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Comparison of resonance integrals of cross sections from JEFF-3.2 library for some problematic reactions

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Abstract

The quality of the capture cross sections in JEFF-3.2 for a selection of nuclides has been assessed in comparison to other evaluated nuclear data libraries (ENDF/B-VII.1, JENDL-4.0, TENDL-2014 and IRDFF v1.05). The incident neutron capture reactions of these nuclides have been compared to experimental data from the EXFOR database in terms of resonance integrals and, where available, energy dependent data. Recommendations for the next version of the JEFF library have been given. For ^{55}Mn , JEFF-3.2 is strongly recommended. For ^{58}Fe and $^{176,178}\text{Hf}$, JEFF-3.2 is recommended. For ^{93}Nb and ^{148}Nd , JEFF-3.2 is not recommended. For these two nuclides, the capture cross section from JENDL-4.0 is recommended.

1. Introduction

Joint Evaluated Fission and Fusion File (JEFF) is an evaluated library produced via an international collaboration of Data Bank member countries co-ordinated by the JEFF Scientific Co-ordination Group, under the auspices of the NEA Data Bank [1]. The current version of the library is JEFF-3.2. The neutron induced cross section subsection represents a very important part of the nuclear data library. It consists of cross sections for different reactions for a large number of nuclides (472 in total). The quality of the evaluated data largely varies from nuclide to nuclide and depends on importance of the nuclide, availability of the experimental data, complexity of the nuclide/reaction, etc. In some cases, evaluations are simply adopted (with optional slight modifications) from other libraries (e.g. ENDF/B, JENDL, TENDL) or older version of the same library. Alternatively, for some nuclides new evaluations are performed, again largely varying in the degree of accuracy and reliability.

The first step in the process of improvement of nuclear data via re-evaluation is to assess the quality of the existing data. This can be done by comparison of the evaluated data with other libraries and/or experimental data.

The resonance integral RI is one of the most important integral parameters which can be used for validation of reaction cross sections. In most cases it is derived from activation measurements in thermalized reactor spectra, however it can also be obtained by integration of energy dependent experimental data. The resonance integral is defined as:

$$RI = \int_{E_{min}}^{E_{max}} \frac{\sigma(E)}{E} dE,$$

where the upper energy bound has been assumed $E_{max} = 100$ keV (unless explicitly stated otherwise) and the lower energy bound E_{min} corresponds to the (cadmium) cutoff energy, which is around 0.55 eV, however slight variations in different experiments are reported. Therefore, for comparison with experiments, the lower integral limit in evaluated libraries has always been adjusted to correspond to the reported experimental value. In principle, the resonance integral is defined for any reaction. In this report the analysis is limited to the neutron capture reaction.

In the following, a comparison of JEFF-3.2 to some other contemporary major nuclear data libraries and experimental data, available in the EXFOR database [2], is presented for a selection of materials for which the reaction cross sections from JEFF-3.2 are deemed problematic. Cross sections are compared both with energy dependent and integral experiments (resonance integrals). The main purpose of this work is to quantitatively assess the quality of the evaluated cross section in JEFF-3.2 for the selected reactions relative to cross sections from other currently available major nuclear data libraries.

2. Comparison of the resonance integrals for the selected reactions

The selection of the reactions was done based on a comparison of the integral resonance data from JEFF-3.2 to other contemporary libraries and the Mughabghab's compilation of resonance parameters [3], published in Ref. [4]. Important nuclides with large discrepancies of the resonance integral between JEFF-3.2 and Mughabghab have been chosen for this analysis. In this report, the evaluated nuclear data libraries are compared only to "raw" experiments (excluding compiled data e.g. those of Mughabghab) available in the EXFOR database [2].

2.1 ^{55}Mn

Reported measurements for the capture resonance integral are shown in Table 1.1. The data of Schuman (1969) (entry 11687.004), Berreth (1962), Macklin (1955) and Harris (1950) are disregarded in the further analysis since they are given without uncertainties. The data of Sampson (1962) and Walker (1960) are disregarded since they obviously strongly underestimate the resonance integral. This could indicate that the results suffer from a bias effect, such that the data cannot be considered as reliable.

Table 1.2 shows that the majority (almost 99%) of the contribution to the resonance integral comes from energies below 3.95 keV, i.e. contributions from bound states and the lowest four resonances are important. In this energy region, the low-resolution fast chopper measurements by Widder are the only relevant energy dependent capture cross section measurements available in EXFOR (Figure 1.3). JEFF-3.2 has slightly different resonance parameters of the first resonance compared to other evaluations, hence a significant difference in the resonance integral. The JEFF-3.2 resonance parameters were fitted to GELINA TOF measurements [5]. Compared to ENDF/B-VII.1, JEFF-3.2 evaluation of the resolved resonance parameters is complemented with additional transmission and measurements with diluted (MnFe alloy) samples, which reduces the multiple interaction for strong *s*-wave resonances and at the same time enables an accurate internal normalization to the 1.15 eV resonance of ^{56}Fe [5]. The comparison of the JEFF-3.2 («This work») and ENDF/B-VII.1 parameters with the experimental capture yield and transmission resulting from the measurements at GELINA is shown in Figure 1.1. The dominant contribution to the ^{55}Mn capture resonance integral come from the three resonances at around 340 eV, 1100 eV and 2370 eV (92-93% in total). In comparison to the list of experiments taken into account (see Table 1.1), **JEFF-3.2 has a statistically much better agreement than other evaluated nuclear data libraries**. However, the χ^2/n is still much larger than expected (~ 1) and (at least) some of the experiments are not consistent within the quoted uncertainties (Figure 1.2). Presumably, some of the results suffer from bias effects related to the normalization or spectral effects.

