Low Neutron Energy Cross Sections of the Hafnium Isotopes

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Context of this Evaluation

- Natural hafnium is composed of **six isotopes**

^{174}**Hf** (0.16%), ^{176}**Hf** (5.26%), ^{177}**Hf** (18.6%), ^{178}**Hf** (27.28%), ^{179}**Hf** (13.62%), ^{180}**Hf** (35.08%)

- Thermal reactor engineering ⇒ BWR, naval propulsion, RJH, EPR, …

- Neutron absorbing material ⇒ Capture Resonance Integral $I_o \approx 2000$ barns

- Control rods ⇒ regulate the fission process

Context of this Evaluation

- Longstanding **reactivity worth underestimation** in specific CEA integral measurements in the EOLE (LWR square lattice) and AZUR (fuel plates of naval reactors) zero-power reactors located at the Cadarache

  Interpreted as an overestimation of the natural Hf capture cross section

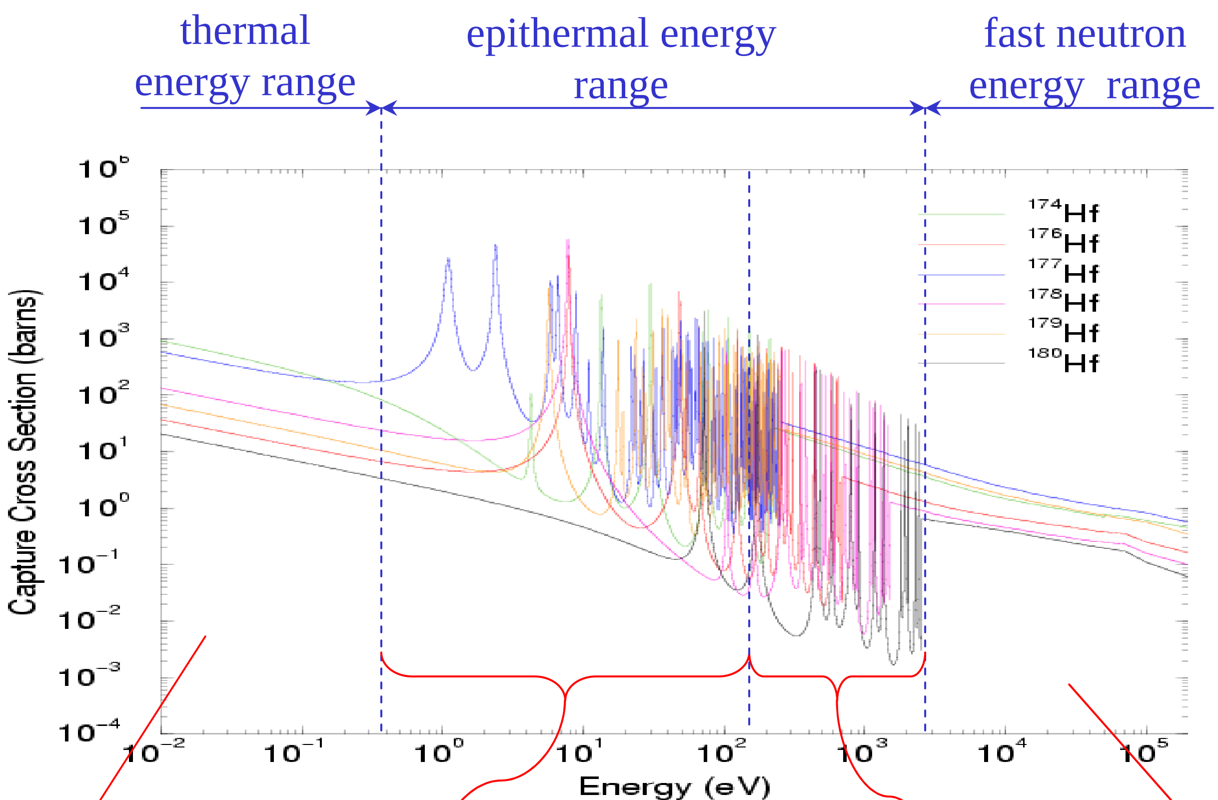
- JENDL-3.3 was the candidate for JEFF-3.1

