

4. Basic Formulae

Theory of nuclear reactions in the resolved resonance region provides a convenient instrument for a description of the reaction cross-sections. The basic concepts of the nuclear reactions theory are a collision matrix $U_{cc'}$ and a reaction channel $c$. The value $|U_{cc'}|^2$ defines a probability of a transition of the system from channel $c'$ to channel $c$. The channel $c$ is defined by a set of parameters $\{x, J, l, s\}$, where $x$ is a partition of the system, $J, l, s$ – the total angular momentum, orbital momentum and spin of the system in units $\hbar$. The collision matrix can be presented in terms of the R-matrix or (identically) the level matrix. In the reactor applications different approximations (formalisms) of the R-matrix (level matrix) are used.

The resonance analysis of the $^{93}$Nb(n,tot), $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction cross-section was carried out within the framework of Reich-Moore formalism of the R-matrix theory. A key point of the Reich-Moore formalism is neglecting contributions from photon channels into off-diagonal elements of the level matrix [51]. In this case the collision matrix $U^{J}_{nc}(c'=n)$ can be presented in following way [51, 52]

$$U^{J}_{nc} = e^{-i(\rho_{nc} + \phi_{nc})} \{2[(I - K)^{-1}]_{nc} - \delta_{nc}\}. \quad (1)$$

Here the matrix $(I - K)$ has a form

$$(I - K)_{nc} = \delta_{nc} - \frac{i}{2} \sum_{\lambda} \frac{\Gamma_{n\lambda}^{1/2} \Gamma_{c\lambda}^{1/2}}{E_{\lambda} - E - i\Gamma_{\gamma\lambda}^{1/2}/2} \quad (2)$$

where $\Gamma_{n\lambda}$ and $\Gamma_{c\lambda}$ – the partial widths in channels $n$ and $c$, $\Gamma_{\gamma\lambda}$ – radiation width, $E_{n\lambda}$ – resonance energy, $\phi_{nc}$– hard-sphere phases. It is convenient to define the following matrix

$$\rho_{nc} = \delta_{nc} - [(I - K)^{-1}]_{nc}. \quad (3)$$

Then the expressions for the total $\sigma_{tot}$, elastic scattering $\sigma_{nn}$ and absorption cross-sections $\sigma_{abs}$ can be written in following way

$$\sigma_{tot} = \frac{2\pi}{k^2} \sum_{lsJ} g(J)[(1 - \cos 2\phi_{l}) + 2Re(\rho_{nn}e^{-2i\phi_{l}})], \quad (4)$$

$$\sigma_{nn} = \frac{\pi}{k^2} \sum_{lsJ} g(J)[2 - 2\cos 2\phi_{l} + 4Re(\rho_{nn}e^{-2i\phi_{l}}) - 4Re(\rho_{nn}) + 4|\rho_{nn}|^2] \quad (5)$$

$$\sigma_{abs} = \frac{4\pi}{k^2} \sum_{lsJ} g(J)[Re(\rho_{nn}) - |\rho_{nn}|^2]. \quad (6)$$

5. Results of the Evaluation

The results of measurements of the $^{93}$Nb(n,$\gamma$)$^{94}$Nb reaction cross section at the thermal point are described in papers [9–11], [14], [22] and [24]. Since 1969 no new experiments have been carried out. For this reason we adopted the Mughabghab’s evaluation of the thermal radiative capture cross-section $(1.15 \pm 0.05 \text{ b})$ [5].
Figure 1. The evaluated $^{93}$Nb(n,tot) reaction cross-section compared to the experimental and ENDF/B-VII.1 (IRDFF) data in the energy range 6–7.5 keV.
Table 1. The numbers of the $s$- and $p$-resonances and the values of resonance integral.

<table>
<thead>
<tr>
<th>Evaluations</th>
<th>Number of resonances</th>
<th>Number of $s$-resonances</th>
<th>Number of $p$-resonances</th>
<th>Value of resonance integral, $b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>ENDF/B-VII.1</td>
<td>194</td>
<td>148</td>
<td>46</td>
<td>9.75</td>
</tr>
<tr>
<td>ROSFOND</td>
<td>199</td>
<td>99</td>
<td>100</td>
<td>9.70</td>
</tr>
<tr>
<td>JENDL-4.0</td>
<td>201</td>
<td>140</td>
<td>61</td>
<td>8.81</td>
</tr>
<tr>
<td>Mughabghab</td>
<td>202</td>
<td>66</td>
<td>136</td>
<td>$8.3 \pm 0.4$</td>
</tr>
<tr>
<td>present</td>
<td>250</td>
<td>70</td>
<td>180</td>
<td>8.6</td>
</tr>
</tbody>
</table>