Table 1.1: Experimental data for $^{55}\text{Mn}(n,\gamma)$ *RI* from the EXFOR database.

Author	Year	EXFOR Entry	E_{min} [eV]	<i>RI</i> [b]	Accepted
Heft	1978	12866.025	0.5	14.0 ± 0.9	Yes
Gleason	1975	10644.020	0.5	15.6 ± 0.5	Yes
Van Der Linden	1972	20643.015	0.55	13.8 ± 0.8	Yes
Breitenhuber	1970	20029.003	0.55	$15.4 (1 \pm 0.05)$	Yes
Nanjyo	1970	20314.002	0.63	14.1 ± 0.6	Yes
Schuman	1969	11687.002	0.5	11.3 ± 0.7	Yes
Schuman	1969	11687.004	0.5	14	No
Sher	1968	11689.002	0.5	14.41 ± 0.48	Yes
Orvini	1968	20633.002	0.55	13.7 ± 0.7	Yes
Borchardt	1967	20647.003	0.55	12.8 ± 1.1	Yes
Koehler	1967	20654.007	0.68	12.6 ± 0.5	Yes
Louwrier	1965	20641.002	0.56	15 ± 1.4	Yes
Sampson	1962	11655.003	0.5	7.5 ± 0.8	No
Berreth	1962	12635.005	0.5	14	No
Feiner	1961	11454.002	0.55	15.6 ± 0.6	Yes
Walker	1960	11525.002	0.5	7.8 ± 0.8	No
Macklin	1955	14388.016	0.55	10.8	No
Harris	1950	11343.007	0.5	12.1	No

Table 1.2: Comparison of resonance integrals for the $^{55}\text{Mn}(n,\gamma)$ reaction.

	JEFF-3.2	ENDF/B-VII.1	JENDL-4.0	TENDL-2014	IRDFF v1.05
<i>RI</i> (> 0.55 eV)	13.99	13.26	13.26	13.26	13.23
<i>RI</i> (< 3.95 keV)	13.81	13.08	13.08	13.08	13.07
χ^2/n	3.43	5.10	5.11	5.10	5.16