  However, capture resonance integral is still too high for reactor applications

- New evaluation of the Resolved Resonance Range

  New resonance parameters have been extracted by **Trbovich** from TOF measurements carried out at the **RPI facility** (E < 200 eV)

Evaluation proposed for JEFF-3.1



New determination of the negative resonances with **SAMMY**

New resonance parameters from Trbovich et al. (**RPI**)

200 eV

ENDF\B-VI.8

JENDL-3.3

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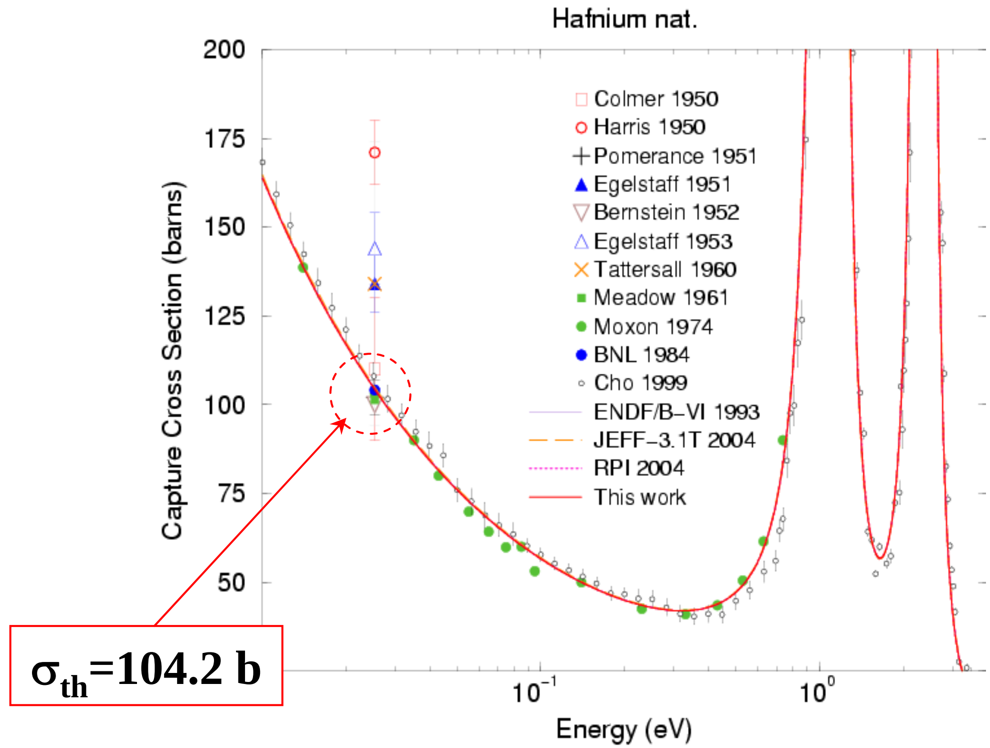
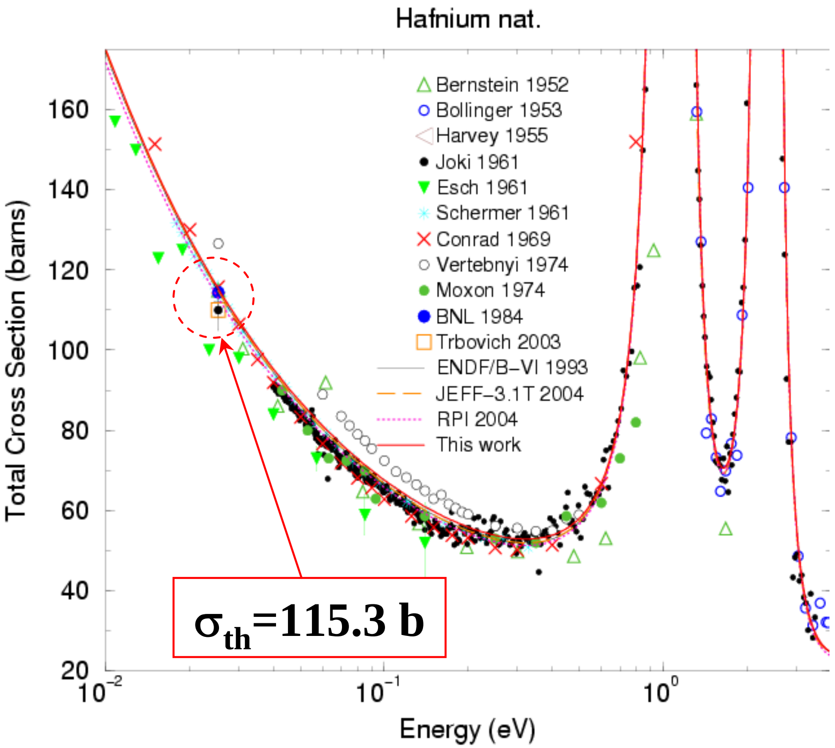
Thermal Energy range

