DIRECTION DE L'ENERGIE NUCLÉAIRE
DIRECTION DU CENTRE DE CADARACHE
DÉPARTEMENT D'ÉTUDES DES RÉACTEURS
SERVICE DE PHYSIQUE DES RÉACTEURS ET DU CYCLE
LABORATOIRE D'ÉTUDES DE PHYSIQUE

NT	**SPRC/LEPH – 05/201**	**0**	**5H64**	**A-DONUT-02-01**		**1/20**
NATURE	CHRONO UNITE	INDICE	UNITE	EOTP	CLASSEMENT UNITE	PAGE

Note Technique

TITRE : **REVISION DU DOMAINE DES RESONANCES RESOLUES DES ISOTOPES DE L'HAFNIUM POUR JEFF-3.1**

TITLE : **REVISION OF THE RESOLVED RESONANCE RANGE OF THE HAFNIUM ISOTOPES FOR JEFF-3.1**

AUTEUR(S) : G. Noguere[1], A. Courcelle[1], P. Siegler[2], J.M. Palau[1], O. Litaize[1]

[1] *CEA-Cadarache, F-13108 Saint Paul Les Durance, France*
[2] *EC-JRC-IRMM, B-2440 Geel, Belgium*

RÉSUMÉ : L'étude des propriétés neutroniques des isotopes de l'hafnium (^{174}Hf, ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf et ^{180}Hf) a été réalisée à l'aide des récentes données mesurées par temps de vol (TOF) auprès du LINAC du Renssealer Polytechnic Institute (RPI). L'objet de ce travail est la ré-évaluation des données importantes pour l'industrie nucléaire, à savoir la section efficace de capture de l'hafnium naturel à basse énergie et son intégrale de résonance.

ABSTRACT : The neutron properties of ^{174}Hf, ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf and ^{180}Hf have been investigated on the basis of the recent experimental work carried out at the linac facility of the Renssealer Polytechnic Institute (RPI) with the Time-Of-Flight (TOF) technique. The focus of this work is a re-evaluation of the key data for the nuclear industry which are the natural hafnium capture cross section at low neutron energies and its capture resonance integral.

MOTS CLÉS : HAFNIUM, SECTION EFFICACE, INTEGRALE DE RESONANCE, SAMMY

	Rédacteur	Vérificateur	A.Q.	Émetteur
Nom et visa	G. Noguere	O. Serot	A. Bernard	Chef du LEPH J.M. Ruggieri
Date	30/06/2005			

Cadre de réalisation du document
DOB .... DSOE
Domaine .... Simulation
Projet .... DONUT
Contrat .... collaboration JEFF

CLASSIFICATION				
DR	CC	CD	SD	Sans
				■

INDICE	OBJET DES RÉVISIONS	DATE	N° CHRONO
1	EMISSION INITIALE	30/06/2005	05/201

Table of Contents

1 Introduction

Hafnium is a ductile metal which does not exist as a free element in nature. The hafnium isotopes ^{174,6,7,8,9}Hf and ^{180}Hf are found combined in zirconium compounds with a respective natural abundance of 0.16%, 5.26%, 18.60%, 27.28%, 13.62% and 35.08% . Hafnium is very corrosion resistant, has impressive mechanical properties and shows good absorption for thermal and epi-thermal neutrons. Due to these properties, it has been selected for a long time in reactor engineering to be used as neutron absorbing material holding in steel clad control rods to regulate the fission process.

Experimental results reported in the literature obtained either from microscopic or integral measurements have pointed out that all existing data libraries show deficiencies concerning description of the nuclear properties of the hafnium isotopes. Transmission Time-Of-Flight (TOF) measurements [1], carried out at the white neutron source GELINA of the Institute for Reference Materials and Measurements (IRMM), have stressed large discrepancies, over a wide energy range from 2 eV to 50 keV between the measured data and the total cross section recommended in the European (JEFF), Japanese (JENDL) and American (ENDF/B) neutron libraries.

In the epi-thermal energy range, specific trends on the natural hafnium capture cross section have been deduced from critical experiments with UOX fuel conducted by CEA in the EOLE (LWR square lattice) and AZUR (fuel plates of naval reactors) zero-power reactors located at Cadarache [2, 3]. The EPICURE or CAMELEON critical experiments have highlighted an overestimate of few percents ($2-4\%$) of the JEF-2.2 natural hafnium effective capture resonance integral. The analysis of oscillation experiments performed in the fast-thermal coupled critical facility STEK of the Energy research Center of the Netherlands has also pointed out C/E biases which may result from an overestimated natural hafnium capture cross section in the epi-thermal energy range [4].

These integral trends were confirmed by the experimental value of the natural Hf capture resonance integral (I_o^{nat}) deduced from the Resonance Shape Analysis (RSA) of several capture and transmission data measured at the linac facility of the Rensselaer Polytechnic Institute (RPI) [5]. The given value of 1959±2 barns is respectively 1.5%, 1.7% and 0.7% lower than the JEF-2.2 (I_o^{nat}=1989 barns), JEFF-3.0 (I_o^{nat}=1993 barns) and ENDF/B-VI.8 (I_o^{nat}=1972 barns) recommended values.

The present work consists in a re-evaluation of the low neutron energy range of the hafnium isotopes on the basis of the resonance parameters obtained at RPI up to 200 eV. Owing to their contribution of about 93% to I_o^{nat}, a special attention is paid to the determination of the capture area of the broad s-wave resonances at 1.1 eV and 2.4 eV of the ^{177}Hf isotope with emphasis on the overlapping ^{176}Hf and ^{178}Hf resonances near 8 eV.

2 Experimental Data

Evaluations of the Resolved Resonance Range in the ENDF/B-VI, JEFF-3 and JENDL-3 neutron libraries are mainly based on the recommended values given in the Brookhaven compilation from Mughabghab [6]. For the low neutron energy range, Hf resonance parameters established by Mughabghab comes from the combination of data reported by Fuketa et al. [14, 15], Moxon [16], Liou et al. [17] and Rohr et al. [18]. Above 2.6 keV, nuclear data are deduced from the capture area values determined by Beer et al. [19]. The list of experimental works dealing with neutron resonance spectroscopy of hafnium is given in Table 1. Resonance parameters for the two ^{177}Hf resonances at 1.1 eV and 2.4 eV and for the ^{176,178}Hf doublet near 8 eV are given in Table 2.

The first resonance spectroscopy which has provided nuclear data for hafnium over a wide energy range has been performed by Fuketa et al. [14, 15]. He carried out two series of independent measurements at the ORNL Fast Chopper (45 m flight path) and at the RPI facility (25 m flight path). Single Level parameters were extracted from transmission of natural and isotopically enriched samples respectively from 1.1 to 210 eV and then up to 1.2 keV. Later on, Moxon analysed a large variety of capture and transmission TOF data measured at Harwell. In Reference [16], he gave accurate Multi-Level Hf parameters up to 30 eV (resonance energy, neutron widths and radiation widths) and was the first to distinguish the ^{176,178}Hf doublet near 8 eV. Beyond 30 eV, Moxon proposed preliminary values which account for the results reported by Fuketa. Two years later, in 1976, a combined area analysis of capture and self-indication measurements of an isotopically ^{177}Hf enriched sample (74%) and of a natural Hf sample carried out at the GELINA facility has lead to a set of E_o, J, Γ_γ and Γ_n values up to 300.0 eV [18]. The comparisons with the Moxon's results highlight large unexplained discrepancies. It is to be noted that, at that time, report from Moxon was classified. The work reported by Beer [19] consists in a shape analysis of Hf capture data measured at the Oak Ridge Linear Accelerator (ORELA) within the 2.5 keV to 10 keV energy range. Results are expressed in term of capture area. Up to now, that energy range has never been re-investigated with modern experimental techniques. Consequently, the ^{178,179}Hf nuclear data sets are still incomplete around 2 keV.