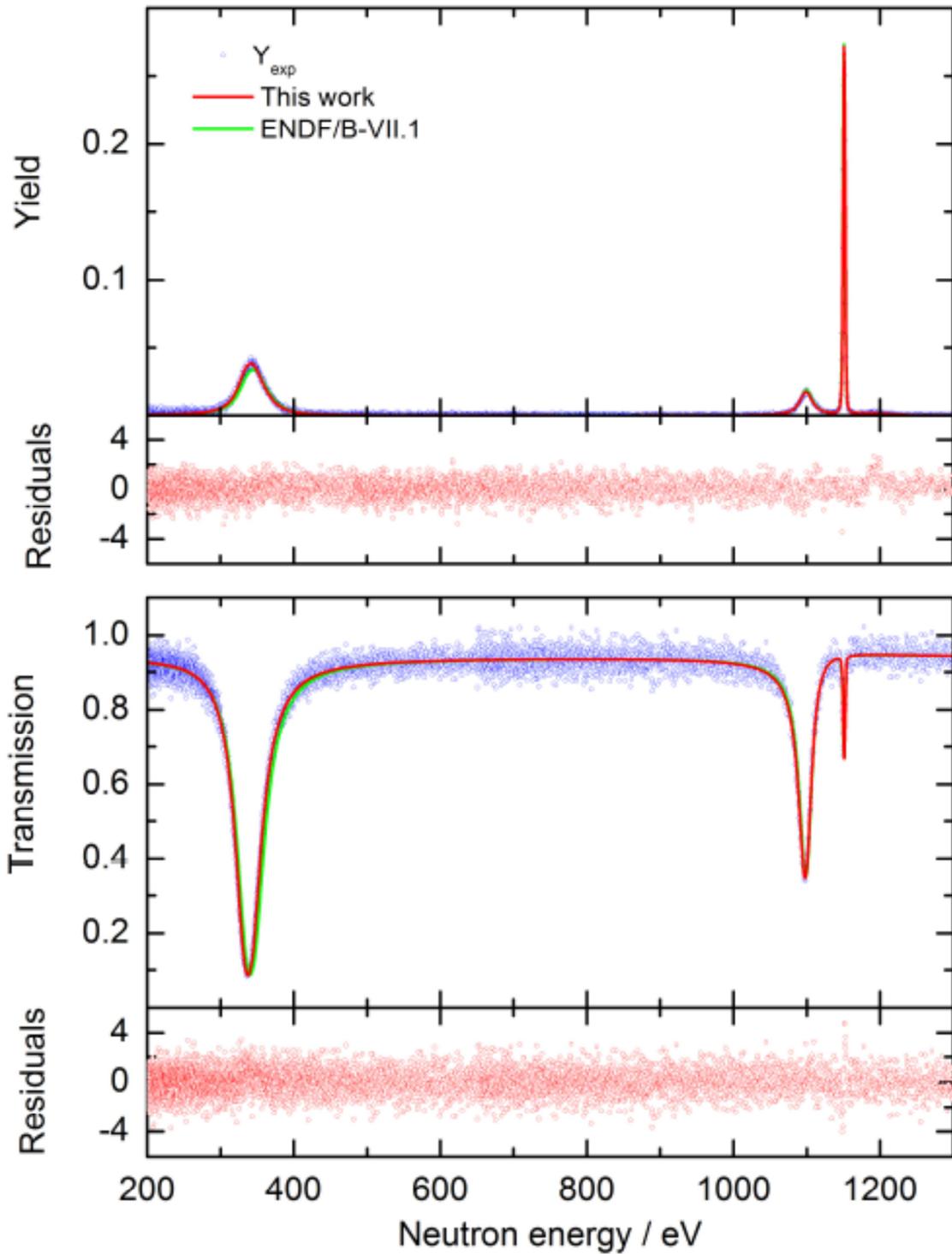


Figure 1.1: Experimental capture yield (top) and transmission (bottom) resulting from measurements at GELINA compared to calculations using fitted resonance parameters (adopted in JEFF-3.2) and the ones adopted in ENDF/B-VII.1 library. [5]

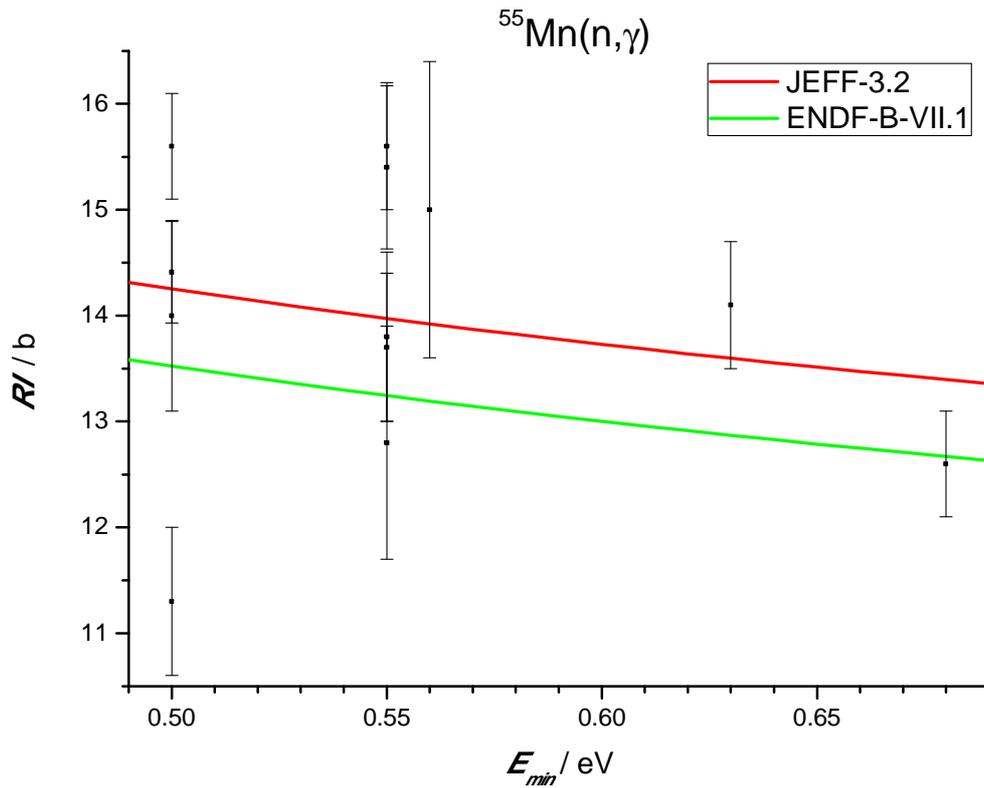


Figure 1.2: $^{55}\text{Mn}(n,\gamma)$ resonance integral RI as a function of lower energy limit E_{min} . JEFF-3.2 (red), ENDF/B-VII.1 (green; almost equal to JENDL-4.0/TENDL-2014), and experimental data (black).