Of the 4 evaluations of the resonance parameters considered in Section 2 the Mughabghab evaluation looks as the most conservative. Mughabghab presented information on the characteristics of 202 resonances. Parameters of 40 resonances are completely identified. Values of $l$ or $J\pi$ for 36 resonances are defined as questionable. For remaining part of the resonances, the parameters are given in part. So, it would be reasonable to take the resonance parameters of this systematics as a starting point in calculations or even incorporate a part of these parameters in the new evaluation. To simplify the calculations a part of the resonance parameters evaluated by Mughabghab was fixed in fitting procedure. The parameters of the 1-st $p$-resonance were adopted from the work [36] to get a consistency with the evaluated thermal radiative capture cross-section. At the first step of calculations the experimental data was approximated by the single level Breit-Wigner formulae. These parameters were taken as an initial approximation. The resulting resonance parameters were calculated with the non-linear least squares method by using an iterative procedure

$$\tilde{\theta}_k = \tilde{\theta}_{k-1} + (X_{k-1}^T V^{-1} X_{k-1})^{-1} X_{k-1}^T V^{-1} (\tilde{y} - f(E, \tilde{\theta}_{k-1})).$$

$$W_k = W_{k-1} + (X_{k-1}^T V^{-1} X_{k-1})^{-1}.$$  

Here $\tilde{y}$ – the vector of the measurements, $f(E, \tilde{\theta}_{k-1})$ – the total and partial cross-sections within the Reich-Moore formalism, $\tilde{\theta}_k$ and $W_k$ – the vector and covariance matrix of the resonance parameters at the $k$-th iteration, $V$ – the covariance matrix of the experimental errors, $X$ – the matrix of the sensitivity coefficients of the cross-sections relative the resonance parameters. 48 new resonance were identified compared to the Mughabghab’s systematics [5]. In Fig. 1 the evaluated $^{93}\text{Nb}(n,tot)$ reaction cross-section is presented in comparison with the ENDF/B-VII.1 data. As seen from Fig. 1 the new evaluation provides an essentially better agreement with the experimental data than the ENDF/B-VII.1 one. It should also be noted that the new evaluation ensures a value of the resonance integral consistent with recommended value of Mughabghab. (see Table 1).

Keeping in mind the approximate dependence of the level density on $J$ ($\rho \sim (2J+1)$) and the relation between the numbers of the $s$- and $p$-resonances for states with $J = 4, 5$ and $J = 3, 4, 5, 6$, respectively, we conclude that the number of $p$-resonances must be about 2 times more than the number of $s$-resonances. As follows from the Table 1 the present and Mughabghab evaluations agree with this conclusion better than any other ones.

In the energy range 7.5 keV – 20 MeV the evaluation is based on the experimental data and theoretical model calculations carried out with a modified version [53] of the code GNASH [54]. The code allows one to estimate the contributions of the statistical, preequilibrium and direct reactions into the cross-sections and angular distributions. The modified version of the code takes the width fluctuation corrections into account. The optical model was used for a calculation of the neutron and charge-particle penetrabilities [55, 56]. The parameters of the discrete levels for $^{93}\text{Nb}$ and all residual nuclei were taken from [57]. Unknown branching ratios were estimated on the basis of statistical calculations of the possible E1, E2 and M1 gamma-ray transitions. Intensities of such transitions were calculated from the radiation strength functions recommended in [58]. The level densities in the continuum were represented
Figure 2. The evaluated $^{93}$Nb(n,$\gamma$) reaction cross-section compared to the experimental data and the ENDF/B-VII.1 (IRDFF) evaluation.
by means of the Gilbert-Cameron model [59] with the Cook parameters [60]. Calculations of the gamma-ray transition probabilities in the continuum region of the excited states of all nuclei were carried out in correspondence with the hypothesis of the dominance of the giant dipole resonance with radiative strength function from Kopecky-Uhl systematics [61]. Recommended parameters for the giant dipole resonances were taken from [62]. In Fig. 2 the evaluated $^{93}$Nb(n,γ) reaction cross-section is presented in comparison with the experimental data and other evaluations.

6. Summary

A new evaluation of the $^{93}$Nb(n,γ) reaction cross-section was carried out. In the energy range $10^{-5}$ eV – 7.5 keV the evaluation is based on the experimental data. A resonance analysis of the $^{93}$Nb(n,tot) and $^{93}$Nb(n,γ) reaction cross-sections was carried out. Unlike the other evaluations the energy dependences of the cross-sections in the resolved resonance region are parameterized within the framework of the Reich-Moore formalism, most accurate approximation of the collision matrix. 48 new resonances were identified compared to the Mughabghab’s systematics. In the energy range 7.5 keV–20 MeV the evaluation is based on the experimental data and results of theoretical model calculations carried out with the code GNASH. The resulting recommended cross-sections and their covariances in this range were calculated by approximation of the experimental and calculated values by means of PADE2 code [63]. The new evaluation provides essentially better consistency with the experimental data than other evaluations.

References