The closer inspection of each work shows that the knowledge of the nuclear properties of the Hf isotopes suffer from large inequalities. The less abundant Hf isotopes, ^{174}Hf and ^{176}Hf, have been evaluated on the basis of the neutron width values extracted by Fuketa et al [14]. For the ^{174,176}Hf radiation widths, Mughabghab proposes a value close to 60 meV. Relevant experimental information has never been reported for determining accurate individual Γ_γ values. The most

Table 1 - Number of Hf resonances reported in the literature. The energy range (in eV) investigated in each work is given in square brackets. Only resonance energy is given when a single one or two resonances are reported.

Authors	Year	Ref.	^{174}Hf	^{176}Hf	^{177}Hf	^{178}Hf	^{179}Hf	^{180}Hf
Bollinger	1953	[8]			6 resonances. [1-14]	1 resonance (7.6 eV)	1 resonance (5.6 eV)	
Igo	1955	[10]			2 resonances (1.1 eV) (2.4 eV)			
Harvey	1955	[11]	1 resonance (30.5 eV)		28 resonances [5.9-105]	1 resonance (7.8 eV)	26 resonances [5.7-110]	1 resonance (73.9 eV)
Levin	1956	[12]			2 resonances (2.4 eV) (6.5 eV)			
Ceulemans	1965	[13]			2 resonances (1.1 eV) (2.4 eV)			
Fuketa	1965-66	[14, 15]	10 resonances [4.2-211]	22 resonances [48.3-1068]	107 resonances [1.1-1019]	18 resonances [7.7-1163]	75 resonances [5.6-1050]	9 resonances [72.5-914]
Moxon[a]	1974	[16]	9 resonances [13.4-211]	22 resonances [7.8-1067]	26 resonances [1.1-202]	25 resonances [7.7-2090]	43 resonances [17.6-189]	40 resonances [72.3-11350]
Liou	1975	[17]			176 resonances [3-700]	12 resonances [3-720]		
Rohr	1976	[18]			98 resonances [10-300]			
Beer	1982-84	[19, 20]		106 resonances [2708-5229]	17 resonances [2653-2767]	138 resonances [2659-8924]	41 resonances [2660-3069]	135 resonances [2700-9865]
Trbovich	2004	[5]	9 resonances [4.2-153.5]	6 resonances [7.8-177.1]	86 resonances [1.1-199.5]	3 resonances [7.7-164.7]	41 resonances [5.7-198.0]	2 resonances (72.46 eV) (171.7 eV)

---

[a] Below 30 eV, Moxon has determined a new set of Hf resonance parameters. Above 30 eV, he took into account in his fitting procedure the results obtainedby Fuketa [14].

Table 2 - Low energy resonance parameters of ^{176,7,8}Hf reported in the literature. E_o is the resonance energy and Γ stands for the total width of the resonance which is defined as the sum of the radiation width Γ_γ and of the neutron width Γ_n.

Isotope	Ref.	E_o (eV)	$\Gamma = \Gamma_\gamma + \Gamma_n$ (meV)	Γ_γ (meV)	Γ_n (meV)	Γ_n/Γ ($\times 10^{-2}$)
^{177}Hf	[8]	1.08±0.02	45±10			
($J^\pi = 3^+$)	[9]	1.095±0.005	67.77±1.0	66±1	1.77±0.02	
	[10]	1.100±0.005	69±2	67±2	2.10±0.05	3.04±0.11
	[13]	1.1				3.66±0.40
	[14]	1.099±0.001	68.3±1.0	66.4±1.0	1.92±0.03	2.81±0.06
	[16]	1.0964±0.0015	67.96±2.86	65.64±2.86	2.32±0.013	3.41±0.14
	[5]	1.1001±0.0001	67.45±0.08	65.23±0.08	2.225±0.003	3.299±0.006
^{177}Hf	[8]	2.34±0.05	<100			
($J^\pi = 4^+$)	[10]	2.39±0.01	69±1	60±1	9.3±0.2	13.5±0.3
	[12]	2.38	70±7	63±7	7.0±0.5	10.0±1.2
	[13]	2.4				12.5±0.8
	[14]	2.384±0.002	70.2±1.5	61.3±1.5	8.9±0.2	12.7±0.4
	[16]	2.3837±0.0002	69.81±0.74	61.74±0.74	8.068±0.068	11.54±0.16
	[5]	2.3868±0.0001	68.7±0.2	60.7±0.2	8.04±0.02	11.70±4.48
^{178}Hf	[8]	7.6±0.1	< 260			
($J^\pi = \frac{1}{2}^+$)	[11]	7.8±0.1			49±3	
	[14]	7.78±0.02			51±3	
	[16]	7.7718±0.0017	109.80±2.14	57.67±1.60	52.13±1.42	47.47±1.59
	[17]	7.770±0.027			49±7	
	[5]	7.7865±0.0001	106.8±0.2	53.0±0.2	53.83±0.08	50.40±0.12
^{176}Hf	[16]	7.886±0.010	61.7±13.2	57±12	~ 4.71	~ 7.63
($J^\pi = \frac{1}{2}^+$)	[5]	7.8891±0.0003	71.9±0.6	61.8±0.6	10.15±0.04	14.11±0.13

important Hf isotope for the nuclear industry, ^{177}Hf, has been widely investigated in the past. The Γ_γ and Γ_n values of the first and second resonances reported in the literature (Table 2) remain consistent within the limit of the uncertainties. The low energy range of the ^{178}Hf and ^{179}Hf isotopes is a combination of the nuclear data proposed by Mughabghab and originally compiled by Moxon [16]. The last stable isotope, ^{180}Hf, is a patchwork of a large variety of results. For the two first resonances, at 72.6 eV and 171.7 eV, Mughabghab recommends neutron widths from Fuketa [14]; parameters from 200 eV to 1 keV are those given in Reference [15]; compilation given by Moxon are used below 2.4 keV [16] and capture area from Beer et al. [19] are listed up to 9.8 keV.

3 RSA Results

The way evaluators interpret the experimental results and then combine the Mughaghab recommendations may explain the lack of consistency between Evaluated Nuclear Data Files. High-resolution transmission measurements of 1 mm, 2 mm, and 15 mm thick natural Hf samples have been performed at the GELINA facility for testing neutron data [1]. The comparison between

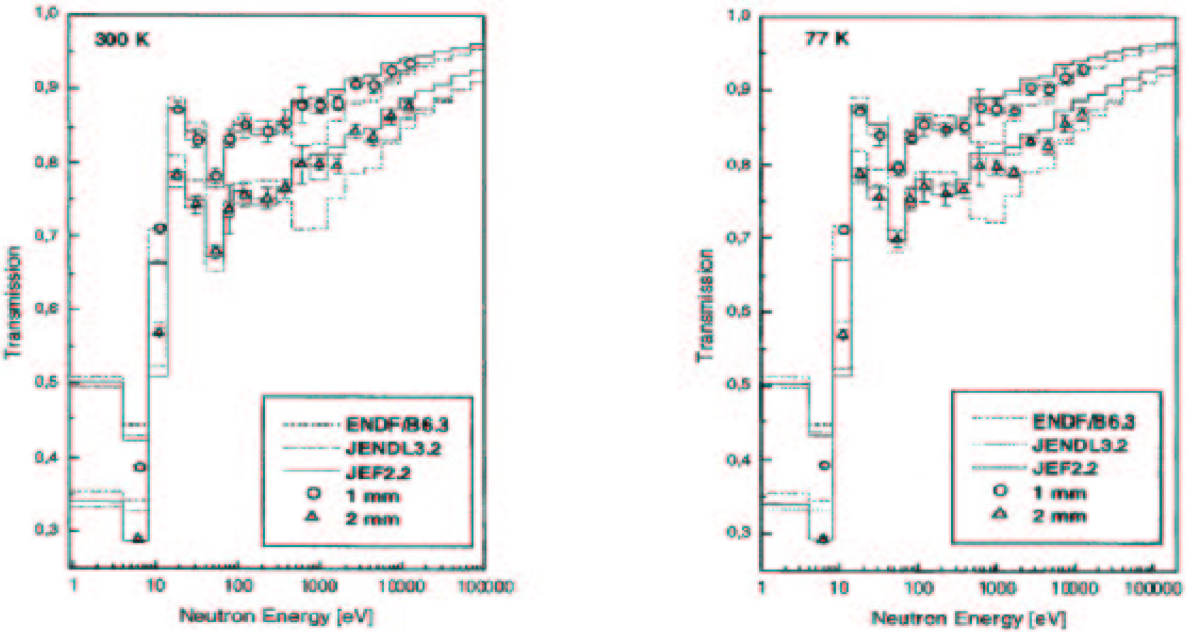

Figure 1 - Comparison of the experimental average transmission for 1 mm- and 2 mm-thick Hf samples with the calculated average transmission values at 300 K and 77 K [1]. Experimental data have been measured at the white neutron source GELINA of the Institute for Reference Materials and Measurements (IRMM). Theoretical calculations have been performed with the CALENDF code [21].

experimental results and CALENDF calculations [21] are presented in figure 1. The level of agreement depends on the neutron libraries and on the energy range. According to the authors, their experimental data give reason for a preferred use of the JENDL library. However, it is obvious that the situation below few ten's of eV needs specific improvements.