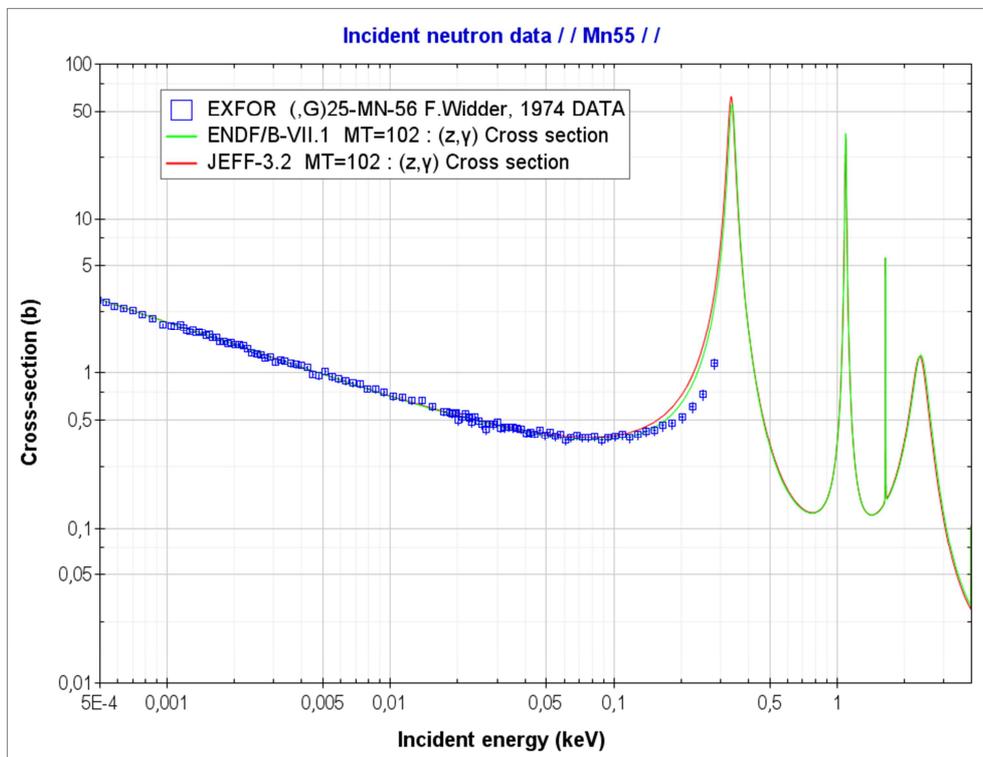


Figure 1.3: $^{55}\text{Mn}(n,\gamma)$ cross section in eV region from different evaluations and experimental data.

2.2 ^{58}Fe

The experimental database for the ^{58}Fe capture resonance integral is relatively scarce (Table 2.1). Only the data points from Heft (1978) and Steinnes (1972) are considered for further analysis. The data point from Alian (1973) lacks reported uncertainty. The point from Sage (1976) does not possess valuable information except a rough estimation of the upper bound which is one order of magnitude higher than the (as of 2016) generally accepted value. Finally, the data points from Van Der Linden (1972) and Brune (1963) are well off and are based on cross section values for thermal capture in ^{58}Fe which differ from the recommended one.

Table 2.1: Experimental data for $^{58}\text{Fe}(n,\gamma)$ RI from the EXFOR database.

Author	Year	EXFOR Entry	E_{min} [eV]	RI [b]	Accepted
Heft	1978	12866.027	0.5	1.21 ± 0.06	Yes
Sage	1976	10637.008	0.5	5.06 ± 5.06	No
Alian	1973	20644.004	0.5	1.17	No
Steinnes	1972	20188.005	0.5	1.27 ± 0.07	Yes
Van Der Linden	1972	20643.016	0.55	1.7 ± 0.1	No
Brune	1963	20050.002	0.6	0.58 ± 0.16	No

In Table 2.2 resonance integrals for the $^{58}\text{Fe}(n,\gamma)$ reaction reported in the EXFOR data base are compared. The contribution to the integral is significant for resonances up to ~ 500 keV, therefore for this specific case, the upper limit of the calculated RI has been set to $E_{max} = 1$ MeV.

Table 2.2: Comparison of resonance integrals for the $^{58}\text{Fe}(n,\gamma)$ reaction.

	JEFF-3.2	ENDF/B-VII.1	JENDL-4.0	TENDL-2014	IRDF v1.05
$RI (> 0.5 \text{ eV})$	1.273	1.492	1.357	1.357	1.272
$RI (> 0.55 \text{ eV})$	1.245	1.467	1.330	1.330	1.244
χ^2/n	0.55	16.0	3.76	3.79	0.54

In EXFOR, there are no high-resolution energy dependent cross section data on samples enriched in ^{58}Fe available. Hence, the capture widths of individual resonances can only be inferred from capture measurements on natural samples and/or transmission measurements with enriched samples, and both methods can show their limitations. **However, the resonance integral, calculated from JEFF-3.2, on average shows better statistical agreement than other three major evaluated libraries** (Figure

2.1). Furthermore, even though the number of measurement points is too small for a definite statement, the JEFF-3.2 and IRDFF version 1.05 are the only evaluations which are statistically consistent ($\chi^2/n < 1$) with experimental data.