Isotopic evaluation of the **negative resonances** based on the experimental data available in EXFOR ⇒ sequential Reich-Moore analysis of the (n,γ) and (n,tot) cross sections with the **SAMMY** code

Final comparison with the capture and total cross sections of the **natural Hf**





⇒ Significant discrepancies between experimental data

⇒ **New accurate Time-Of-Flight measurements are needed**

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Epithermal Energy range

Authors	Year	Ref.	^{174}Hf (0.14%)	^{176}Hf (5.26%)	^{177}Hf (18.60%)	^{178}Hf (27.28%)	^{179}Hf (13.62%)	^{180}Hf (35.08%)
Bollinger	1953	[5]			6 resonances. [1-14]	1 resonance (7.6 eV)	1 resonance (5.6 eV)	
Igo	1955	[7]			2 resonances (1.1 eV) (2.4 eV)			
Harvey	1955	[8]	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	[9]			2 resonances (2.4 eV) (6.5 eV)			
Ceulemans	1965	[10]			2 resonances (1.1 eV) (2.4 eV)			
Fuketa	1965-66	[11, 12]	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	[13]	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	[14]			176 resonances [3-700]	12 resonances [3-720]		
Rohr	1976	[15]			98 resonances [10-300]			
Beer	1982-84	[16, 17]		106 resonances [2708-5229]	17 resonances [2653-2767]	138 resonances [2659-8924]	41 resonances [2660-3069]	135 resonances [2700-9865]
Trbovich	2004	[2]	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)

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1965 Fuketa (E < 240 eV)

- ORNL Fast Chopper
- Transmission measurements of isotopically enriched samples
- Area Analysis (E_o and Γ_n)

⇒ First Hf resonance spectroscopy over a wide energy range
⇒ Significant number of resonances are missed (low energy resolution)

1974 Moxon (E < 30 eV)

- Harwell 45 MeV linac
- Capture and transmission measurements of natural Hf and isotopically enriched samples
- Multi-Level formalism (E_o, Γ_γ, Γ_n and spin assignement for ^{177,179}Hf)

⇒ Discovery of the existence of the ^{178,176}Hf doublet near 7.8 eV
⇒ Major influence on the cross section of ^{176}Hf in the sub-thermal energy range

2004 Trbovich (E < 200 eV)

- RPI linac facility
- Capture and transmission measurements of natural Hf and isotopically enriched samples
- Reich-Moore analysis with the SAMMY code (E_o, Γ_γ and Γ_n)

⇒ Confirms the existence of the doublet near 7.8 eV
⇒ Gives a consistent set of resonance parameters