As pointed out by Moxon [16], owing to the large variation in cross section present in natural hafnium, accurate Hf Resonance Shape Analysis would require the use of at least 10 different sample thicknesses in order to reduce the effect of uncertainties in normalisation and background determination. Recent resonance parameters determined by Trbovich [5] result from a Reich-Moore analysis of 8 metallic natural Hf samples with thicknesses ranging from 4.621×10^{-5} to 1.154×10^{-2} atoms per barns, and of two liquid samples isotopically enriched in ^{176}Hf (56.17%) and ^{178}Hf (83.37%). As shown in Table 2, RPI work confirms the resonance parameters given by Moxon for the 1.1 eV of ^{177}Hf, it suggests to decrease the radiation width of the second ^{177}Hf resonance, and it gives the best Reich-Moore description of the ^{176,178}Hf doublet near 8 eV. Interpretation of the present results in term of improved capture resonance integral is discussed later on in Section 5.

Below 200 eV, we have taken into account the set of resonance parameters determined at RPI. The given E_o, Γ_γ and Γ_n values were extracted with the SAMMY code [22]. Prior resonance parameters used by Trbovich were those suggested in the ENDF/B-VI library. The low-neutron cross sections depend on the levels above 200 eV and on the potential scattering of each isotope which have been determined to accurately reproduce the total cross section of the natural hafnium. For a consistent evaluation work, we have followed a similar approach than the one used at RPI. We decided to include the RPI parameters into the Resolved Resonance Range of the

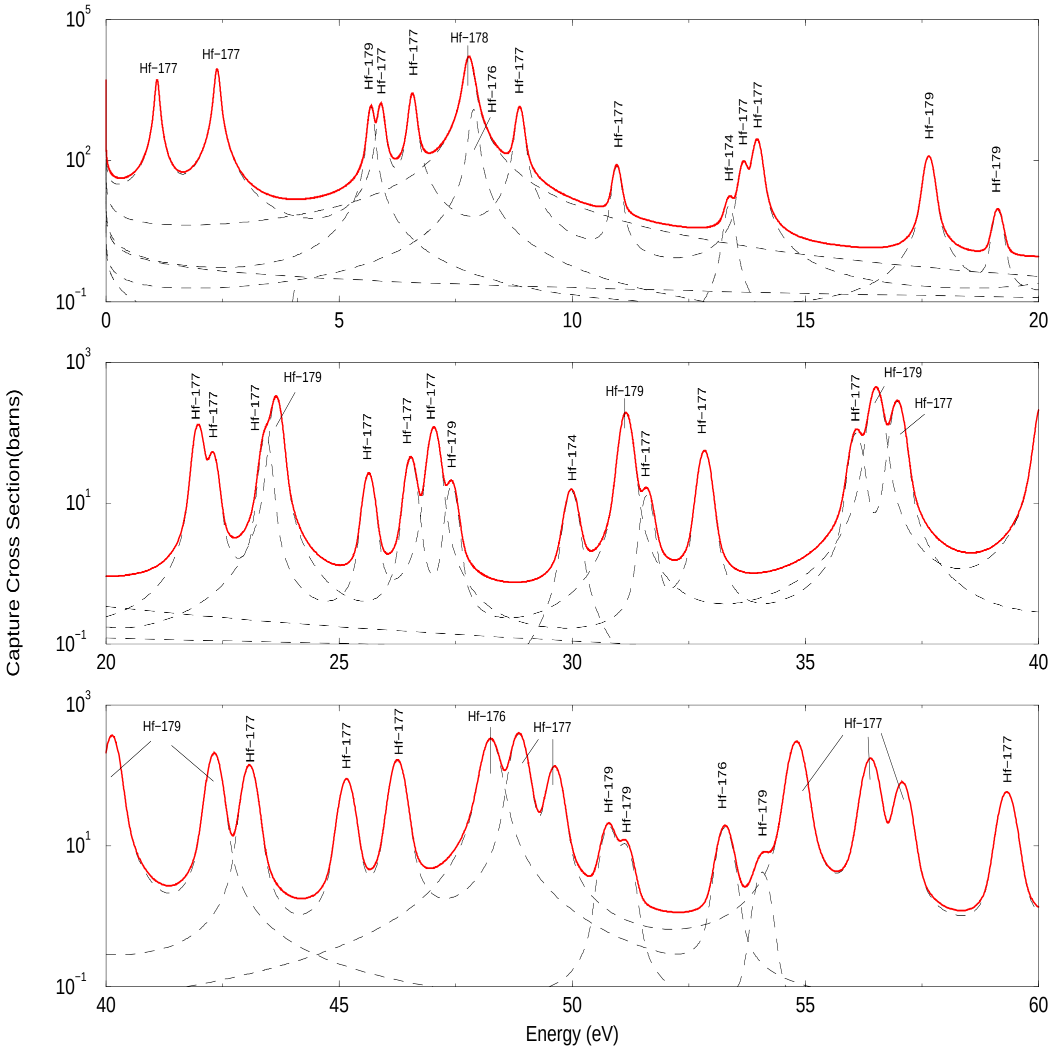

Figure 2 - The solid line represents the natural hafnium capture cross section calulated with the Reich-Moore formalism of the SAMMY code by using the resonance parameters given by Trbovich [5]. The dashed lines are contributions of the six hafnium isotopes.

ENDF/B-VI library and to use the Reich-Moore approximation (RM) of the R-Matrix theory rather than the original Multi-Level Breit-Wigner formalism (MLBW). The RM approximation consists in neglecting the off-diagonal contribution of photon channels in the R-Matrix. The only reactions requiring explicit channel definitions are then elastic scattering and fission. Therefore, for non-fissile heavy nucleus, effect of that change on the neutron cross section modeling can be disregarded. Calculations with the nuclear data processing system NJOY [23] performed on the Hf isotopes with the RM or MLBW formalism give similar results. The discrepancies are within the fractional reconstruction tolerance as specified in input of the NJOY code ($ERR < 0.1\%$).

Figure 2 shows the natural hafnium capture cross section calculated with the RM approach of the SAMMY code. The complex resonance structure is dominated by the ^{177}Hf isotope. The peak cross-section values of the resonances at 1.1 eV and 2.4 eV reach respectively 5180 barns and 8789 barns. Near 8 eV, one can distinguish the non-negligible contribution of the ^{178}Hf isotope. Between 15 eV and 45 eV, the natural Hf capture cross section is significantly affected by the

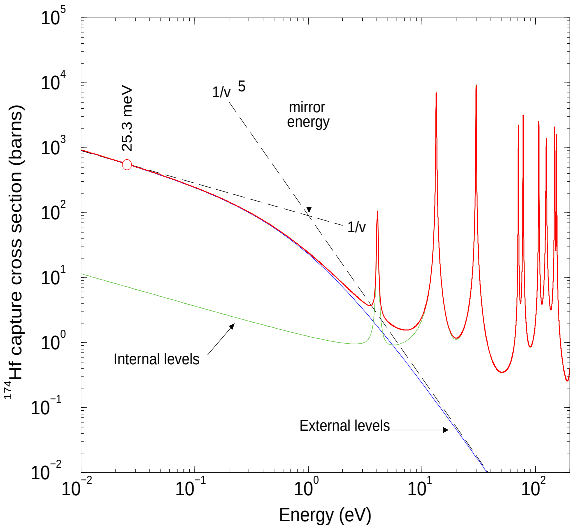

Figure 3 - Contribution of the bound level (-1 eV), recommended in ENDF/B-VI, in the ^{174}Hf capture cross section. In the thermal energy range, one can observed the two asymptotic behaviours of the cross section ($1/v$ at low and $1/v^5$ at high energies) around the so-called "mirror-energy" $E_o = 1$ eV.

resonances of the ^{179}Hf(n,γ) reaction.