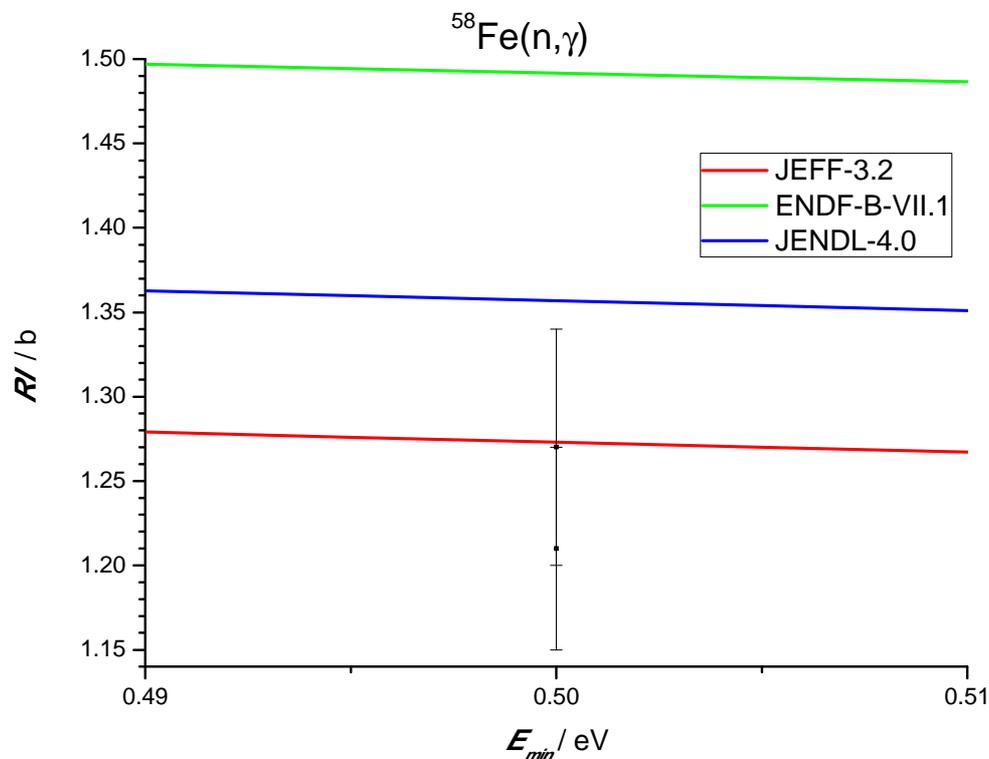


Figure 2.1: $^{58}\text{Fe}(n,\gamma)$ resonance integral RI as a function of lower energy limit E_{min} . JEFF-3.2 (red), ENDF/B-VII.1 (green), JENDL-4.0/TENDL-2014 (blue), and experimental data (black).

2.3 ^{93}Nb

Even though in EXFOR there are several experiments reported for the ^{93}Nb capture resonance integral (Table 3.1), most of them are incomplete and/or highly discrepant. Only the data point from Schuman (1969) is considered for further analysis. The data points from Hayodom (1969), Druschel (1968), Macklin (1955) and Harris (1950) lack reported uncertainties. The points from Le Sage (1966) and Feiner (1961) are clearly discrepant from Schuman (1969) and from other data in Table 3.1 that are reported without uncertainties from Table 3.1. They are also significantly higher than the (as of 2016) generally accepted value.

The comparison of the resonance integral for the $^{93}\text{Nb}(n,\gamma)$ reaction is shown in Table 3.2. The contribution to the resonance integral is significant up to ~ 1 MeV, therefore for this specific case, the upper limit of the calculated RI has been set to $E_{max} = 1$ MeV. The resonance integrals in JEFF-3.2 and ENDF/B-VII.1 are clearly higher compared to other evaluations. The differences mainly originate from the additive »background« contribution to the cross section in those two libraries, and only partly also from different values of the resonance parameters. Above the resolved resonance region (above ~ 10 keV, depending on the library) JEFF-3.2 and ENDF/B-VII.1 mainly rely on linac TOF measurements by Macklin (EXFOR entry: 10537.002), while JENDL-4.0 (as well as

TENDL-4.0 and IRDFF version 1.05) relies on Van De Graaff measurements by Reffo (EXFOR entry: 21796.005) and Kompe (EXFOR entry: 20358.002) (Figure 3.1). Due to higher resolution of the linac measurement, JEFF-3.2 and ENDF/B-VII.1 include more details than other libraries, however, this does not affect the resonance integral. **The resonance integral, calculated from JEFF-3.2 (and ENDF/B-VII.1), on average has worse agreement with the regarded measurement point than other three evaluated libraries.**

Table 3.1: Experimental data for $^{93}\text{Nb}(n,\gamma)$ *RI* from the EXFOR database.