[177]**Hf** resonances at 1.1 eV and 2.3 eV - [176,178]**Hf** doublet near 7.8 eV

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 ($J^\pi = 3^+$)	[5]	1.08±0.02	45±10			
	[6]	1.095±0.005	67.77±1.0	66±1	1.77±0.02	
	[7]	1.100±0.005	69±2	67±2	2.10±0.05	3.04±0.11
	[10]	1.1				3.66±0.40
	[11]	1.099±0.001	68.3±1.0	66.4±1.0	1.92±0.03	2.81±0.06
	[13]	1.0964±0.0015	67.96±2.86	65.64±2.86	2.32±0.013	3.41±0.14
	[2]	1.1001±0.0001	67.45±0.08	65.23±0.08	2.225±0.003	3.299±0.006
^{177}Hf ($J^\pi = 4^+$)	[5]	2.34±0.05	<100			
	[7]	2.39±0.01	69±1	60±1	9.3±0.2	13.5±0.3
	[9]	2.38	70±7	63±7	7.0±0.5	10.0±1.2
	[10]	2.4				12.5±0.8
	[11]	2.384±0.002	70.2±1.5	61.3±1.5	8.9±0.2	12.7±0.4
	[13]	2.3837±0.0002	69.81±0.74	61.74±0.74	8.068±0.068	11.54±0.16
	[2]	2.3868±0.0001	68.7±0.2	60.7±0.2	8.04±0.02	11.70±4.48
^{178}Hf ($J^\pi = \frac{1}{2}^+$)	[5]	7.6±0.1	< 260			
	[8]	7.8±0.1			49±3	
	[11]	7.78±0.02			51±3	
	[13]	7.7718±0.0017	109.80±2.14	57.67±1.60	52.13±1.42	47.47±1.59
	[14]	7.770±0.027			49±7	
	[2]	7.7865±0.0001	106.8±0.2	53.0±0.2	53.83±0.08	50.40±0.12
^{176}Hf ($J^\pi = \frac{1}{2}^+$)	[13]	7.886±0.010	61.7±13.2	57±12	~ 4.71	~ 7.63
	[2]	7.8891±0.0003	71.9±0.6	61.8±0.6	10.15±0.04	14.11±0.13

Neutron radiation widths reported by **Moxon** [13] are confirmed by **Trbovich** [2]