The upper energy bounds of the Resolved Resonance Range of the ^{174,6,7,8,9}Hf and ^{180}Hf are respectively set to 220 eV (10 resonances), 700 eV (17 resonances), 250 eV (94 resonances), 1500 eV (22 resonances), 250 eV (49 resonances) and 2500 eV (14 resonances). For the high neutron energy region, we have taken into account conclusions of the work performed at the GELINA facility [1]. The Japanese evaluations have been considered as good candidates for describing the Unresolved Resonance Range and continuum region of the Hf isotopes.

4 Thermal Energy Range

In this work, the low Hf neutron energy range is described with the resonance parameters determined at RPI. However, no relevant information on the negative resonances has been reported in the main Reference [5]. Description of the Hf thermal energy range has been revised to account for new Reich-Moore description of the Resolved Resonance Range, to fit experimental data (Tables 3 and 4), to include recent ^{174}Hf recommended values proposed by Mughaghab [7] and to improve description of the sub-thermal behaviour of the ^{179}Hf and ^{180}Hf cross sections.

In practice, considering only the observed resonances does not allow a satisfactory fit of the experimental data. The R-matrix theory shows that the cross sections in a limited energy range depend not only on the resolved resonances in that range (*internal levels*), but also on the *external* levels outside. Typically, some difficulties arise in resonance analysis whenever compound levels below the neutron separation energy ($E < 0$) happen to be omitted. Although these external levels are unobservable and therefore unknown, they must be introduced in the R-matrix formalism. Various ad-hoc approaches exist to describe their contributions. A convenient approximation consists of accounting for the effect of such distant levels by using the tails of broad resonances having negative energies [55]. The appropriate resonance energy E_o and partial widths (Γ_γ and

Table 3 - Hf thermal capture cross sections σ_γ^{th} in barns.

Author	Ref.	Year	^{174}Hf	^{176}Hf	^{177}Hf	^{178}Hf	^{179}Hf	^{180}Hf	^{nat}Hf
Seren	[24]	1947						10±2	
Colmer	[25]	1950							110±20
Harris	[26]	1950							171±9
Pomerance	[27]	1951							102±5
Egelstaff	[28]	1951							134±8
Berstein	[28]	1952							100
Pomerance	[29]	1952	500	15	375±30	72±11	50±25	13±5.2	
Egelstaff	[30]	1953							144±10
Bollinger	[8]	1953			350±50	90±20	75±15		
Tattersall	[32]	1960							134±40
Meadow	[34]	1961							101.4±0.5
Esch	[35]	1961	390±55						95±14
Schermer	[36]	1961							93.0±0.7
Scoville	[16]	1964		21.5±3.5	289±27	86±7	38±3	11±2	124±8
Carre	[38]	1966							101±0.5
Scharf-Gol.	[39]	1967						12.6±0.7	
Holden	[47]	1968	390					12.2	
Conrad	[43]	1969		31.5±1.8	339±12	88.7±1.4	45±1	26±2	
Moxon	[16]	1974		13.7±3.2	366.4±82.0	77.4±2.2	38.6±2.12	12.06±2.12	103±2
Vertebnyi	[48]	1974	635±50	14.9±1.5	318.3±5.5	94±4	58±2	29±3	
Pavlenko	[49]	1975	549±12	24.3±2.7	360.5±3.5	84.1±1.5			
Mannhart	[50]	1975						13.04±0.07	
Heft	[51]	1978	620±20					13.1±0.1	
Kim	[53]	1999						13.04±0.3	
Cho	[54]	1999							109.2±3.7
Weighted Average			565±10	21±1.0	347±3	85±1	46±1	13.1±0.1	100.0±0.3
Mughabghab*	[6]	1984	561±35	23.5±3.1	373±10	84±4	41±3	13.04±0.07	104.1±0.5
Mughabghab*	[7]		549±7	23.5±3.1	373±10	84±4	41±3	13.04±0.07	104.1±0.5
This work			549.5	21.3	371.8	83.9	40.8	13.1	104.2

* Recommended values

Table 4 - Hf thermal total cross sections σ_t^{th} in barns.

Author	Ref.	Year	^{174}Hf	^{176}Hf	^{177}Hf	^{178}Hf	^{179}Hf	^{180}Hf	^{nat}Hf
Berstein	[28]	1952							115
Joki	[9]	1961							110±2
Esch	[35]	1961							104±13
Schermer	[36]	1961							113.7±0.3
Conrad	[43]	1969		36±1	360.5±3.5	90±1	50±1	40±1	
Okamoto	[44]	1971	611±161			97±8	73±11	55±8	
Vertebnyi	[48]	1974	650±50	19.3±1.1	376±4	99.4±4.2	64.7±2.2	51±3	
Trbovich	[5]	2004							110±5
Weighted Average			646±48	28.4±0.7	367±3	90.6±1.0	52.6±1.0	41±1	113.6±0.3
Mughabghab*	[6]	1984	576±35	29.1±3.1	373.2±10	88.5±4	48±3	35.04±1	114.4±0.6
This work			597.7	26.8	371.9	90.6	48.6	35.4	115.3