Author	Year	EXFOR Entry	E_{min} [eV]	<i>RI</i> [b]	Comment
Schuman	1969	11899.003	0.5	8.5 ± 0.5	Acceptable
Hayodom	1969	30368.012	0.5	5.8	No unc. given
Druschel	1968	10347.003	0.5	8.5	No unc. given
Le Sage	1966	11754.003	0.5	18.8 ± 3	Clearly discrepant
Feiner	1961	11454.003	0.55	13.8 ± 2.2	Marginally acceptable
Macklin	1955	14388.031	0.49	8.3	No unc. given
Harris	1950	11343.015	0.49	4.19	No unc. given

Table 3.2: Comparison of resonance integrals for the $^{93}\text{Nb}(n,\gamma)$ reaction.

	JEFF-3.2	ENDF/B-VII.1	JENDL-4.0	TENDL-2014	IRDFF v1.05
<i>RI</i> (> 0.5 eV)	9.90	9.90	8.96	9.39	8.74
<i>RI</i> (< 10 keV)	9.08	9.08	8.14	8.43	7.92

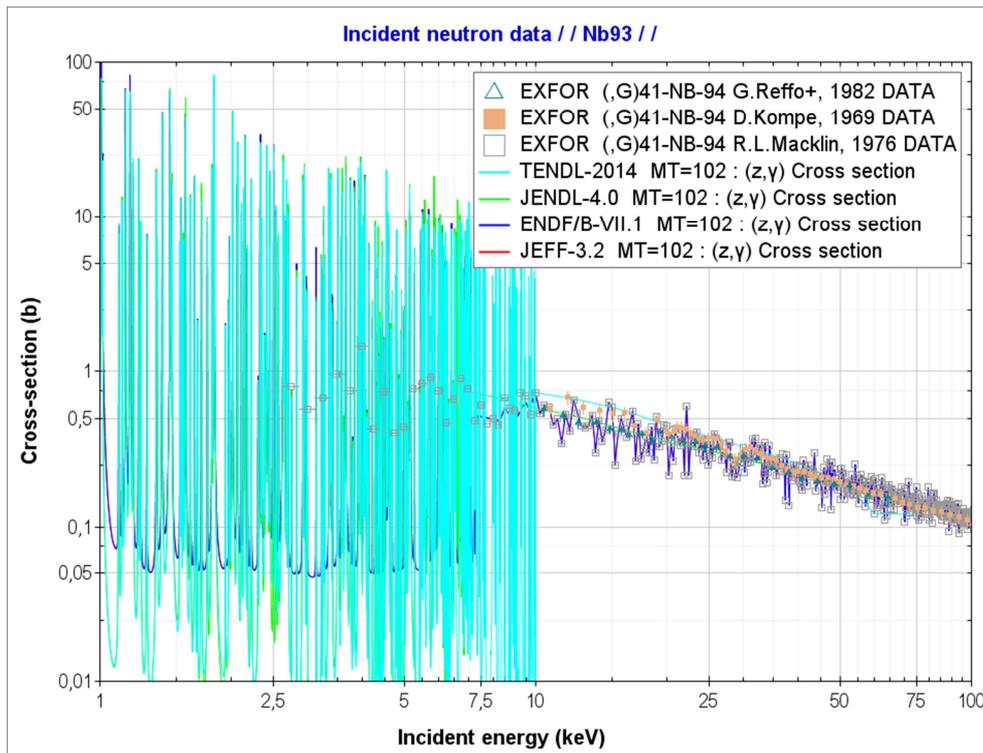


Figure 3.1: $^{93}\text{Nb}(n,\gamma)$ cross section in keV region from different evaluations and experimental data.

2.4 ^{148}Nd

The experimental data that are available in EXFOR for the $^{148}\text{Nd}(n,\gamma)$ resonance integral are listed in Table 4.1. The value of Barry (2006) is the only one in Table 4.1 that was obtained from a time-of-flight capture cross section experiment. It is the most precise and presumably also the most reliable result. The result of Ruiz (1964) is the closest to the JEFF-3.2 evaluation, however discrepant with all other (newer) experimental data and evaluated cross section libraries. All other measurements have been taken into account in the comparison.

In Table 4.2 resonance integrals for the $^{148}\text{Nd}(n,\gamma)$ reaction obtained with different data libraries are compared. The contribution to the resonance integral is significant up to ~ 1 MeV, therefore for this specific case, the upper limit of the calculated RI has been set to $E_{max} = 1$ MeV. There are large discrepancies in RI with different libraries. There are no high-resolution time-of-flight capture measurements available in EXFOR for the energy region of interest. The capture widths of individual resonances can thus be inferred only from capture measurements on natural samples and/or transmission measurements with enriched samples, and both methods can show limitations. **The resonance integral, calculated from JEFF-3.2, on average shows the worst statistical agreement of all compared evaluated libraries** (Figure 4.1). **Furthermore, the evaluation is also very outdated, since the last small modifications were done in 1990 in version JEF-2.2, while the evaluation dates back to version JEF-1 (based on Ref. [6])!** From the remaining evaluations, only JENDL-4.0 is statistically consistent ($\chi^2/n < 1$) with the selection of experimental data. ENDF/B-VII.1 and TENDL-2014 overestimate the RI , however they are much closer to JENDL-4.0/experiments than JEFF-3.2.