Γγ lowered by 1.7 %
⇒ decrease of the Effective Capture Resonance Integral

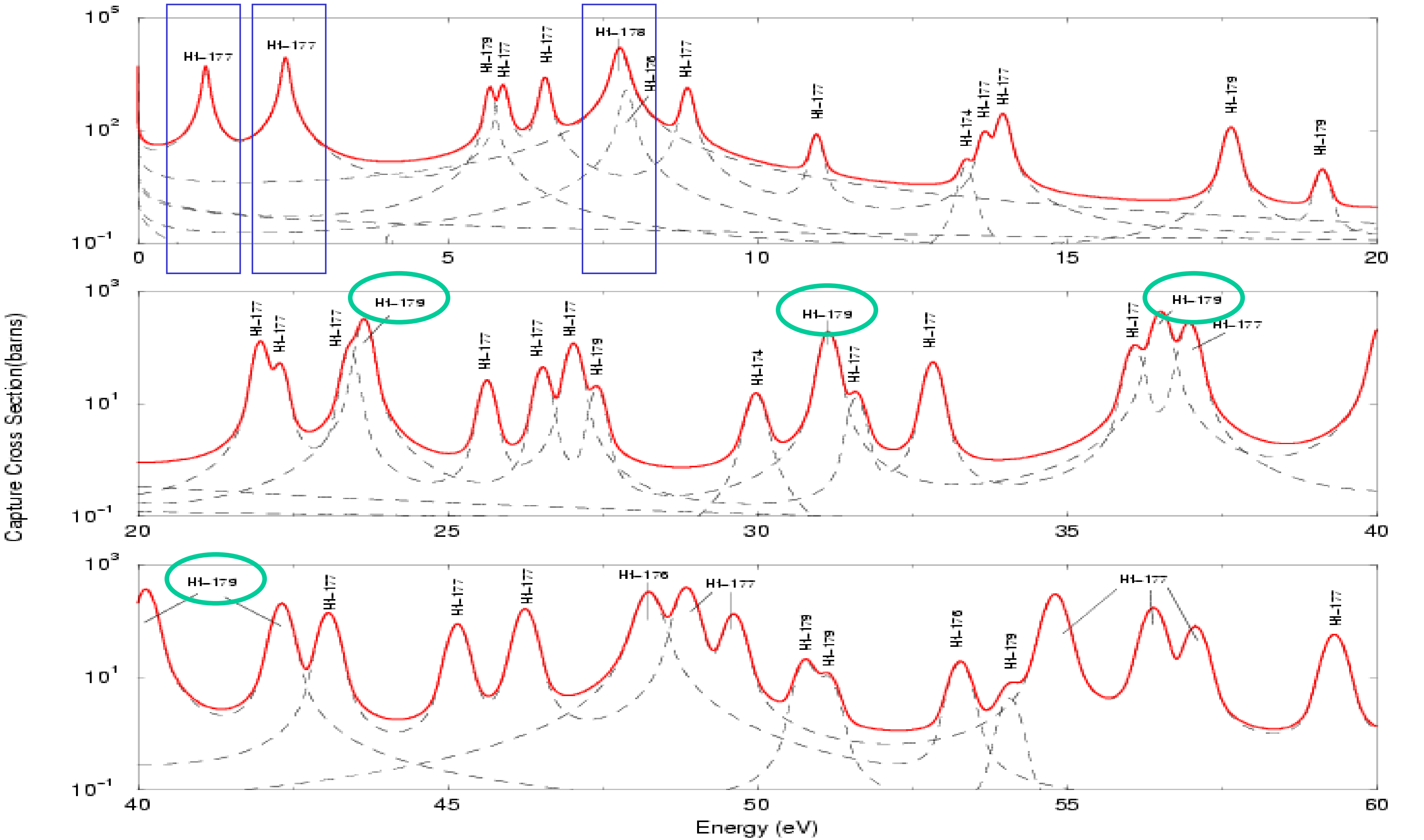
Γγ lowered by 8.8 %
⇒ decrease of the Effective Capture Resonance Integral

Γn × 2.1 ⇒ increase of the Capture Resonance Integral

- ✓ Resonance parameters agree with the integral trends.
- ✓ Uncertainties quoted by Trbovich are underestimated ⇒ systematic uncertainties not included

Trbovich et al. (below 200 eV)