* Recommended values

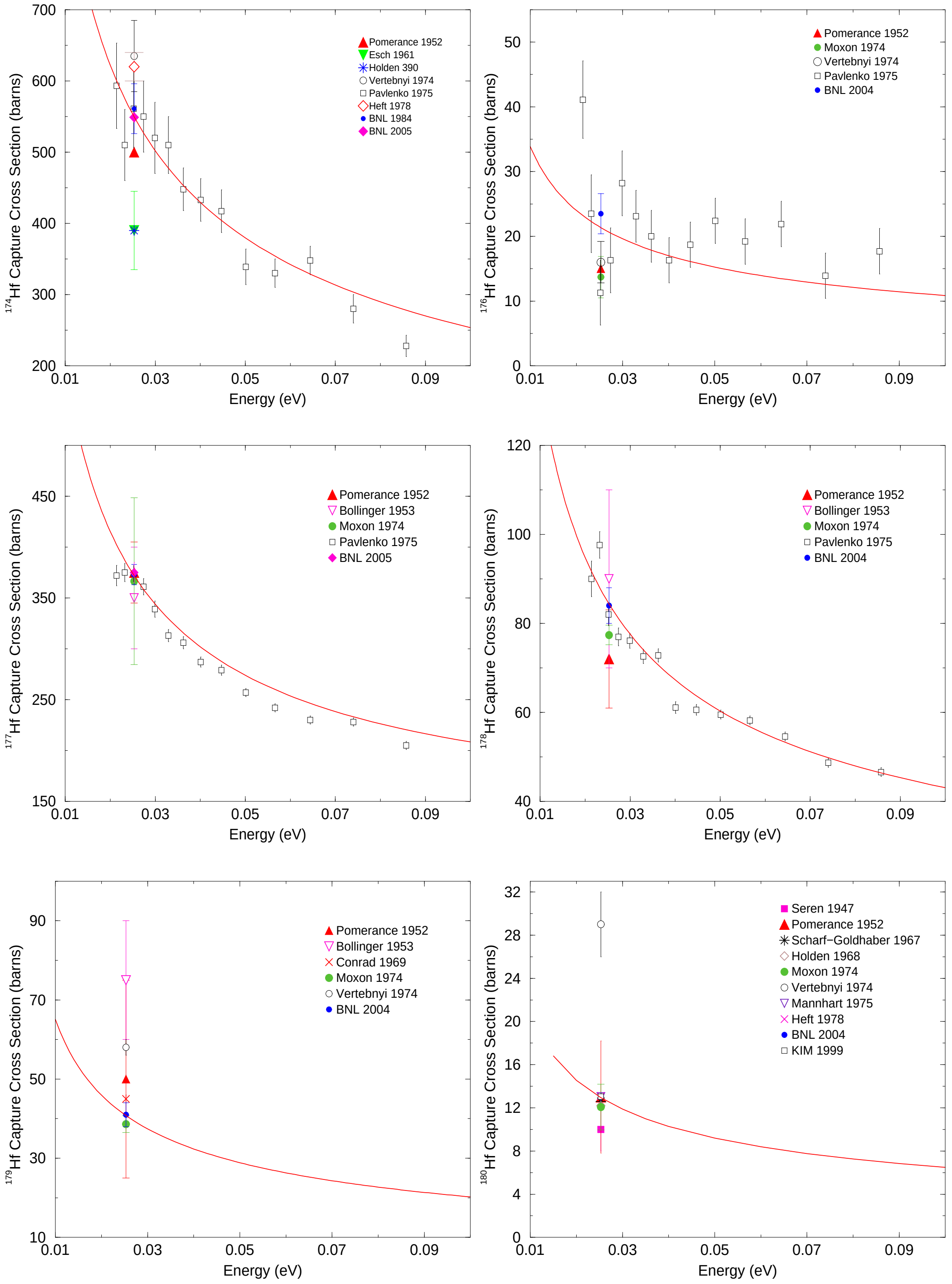

Figure 4 - The solid curve represents the Reich Moore description of the thermal and sub-thermal energy range of the ^{174}Hf, ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf and ^{180}Hf capture cross sections.

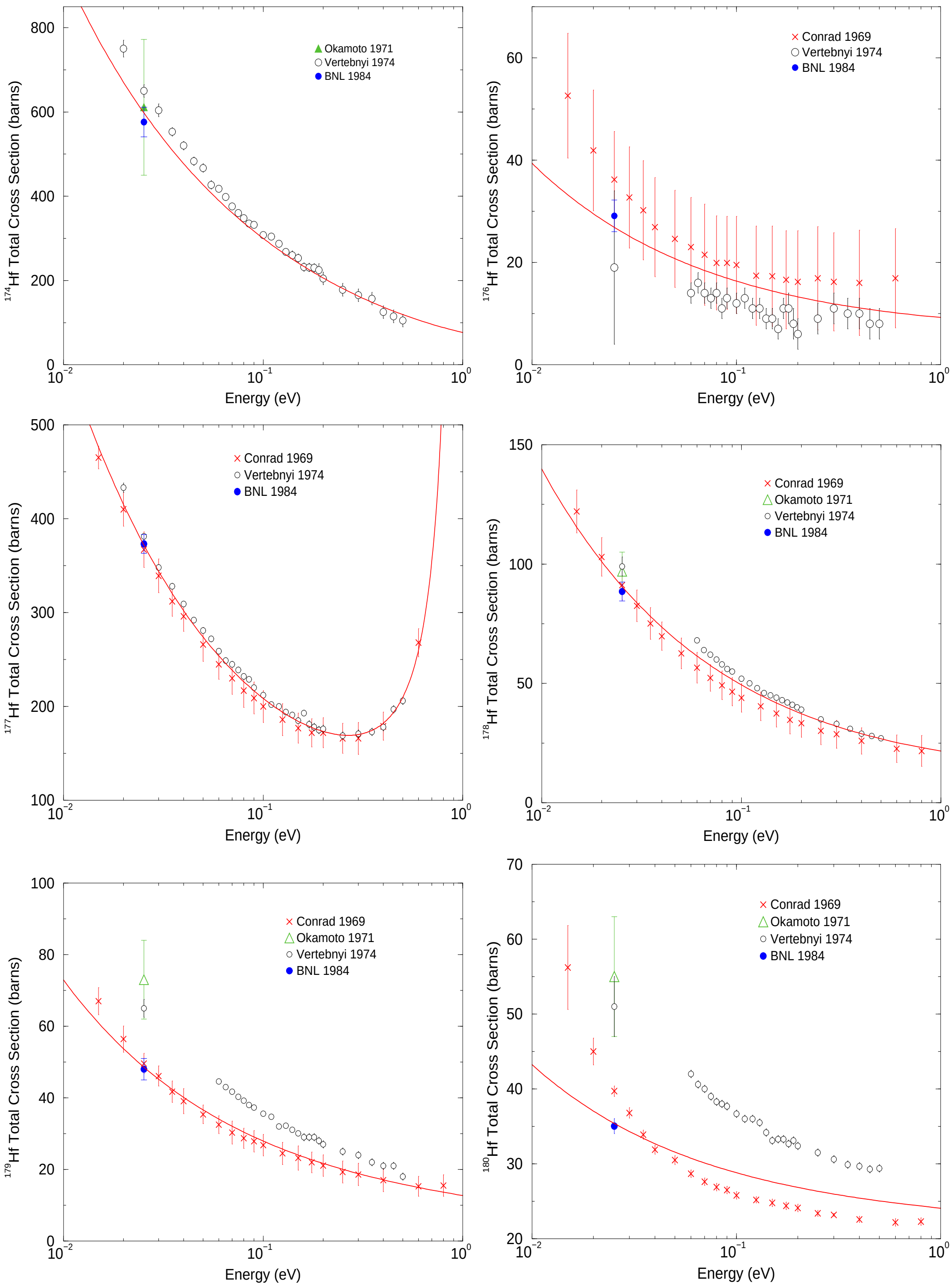

Figure 5 - The solid curve respresents the Reich Moore description of the thermal and sub-thermal energy range of the ^{174}Hf, ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf and ^{180}Hf total cross sections.

Γ_n for the Hf isotopes) are those which reproduce accurately the thermal values as well as the sub-thermal behaviour of the cross sections. Results of the least-squared adjustment performed with the SAMMY code are shown in Figures 4 and 5. The nuclear parameters of the resonances are listed in Table 5.

In the existing neutron libraries, the theoretical description of the sub-thermal energy range of the ^{174}Hf and ^{180}Hf isotopes presents a spurious behaviour around the so-called "mirror energy". That term has been introduced by Froehner to explain the contribution at the positive neutron energy $|E_o|$ of a bound level set at the negative energy E_o [55]. If the experimental data show deviation from the expected $1/v$ behaviour, this could contribute clear evidence for a bound level close to the neutron binding energy. Figure 3 shows this effect for an ^{174}Hf bound level at 1.0 eV as recommended in the ENDF/B-VI library. Similar deviation affects the ^{180}Hf capture cross section. However, in the case of these two isotopes neither experimental data nor "Westcott factor" different from unity could confirm this deviation. New ^{174}Hf and ^{180}Hf resonance parameters have been determined to shift the negative resonances to energy above the first resolved resonance. For ^{180}Hf, the original potential scattering R' of 8 fm has been decreased to 7.2 fm to get a satisfactory agreement with the Mughaghab values. For that reason, we have rejected the data reported by Vertebnyi because its capture and total cross sections deviate significantly from the BNL recommendations.

For ^{176}Hf, the contribution of the internal levels to the capture cross section is close to 16.7 barns. A σ_γ^{th} value higher than 17 bans is then expected. Therefore, results reported by Pomerance (15 barns) [29], Moxon (13.7 barns) [16] and Vertebnyi (14.9 barns) [48] are underestimated.

The ^{177}Hf thermal capture cross section is entirely defined by the nuclear properties of the internal levels. The contribution of the tails of the bound levels is negligible compared to the strength of the broad s-waves at 1.1 eV and 2.4 eV. The ^{177}Hf resonance parameters reported by Trbovich lead to a thermal capture cross section close to 372 barns which is in good agreement with the central value reported by Moxon (366.4±82.0) [16].

By contrast, the tails of the ^{178}Hf bound levels contribute to $\sim$ 13.4 barns to the capture cross section. New resonance parameters has been suggested by Trbovich to account for this significant contribution. A SAMMY fit has been performed to improve the description of the total cross section. A single one negative resonance at -49.1 eV was used to reproduce the weighted mean values of $\sigma_{tot}^{th} = 90.6$ barns.