Table 4.1: Experimental data for $^{148}\text{Nd}(n,\gamma)$ *RI* from the EXFOR database.

Author	Year	EXFOR Entry	E_{min} [eV]	RI [b]	Accepted
Karadeg	2014	23202.005	0.55	14.6 ± 1.3	Yes
<i>Barry</i>	<i>2006</i>	<i>14093.012</i>	<i>1 (E_{max} = 500 eV)</i>	<i>11.0 ± 0.2</i>	<i>Yes (TOF)</i>
Heft	1978	12866.139	0.5	16.5 ± 3	Yes
Gryntakis	1976	20625.021	0.55	13.77 ± 1	Yes
Steinnes	1975	20635.003	0.5	14.1 ± 1.3	Yes
Van Der Linden	1974	20645.020	0.55	14 ± 0.7	Yes
Riccabarra	1973	30239.003	0.6	11.7 ± 1	Yes
Alstadt	1967	20044.009	0.4	14 ± 2	Yes
Ruiz	1964	12049.002	0.5	18.7 ± 0.5	No

Table 4.2: Comparison of resonance integrals for the $^{148}\text{Nd}(n,\gamma)$ reaction.

	JEFF-3.2	ENDF/B-VII.1	JENDL-4.0	TENDL-2014
RI (> 0.5 eV)	17.02	13.57	11.19	13.68
χ^2/n	30.9	4.88	0.78	5.68
RI (1-500 eV)	17.0	13.6	11.2	13.7

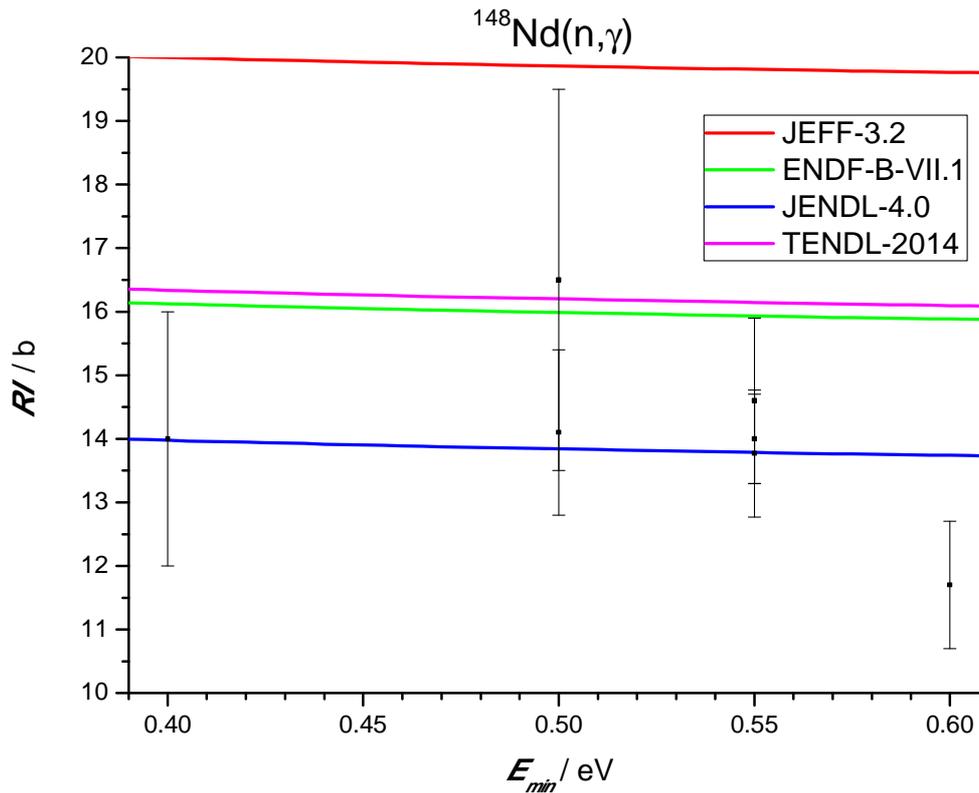


Figure 4.1: $^{148}\text{Nd}(n,\gamma)$ resonance integral RI as a function of lower energy limit E_{min} . JEFF-3.2 (red), ENDF/B-VII.1 (green), JENDL-4.0 (blue), TENDL-2014 (purple), and experimental data (black).