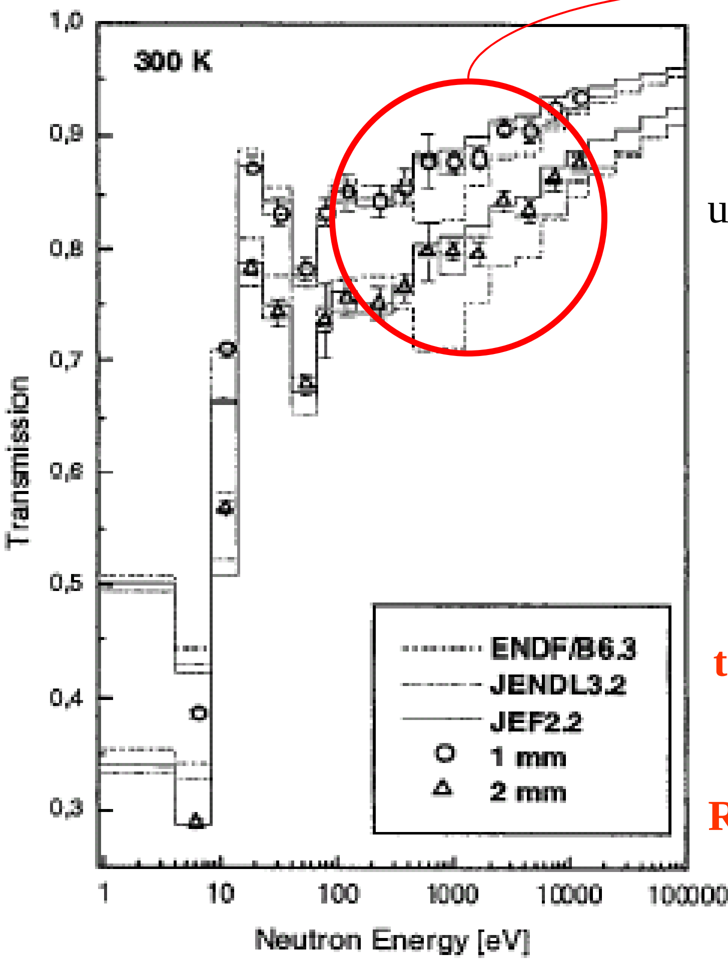
Epithermal Energy range



- ✓ Natural Hf capture cross section dominated by the ^{177}**Hf** levels
- ✓ E = 7.8 eV ⇒ significant contribution of the ^{178}**Hf** resonance
- ✓ E < 100 eV ⇒ non negligible contributions of the ^{179}**Hf** resonances

Unresolved-Resonance Range and Continuum

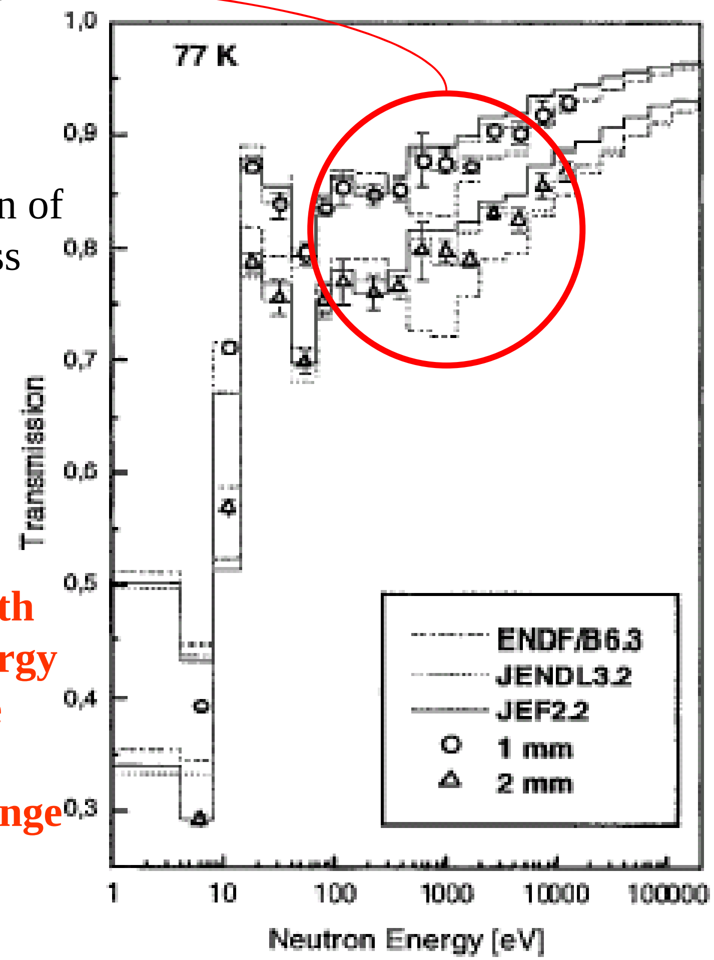
Transmission of thin natural Hf samples measured at the **GELINA** facility with the TOF technique (T=77 K, T=300 K)*