Owing to the half-integer spin value of the ^{179}Hf isotope ($9/2^+$), two J values for s-wave resonances

Table 5 - Resonance parameters of the Hf negative resonances

E_o (eV)	J	Γ_n (meV)	Γ_γ (meV)	Isotope
-123.0	0.5	3422.27	112.3	^{180}Hf
-100.5	0.5	795.15	102.7	^{180}Hf
-80.6	0.5	350.99	58.3	^{176}Hf
-49.1	0.5	950.46	57.8	^{178}Hf
-20.1	0.5	24.66	56.3	^{176}Hf
-11.7	0.5	194.28	91.8	^{174}Hf
-9.7	4.0	26.41	52.4	^{179}Hf
-5.8	5.0	15.84	53.0	^{179}Hf
-1.2	0.5	2.47	59.8	^{174}Hf

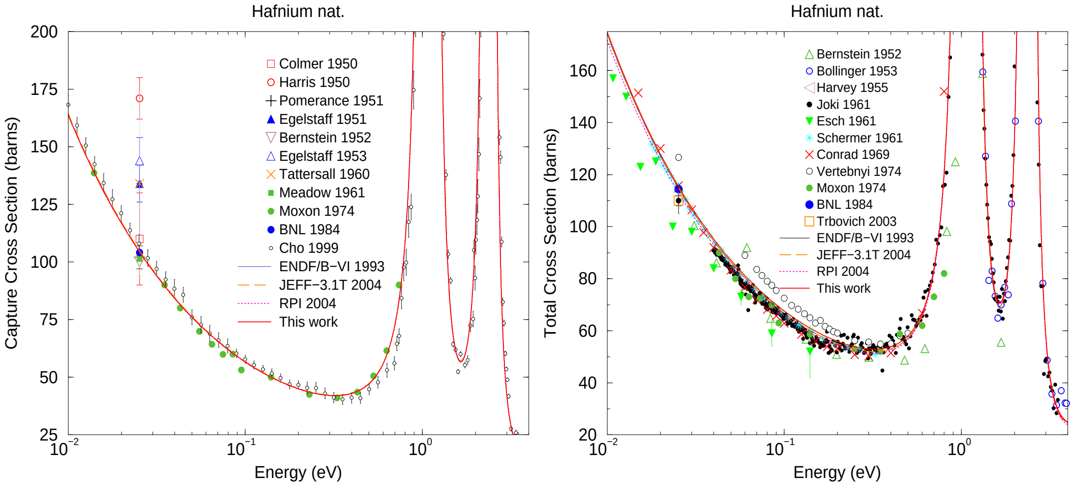

Figure 6 - Neutron capture and total cross sections of Natural hafnium below 3 eV.

are possible ($J = 4$ and $J = 5$). It is convenient to determine a set of negative resonances for each spin configuration. We have included in the SAMMY calculation two negative resonances having respectively a spin and parity of 4^+ and 5^+. Experimental data reported by Bollinger (1953), Vertebnyi (1974) and Okamoto (1971) have been rejected from the least-squared fitting procedure in accordance with the Mughabghab values.

In Figure 6 are compared the experimental data for the natural hafnium with the capture and total cross sections reconstructed with the MIXER module of the PREPRO2004 code system. The capture cross section of 104.2 barns obtained in this work remains in good agreement with the value given by Moxon (103±2 barns) and with the recent result of 109.2±3.7 reported by Cho et al [54]. Our total cross section near the thermal energy is in satisfactory agreement with the Mughaghab value of 114.4±0.6 barns. This recommendation has been probably calculated with the isotopic results reported by Conrad [43] for the ^{176,7,8,9}Hf and ^{180}Hf. The recent measurements carried out at the RPI facility have lead to a total cross section of about 110 barns. That value is about 5% lower than our evaluation. That discrepancy could be reduced by a deeper investigation of the potential scattering cross section.

5 Integral Results

For thermal reactor applications, the capture resonance integrals (I_o) of the natural hafnium is a key information for determining the probability that a neutron will be absorbed before reaching the thermal energy. The I_o values calculated with the NJOY code for each Hf isotope and for the natural element are given in Table 6. Our results are compared with the experimental I_o values reported in the literature and with recommended values from Mughabghab. Results for the European, American and Japanese neutron libraries are listed in Table 7.

The previous evaluation of the I_o^{174} capture resonance integral was based on two experimental trends close to 304 barns [47] and 350 barns [51]. With the resonance parameters determined by Trbovich, the contribution of the internal levels to I_o^{174} reaches 367.5 barns. The latter contribution remains in good agreement with the previous experimental trends. That observation suggests a negligible impact of the negative resonances to I_o^{174}. However, it was not possible to determine

Table 6 - Hf capture resonance integral I_o in barns. Values in brackets are experimental or evaluated I_0/σ_γ^{th} ratios.

Author	^{174}Hf	^{176}Hf	^{177}Hf	^{178}Hf	^{179}Hf	^{180}Hf	^{nat}Hf
Harris [26] 1950						19.7	1400
Spivak [31] 1956							1750
Macklin [31] 1956						21.8	1300
Klimentov [31] 1959							1470±200
Tattersall [32] 1960							2850±350
Feiner [33] 1961							1950
Le Sage [37] 1966							2014±150
Carre [38] 1966							2080±50
Fulmer [40] 1968		640±30	7380±500	1950±120	660±30	43±8	
Roger [41] 1968		880±40	7000±240	1710±50	595±55	52±7	{ 1905 ± 70† 2300 ± 60‡
Ricabara [42] 1969						24.3±1.6	
						(1.92±0.08)	
Steinnes [45] 1972						32±3	
Alian [46] 1973						30.8	
VandDerLinden [47] 1974	304±15					31.5±1.6	
Heft [51] 1978	350±20					33.7±2.1	
Simonits [52] 1980						(2.52)	
Ahmad [52] 1982						(2.437±0.158)	
Trbovich [5] 2004	375±20	692±2	7196±8	1872±4	506±3	28.8±0.1	1959±2
Mughabghab* [6] 1984	436±35	880±40	7173±200	1950±120	630±30	35.±1.0	1992±50
	(0.78±0.08)	(37.4±5.2)	(19.2±0.7)	(23.2±1.8)	(15.4±1.3)	(2.68±0.08)	(19.1±0.5)
This work	442.3	694.3	7211.1	1871.5	509.2	29.7	1968.7
	(0.8)	(32.6)	(19.4)	(22.3)	(12.5)	(2.3)	(18.9)

* Recommended values; † from isotopic I_o values; ‡ from measurement

a set of negative resonances which could reproduce simultaneously the experimental I_o^{174} trends reported in the literature, the high value of the thermal capture cross section (549.5 barns) and the 1/v behaviour of the (n,γ) reaction. In this work, we have chosen to respect the 1/v law together with the thermal capture cross section. The calculations performed with the SAMMY code, leads to a capture resonance integral of about 442.3 barns. The present discrepancies cannot be solved without a new experimental investigation of the thermal capture cross section.

For ^{176}Hf, a closer inspection of the experimental data and of the recommended value shows some significant discrepancies. In the American library, I_o^{176} is close to 400 barns, while the experimental trend from RPI gives a value close to 692 barns. The American recommendation comes from the results reported by Moxon [16]. This surprising discrepancy explains the low I_o^{nat} value calculated with the isotopic evaluations proposed in ENDF/B-VI. The work performed at RPI on an enriched sample is the first which give a precise description of the ^{176}Hf nuclear properties.

The situation for I_o^{177} does not reveal any ambiguous results. The experimental data and the evaluated values are in good agreement. That could be explained from one hand by the negligible impact of the bound levels to the capture resonance integral and on the other hand by the

Table 7 - Evaluated capture resonance integrals (infinite dilution) in barns calculated with the NJOY code.

Libraries	^{174}Hf	^{176}Hf	^{177}Hf	^{178}Hf	^{179}Hf	^{180}Hf	^{nat}Hf
ENDF/B-VI	355.68	400.77	7212.45	1914.24	549.53	34.42	1972.29
JEF-2.2	321.92	614.07	7232.77	1922.51	543.88	35.59	1989.13
JEFF-3.0	363.49	893.25	7210.04	1914.16	522.61	34.03	1993.93
JENDL-3.3	363.49	893.25	7210.04	1914.16	522.61	34.03	1993.93
This work	442.3	694.3	7211.1	1871.5	509.2	29.7	1968.7

extensive studies which have been performed in the resonance range of this key isotope.