2.5 $^{176,178}\text{Hf}$

The experimental data (available in EXFOR) for the capture resonance integrals in hafnium isotopes $^{176,178}\text{Hf}$ is very limited (Table 5.1). The only available data points were derived by Trbovich (2009) from capture and transmission time-of-flight measurements at RPI in the energy region 0.5-200 eV. These data were included in the evaluation adopted in ENDF/B-VII.1 (^{176}Hf and ^{178}Hf), JENDL-4.0 (^{176}Hf) and TENDL-2014 (^{178}Hf). The resonance parameters in JEFF-3.2 are based on more recent measurements at GELINA [7]. The contribution of the cross section above 200 eV to the resonance integral (Table 5.2) is about 3-4% and <1% for ^{176}Hf and ^{178}Hf , respectively. Both for ^{176}Hf and ^{178}Hf , the contribution of the first resonance (at around 8 eV) dominates the resonance integral and the differences in RI mainly originate from different resonance parameters of the first resonance (Table 5.2). The analysis of the measurements at GELINA excluded the 8 eV doublet as most of the samples were too thick to adequately resolve the doublet. Trbovich's measurements were on liquid samples with sufficiently low number density to resolve the 8 eV doublet. However, Mick Moxon identified an issue with the normalisation of the 8 eV doublet in Trbovich's analysis; reanalysis of the RPI data was reported in Ref. [8] and the reanalysed parameters were adopted for JEFF-3.1.2.¹ **Since JEFF-3.2 is based on newer measurements and complemented by re-analyzed Trbovich (2009) measurements, one could assume that JEFF-3.2 is a more**

¹ T. Ware, private communication.

reliable evaluation. However, additional integral tests would have to be performed in order to clearly confirm that statement.

Table 5.1: Experimental data for $^{176,178}\text{Hf}(n,\gamma)$ RI from the EXFOR database.

Author	Year	Isotope	EXFOR Entry	E_{min} [eV]	RI [b]	Accepted
Trbovich	2009	^{176}Hf	14239.009	0.5	692 ± 2	Yes
Trbovich	2009	^{178}Hf	14239.011	0.5	1872 ± 4	Yes

Table 5.2: Comparison of resonance integrals for the $^{176,178}\text{Hf}(n,\gamma)$ reaction.

		JEFF-3.2	ENDF/B-VII.1	JENDL-4.0	TENDL-2014
	RI (> 0.5 eV)	633	693	698	526
^{176}Hf	RI (< 200 eV)	613	676	676	504
	RI (~8 eV res.)	518	585	585	421
	RI (> 0.5 eV)	1799	1872	1915	1871
^{178}Hf	RI (< 200 eV)	1784	1859	1902	1859
	RI (~8 eV res.)	1773	1849	1893	1849

3. Conclusion

The quality of the evaluated nuclear data library JEFF-3.2 has been assessed in comparison to other contemporary major evaluated nuclear data libraries for a selection of nuclides. Cross sections and resonance integrals for neutron capture reactions on these nuclides have been compared. The conclusions for individual materials can be summarized as follows:

- ^{55}Mn : Qualitatively and quantitatively (statistically), JEFF-3.2 performs better compared to other libraries. A strong recommendation to stick with the JEFF-3.2 evaluation for the next (future) version of the JEFF library can be expressed.
- ^{58}Fe : JEFF-3.2 shows better statistical agreement with integral experiments (i.e. measurements of the capture resonance integral), however their accuracy and reliability is limited, added to lack of high-resolution TOF capture measurements. No definite conclusions can be drawn, however JEFF-3.2 stays the recommended library.
- ^{93}Nb : Very unreliable and scattered integral experiments, however JEFF-3.2 has worse statistical agreement with them compared to JENDL-4.0, TENDL-2014 and IRDFF. Re-evaluation or adoption of the evaluation from JENDL-4.0 is to be considered for the next version of the JEFF library.
- ^{148}Nd : JEFF-3.2 has the worst statistical agreement with integral experiments, the evaluation is also the most outdated (adopted from JEF-1/Mug-81 with minor modifications) compared to other major nuclear data libraries. Re-evaluation is necessary! From the existing libraries, JENDL-4.0 is the recommended evaluation.
- $^{176,178}\text{Hf}$: Lack of integral experimental data. JEFF-3.2 is based on various high-resolution TOF transmission and capture measurements. The differences in RI mainly originate from differences of the parameters of the first resonance. JEFF-3.2 is based on newer measurements at GELINA which were found to agree with a re-analysis (Moxon) of the Trbovich (2009) data. For lack of independent good quality RI data JEFF-3.2 stays the recommended library.

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List of abbreviations and definitions

EXFOR	Experimental Nuclear Reaction Data
ENDF/B	Evaluated Nuclear Data File, format B
GELINA	GEel LINear Accelerator
IRDF	International Reactor Dosimetry and Fusion File
JEFF	Joint Evaluated Fission and Fusion file
JENDL	Japanese Evaluated Nuclear Data Library
OECD/NEA	Organisation for Economic Co-operation and Development / Nuclear Energy Agency
<i>RI</i>	Resonance Integral
RPI	Rensselaer Polytechnic Institute
TENDL	TALYS-based Evaluated Nuclear Data Library
TOF	Time-Of-Flight

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