ENDF\B-VI

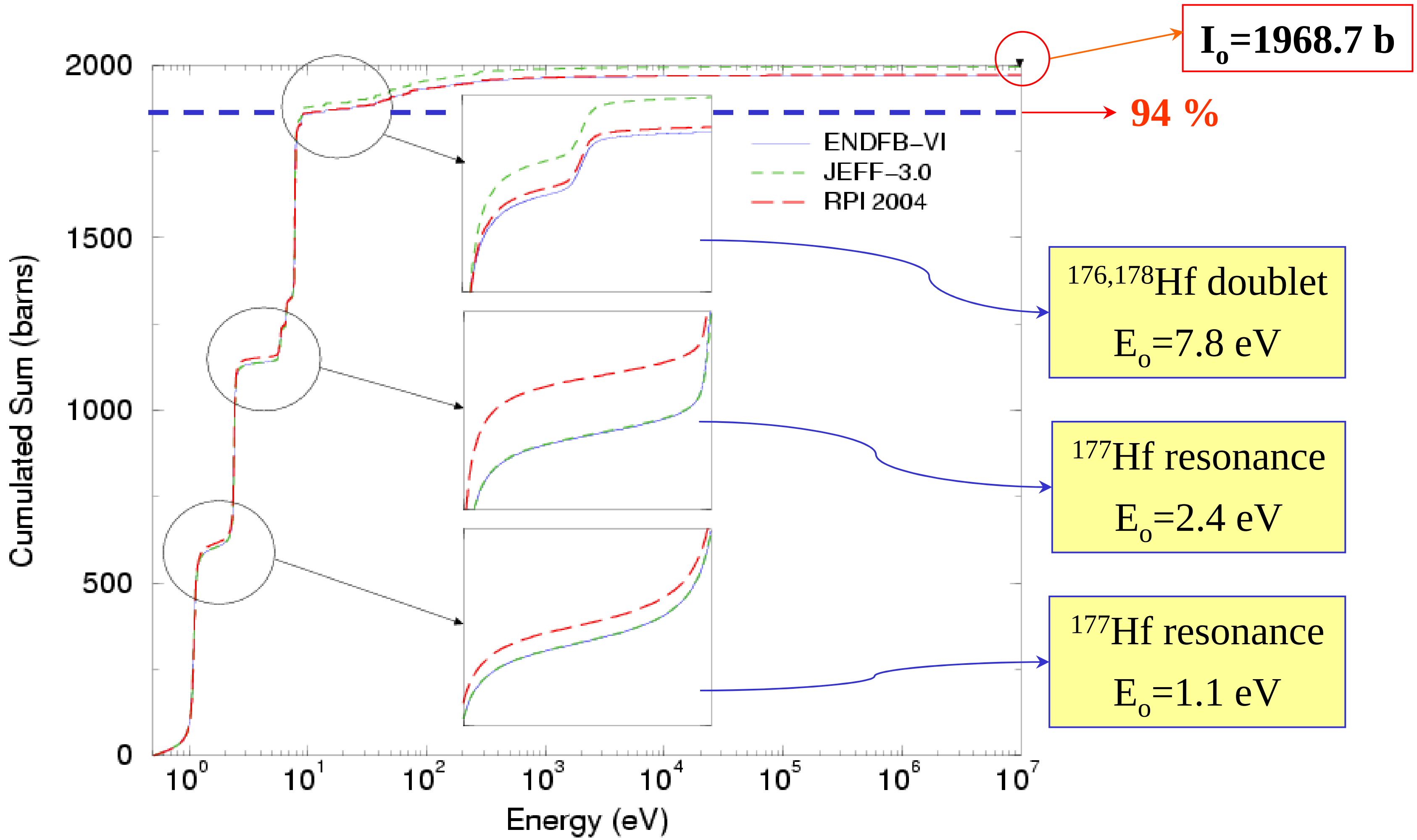
Significant underestimation of the total cross section