For the ^{178}Hf, ^{179}Hf and ^{180}Hf isotopes, experimental results from RPI suggest a decrease the respective capture resonance integral by at least 2%, 3% and 18%. The huge variation of I_o^{180} remains marginal with respect to its contribution to I_o^{nat} (10.4 barns).

The capture resonance integral for the natural hafnium obtained in the present study, reaches 1968.7 barns. That value is respectively 0.2% and 1.2% lower than the evaluated data proposed in the ENDF/B-VI library and in Reference [6]. The energy dependence of I_o^{nat} obtained in the present work is compared in Figure 7 with the American and European evaluations. The RPI work suggests an increase of the contribution of the two first ^{177}Hf resonances compared to the previous evaluations. By contrast, the specific analysis of the ^{176,8}Hf doublet near 8 eV lead to a better agreement with the American evaluations. That trend is consistent with the work of Moxon, in which the first identification of the doublet was reported. To conclude, the RPI results suggests an increase of the contributions of the two first ^{177}Hf resonances and of the ^{176}Hf isotopes. The latter is then compensated by a decrease of the radiation width of the first ^{178}Hf resonance at 7.6 eV.

The present trends seem to confirm the overestimation of the low-energy capture cross section in the JEF-2.2 library as suggested by the integral measurements carried out in the AZUR and EOLE facilities. However, preliminaries Monte-Carlo calculations have demonstrated that the RPI results cannot reproduced accurately the integral measurements [56]. A better agreement with the integral results could be achieved by exploring the neutron energy range between 10 eV and 100 eV with the help of new appropriate TOF data.

6 Conclusion

The low-energies cross sections of the Hf isotopes has been re-evaluated thanks to the extensive experimental work carried out at the RPI facility up to 200 eV. Up to now, resonance region of the Hf isotopes were a compilation of few works performed more than 30 years ago. The resonance parameters reported by Trbovich in 2004 give the best description of the isotopic hafnium capture cross sections below 200 eV.

In this work, the sub-thermal energy range has been revisited on the basis of a very limited number of experimental data reported in the EXFOR database. The negative resonances have been adjusted with the SAMMY code to get a satisfactory agreement with the experimental data without changing the effective potential scattering length values of each isotope. Above 200 eV, we have taken into account the resonance parameters recommended in the American library. The

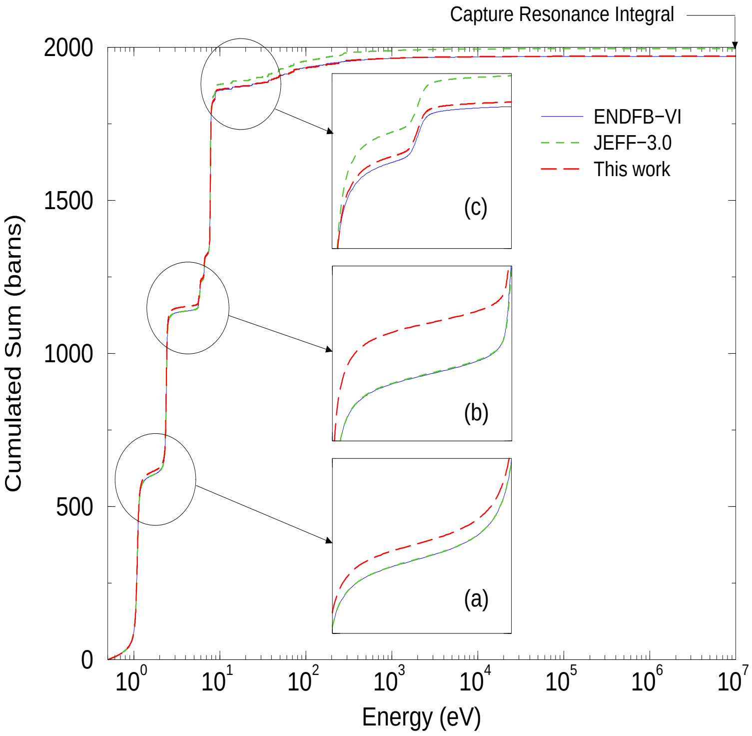

Figure 7 - Energy dependence of the capture resonance integral (infinite dilution) of the natural Hafnium. The cumulated sum obtained with the RPI resonance parameters are compared with the American and European evaluations. In the enlargements, one can distinguish the contributions of the first ^{177}Hf resonance (a), of the second ^{177}Hf resonance (b) and of the ^{176,8}Hf doublet near 8 eV (c).

unresolved resonance range and the continuum region come from the JENDL-3.3 evaluation.

The present Evaluated Nuclear Data File (included in JEFF-3.1) remains a compilation of several sources of information. The consistency of the nuclear parameters (effective potential scattering length, resonance parameters, average parameters, ...) has to be improved below and above 200 eV, and between the resolved and unresolved resonance range. At low neutron energies, a better agreement with the integral information cannot be achieved without new experimental campaigns using enriched Hf samples.

In the high energy range ("continuum" region), future evaluations could be partially based on the recent experimental work carried out with the Karlsruhe 4πBaF$_2$ detector [57] and on the new evaluations performed at the CEA/DAM. The latter will provide key information to ensure a better continuity between the low and high neutron energy regions.

Acknowledgements

The authors thank M. Trbovich for the valuable information concerning his Resonance Shape Analysis of the RPI Time Of Flight data.

References

[1] P. Siegler et al., *Testing of neutron data by comparison of measured and calculated average transmissions*, Int. Conf. on Nuclear Data for science and technology, Tsukuba, Japan, 2001.

[2] A. Santamarina et al., *Rapport de qualification R1 du produit APOLLO2.5/CEA93.V6*, Technical Report CEA-SPRC/LEPh/02-003 (2002).

[3] J.M. Palau, *Coorélations entre données nucléaires et expériences intégrales à plaques: le cas du Hafnium*, Report CEA-R-5843 (1999).

[4] K. Dietze et al.,*Integral test of neutron data and comparison of codes by re-analysis of the SEG and STEK experiments*, JEF/DOC-861 (2001).

[5] M.J. Trbovich et al., *Hafnium resonance parameter analysis using neutron capture and transmission experiments*, Int. Conf. on Nuclear Data for Science and Technology, Sept. 26 - Oct. 1 , Santa Fe, New Mexico (2004), in press.

[6] S.F.Mughabghab et al., *Neutron cross sections, neutron resonance parameters and thermal cross sections* vol 1., Academic Press (1984).

[7] S.F.Mughabghab, *Atlas of Neutron Resonances*, to be published.

[8] L.M. Bollinger et al., *Neutron absorption and scattering by Hafnium*, Phys. Rev. 92(1953)1527

[9] E.G. Joki et al., *Total Neutron cross section measurements of titanium, zirconium and hafnium*, Nucl. Sci. Eng. 11(1961)298

[10] G. Igo and H.H. Landon, *Radiation widths in the slow neutron resonances of Hafnium*, Phys. Rev. 101(1955)726.

[11] J.A. Harvey et al., *Spacings and neutron widths of nuclear energy levels*, Phys. Rev. 99(1955)10.

[12] J.S. Levin and D.J. Hughes, *Radiation widths of nuclear energy levels*, Phys. Rev. 101(1956)1328.

[13] H. Ceulemans and F. Poortmans, *Spins of slow neutron resonances in Hf, Tm and Dy*, Nucl. Phys. 62(1965)641.

[14] T. Fuketa and J.A. Harvey, *Level spacings and s-wave neutron strength functions of the isotopes of Hafnium*, ORNL report, ORNL-3778 (1965).

[15] T. Fuketa et al., *Analysis of total neutron cross section data for the hafnium isotopes*, RPI report, RPI-328-68 (1966).

[16] M.C. Moxon et al., *Differential neutron cross-sections of natural hafnium and its isotopes for neutron energies up to 30 eV*, Harwell report, AERE-R7864 (1974).

[17] H.I. Liou et al., *Neutron resonance spectroscopy:* ^{177}Hf, Phys. Rev. C 11(1975)2022.