Problems with the upper energy limit of the Resolved Resonance Range



* P. Siegler et al., Int. Conf. ND2001

Natural Hf Capture Resonance Integral



Trbovich (RPI 2004):

⇒ compensation between the contributions of the ^{177}Hf , ^{176}Hf and ^{178}Hf

⇒ **I_o(JEFF-3.1) ≈ I_o(ENDF\B-VI.8) (Hfnat)**

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Integral Quantities

libraries		^{174}Hf (0.16%)	^{176}Hf (5.26%)	^{177}Hf (18.6%)	^{178}Hf (27.28%)	^{179}Hf (13.62%)	^{180}Hf (35.08%)	^{nat}Hf
BNL	σ_{th}	549±7	23.5±3.1	375±10	84±4	41±3	13.04 ±0.07	104.1±0.5
	I_o	436±35	880±40	**7173±200**	1950±120	630±30	35±1	1992±50
ENDF\B-VI	σ_{th}	577.2	**13.8**	373.6	84.0	43.6	13.0	104.5
	I_o	355.7	**400.8**	7212.4	**1914.2**	**549.5**	**34.4**	1972.3
JENDL-3.3	σ_{th}	561.5	23.5	373.6	84.0	42.8	12.99	104.9
	I_o	363.5	893.2	7210.0	1914.1	522.6	34.0	**1993.9**
JEF-2.2	σ_{th}	403.4	14.0	376.4	78.4	39.1	13.1	102.7
	I_o	321.9	614.1	7232.8	1922.5	543.9	35.6	1989.1
JEFF-3.0	σ_{th}	561.5	23.5	373.6	84.0	42.8	13.0	104.9
	I_o	363.5	893.2	7210.0	1914.2	522.6	34.0	1993.9
JEFF-3.1	σ_{th}	**549.5**	**21.3**	**371.8**	**83.9**	**40.8**	**13.1**	**104.2**
	I_o	**442.3**	**694.3**	**7211.1**	**1871.5**	**509.2**	**29.7**	**1968.7**

- ✓New trend for the capture cross sections and the Capture Resonance Integrale of ^{176}Hf
- ✓No significant modifications for ^{177}Hf
- ✓Decrease of the ^{178}Hf, ^{179}Hf and ^{180}Hf Capture Resonance Integral

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Preliminary Validation with TRIPOLI calculations

Simulation of two reactivity worth measurements carried out in the EOLE* (LWR square lattice) and AZUR* (fuel plates of naval reactors) zero-power reactors of the CEA-Cadarache.

CAMELEON experiment ⇒LWR square lattice containing 25 Hf rods.

	EOLE CAMELEON experiments (Hf rw. ~ 9000 pcm)	**AZUR** (Hf rw. ~ 7000 pcm)
Hf JEF-2.2	-352±30 pcm	-343±17 pcm
Hf JENDL-3.3	-398±33 pcm	
Hf JEFF-3.1	-333±31 pcm	-300±17 pcm

JEFF-3.1 still underestimates by about **~4%** the natural Hf reactivity worth

* O. Litaize and J.M. Palau, CEA-Cadarache

Conclusions and Perspectives

- This evaluation provides a body of consistent resonance parameters up to 200 eV

  However: underestimation of the reactivity worth in specific integral measurements are still not solved (~4 %)

- Hafnium isotopes eval. remains a compilation of several source of information:

  Accuracy of the **effective potential scatering length** (R') ?

  Consistency of the **average resonance parameters** (S_o, $<\Gamma_\gamma>$ and D_o) ?

  Determination of the upper energy limit of the Resolved Resonance Range ?

- For the next release:

  Experimental data in the Resolved Resonance Range would be valuable

  New modeling of the Unresolved Resonance Range are needed
  (Cf. recent experimental data from FZK*)

  Evaluation of the fast range performed by CEA/BRC to be considered

* K. Wisshak et al., *FZK 6962* (2004)