[18] G. Rohr and H. Weigmann, *Short range energy dependence of the neutron widths of* ^{177}Hf *resonances*, Nucl. Phys. A264(1976)93.

[19] H. Beer and R.L. Macklin, $^{178,179,180}Hf$ *and* $^{180}Ta(n,\gamma)$ *cross sections and their contribution to stellar nucleosynthesis*, Phys. Rev. C 26(1982)1404.

[20] H. Beer et al., *Neutron capture cross sections and solar abundances of* ^{160,161}*Dy,* ^{170,171}*Yb,* ^{175,176}*Lu and* ^{176,177}*Hf for the s-process analysis of the radionuclide* ^{176}*Lu*, Phys. Rev. C 30(1984)464.

[21] J.C. Sublet et al., *CALENDF-2002, user manual*, CEA Report CEA-R-6020 (2003).

[22] N.M. Larson, *RISC peripheral shielding routine collection, SAMMY-M7*, Oak Ridge National Laboratory, ORNL/TM-9179/R7 (2004).

[23] R.E.MacFarlane, *The NJOY nuclear data processing system*, version 99-90, Los Alamos National Laboratory (1999).

[24] L. Seren et al., *Thermal neutron activation cross section*, Phys. Rev. 72(1947)888

[25] F.C.W. Colmer and D.J. Litler, *Pile neutron absorption cross sections of some elements*, Proc. Phys. Soc., London, Section A, 63(1950)1175

[26] S.P. Harris et al., *Pile neutron absorption cross sections*, Phys. Rev. 80(1950)342

[27] H. Pomerance, *Thermal neutron capture cross sections*, Phys. Rev. 83(1951)641

[28] S. Berstein et al., *Slow neutron crystal spectrometry: the total cross sections of Co, Er, Hf,* ^{58}*Ni,* ^{60}*Ni, Ho and fission Sm*, Phys. Rev. 87(1952)487

[29] H. Pomerance, *Thermal neutron capture cross sections*, Phys. Rev. 88(1952)412

[30] P.A. Egelstaff, *The thermal neutron cross sections of Hf, Br, V, Se, Mo and Bi*, AERE Harwell Report (1953)

[31] V.B. Klimentov, V.M. Griazev, *Some neutron resonance-absorption integrals*, J. Nucl. Eng. 9(1959)20

[32] R.B. Tattersall et al., *Pile oscillation measurements of resonance absorption integrals*, J. Nucl. Eng. A 12(1960)32

[33] F. Feiner, *Resonance integrals of Mg, Hf and Nb*, Report KAPL-200-16 (1961)

[34] J.W. Meadows, J.F. Whalen, *Thermal neutron absorption cross sections by the pulsed source method*, Nucl. Sci. Eng. 9(1961)132

[35] L. Esch, W. Moore, *Thermal neutron activation cross sections of* ^{174}*Hf and* ^{120}*Te*, Bull. Amer. Phys. Soc. 6(1961)70

[36] R. Shermer, USAEC Washington Office Reports, WASH-1031(1961)

[37] L. Le Sage and R. Sher, *A measurement of the infinite dilution capture resonance integrals with a Moxon-Rae detector*, ANS Conf. on Reactor Physics in the Resonance and Thermal Region, San Diego (1966)

[38] J.C. Carre and R. Vidal, *Measurement of cross sections and resonance integrals by the oscillation method*, IAEA Conf. on Nuclear Data for Reactors, Paris, France (1966)

[39] G. Scharff-Goldhaber, M. McKeown, *Anomalous L-subshell conversion coefficients of the highly K-forbidden E1 transition in* ^{180m}*Hf (5.5 h)*, Phys. Rev. 158(1967)1105

[40] R.H Fulmer et al., *Measurement of absorption resonance integrals for* $^{176-180}$*Hf*, 2^{nd} Conf. on Nuclear Cross Sections and Technology, Washington (1968)

[41] J.W Rogers, J.J. Scoville, *Infinite dilution resonance absorption integrals of hafnium*, Nucl. Sci. Eng 33(1968)350

[42] M.D. Ricabarra et al., *Neutron activation resonance integrals of* ^{64,68}*Zn,* ^{85}*Rb,* ^{100}*Mo,* ^{102}*Ru,* ^{113}*In,* ^{123}*Sb and* ^{180}*Hf*, Can. J. Phys. 47(1969)2031

[43] W. Conrad et al., *Total cross section of the hafnium isotopes at low energies*, Bull. Amer. Phys. Soc. 14(1969)496

[44] K. Okamoto et al., *Total cross section of the hafnium isotopes at thge neutron energy of 0.0253 eV*, JAERI-MEMO-4301 (1971)

[45] E. Steinnes, *Resonance integrals of some nuclides of interest in neutron activation analysis*, J. Inorg. Nucl. Chem. 34(1972)2699

[46] A. Alian et al. *Thermal and epithermal neutron activation analysing using the monostandard method*, J. Radio. Chem. 15(1973)535

[47] R. Van Der Linden et al.,*A compilation of infinite dilution resonance integrals*, J. Radio. Chem. 20(1974)695

[48] V.P. Vertebnyi et al.,*Total neutron cross sections of 174, 176, 177, 178, 179, 180 hafnium isotopes in the energy range 0.06 - 0.5 eV*, 2nd National Soviet Conf. on Neutron Phys., Kiev (1974)

[49] E.A. Pavlenko, N.L. Gnidak, *Neutron capture cross sections for vanadium and hafnium isotopes*, 3rd All Union Conf. on Neutron Phys., Kiev (1975)

[50] W. Mannhart, *Thermal neutron activation cross sections with high accuracy*, Z. Phys. A 272(1975)273

[51] R.E. Heft, *A consistent set of nuclear-parameter values for absolute instrumental neutron activation analysis*, Computer in Activation Analysis and Gamma-ray Spectroscopy, Mayagues, Puerto Rico (1978)

[52] A. Ahmad et al., *Neutron activation analysis without multielement standards*, J. Radio. Chem. 72(1982)335

[53] Do-Sung Kim et al., *Measurement of thermal neutron cross section for* $^{164}Dy(n,\gamma)^{165}Dy$ *and* $^{180}Hf(n,\gamma)^{181}Hf$ *reactions*, Report JINR, Dubna (1999)

[54] H.J. Cho et al., *Measurement of neutron capture cross section of Dy and Hf in the energy range from 0.003 eV to 50 KeV*, J. Kor. Phys. Soc. 34(1999)333

[55] F.H. Frohner and O. Bouland, *Treatment of external levels in neutron resonance fitting: application to the non-fissile nuclide* ^{52}*Cr*, Nucl. Sci. Eng. 137(2001)70.

[56] G. Noguere et al., *Low-energy neutron cross sections of the hafnium isotopes*, JEFF/DOC-1077 (2005).

[57] K. Wisshak et al., *Stellar neutron capture cross sections of the Hf isotopes*, FZK Report FZKA-6962 (2004).

FICHE DE VERIFICATION DU DOCUMENT

TITRE DU DOCUMENT :

Revision du Domaine des Resonances Resolues des Isotopes de l'Hafnium pour JEFF-3.1

Rédacteur : G. Noguere

Vérificateur : O. Serot Date : Visa :

ACTIONS	SUFFISANT*	
1) Vérification de la forme		
- Résumé	oui	non
- Plan du document	oui	non
- Clarté	oui	non
- Texte	oui	non
- Figures (titres, échelles, légendes)	oui	non
- Tableaux (titres, unités)	oui	non
2) Vérification du fond		
- L'étude s'appuie-t-elle sur une analyse de l'état de l'art ?	oui	non
- Le problème est-il clairement posé ?	oui	non
- Les hypothèses sont-elles clairement définies ?	oui	non
- Les données sont-elles exactes et appropriées ?	oui	non
- Les méthodes employées sont-elles justifiées et validées ?	oui	non
- Les ordres de grandeurs sont-ils cohérents ?	oui	non
- Les documents de références et les documents applicables sont-ils mentionnés ?	oui	non
3) Vérification des conclusions Les conclusions sont-elles justifiées ?	oui	non
3) Observations		

---

*Si la rubrique ne s'applique pas, inscrire S.O. (sans objet).