30th NENE 2021
September 6 - 9
BLED-SLOVENIA
International Conference Nuclear Energy for New Europe

Burnup-dependent isotopic compositions of PWR fuel pins using OpenMC and WIMS with ENDF/B-VIII nuclear data library

Jan Malec, Dušan Čalič, Andrej Trkov
Jožef Stefan Institute
Jamova cesta 39
1000, Ljubljana, Slovenija
jan.malec@ijs.si, dusan.calic@ijs.si, andrej.trkov@ijs.si

ABSTRACT

The OpenMC Monte Carlo transport code has been identified as a possible candidate for generating homogenized cross sections for the CORD-2 core design package. In this work we present a comparison between OpenMC, Serpent and WIMS-D calculations on a burnup example for the central cell in a 3x3 cell geometry. A new WIMS-D library with 191 nuclides based on data from the ENDF/B-VIII.0 library was created. The differences in isotopic compositions calculated with Serpent, OpenMC and WIMS-D were analyzed. The newly created WIMS-D library has helped to narrow down many differences in the calculated burnup-dependent isotopic compositions, but some differences remain. A method for calculating the multiplication factor from cross sections based on two group diffusion approximation was derived to bring reactivity estimates by different codes on the same basis. OpenMC proved to be a promising method for generating homogenized cross sections for reactor core design calculations within the CORD-2 system.

1 INTRODUCTION

Core design calculations based on deterministic methods can be performed in several stages. The CORD-2 core design package [1] for pressurized water reactors uses three stages, the first of which is a fuel cell calculation including the nearest neighbours, making a 3x3 cell grid with periodic boundary conditions, the second is a fuel assembly level calculation with critical albedo boundary condition [2]. Both stages include the effective diffusion cross section homogenisation (EDH) [3] that preserves partial currents on the boundary of the region of interest. The third stage is a global calculation of the entire reactor core. The first stage is performed with the WIMSD-5B transport code [4], which calculates homogenized cross sections for the central cell in a 3x3 cell geometry and the burnup-dependent isotopic composition for the entire fuel lifetime. The isotopic composition of the fuel cell is exported to a text-based ISOlib library, which is interpolated by the CORD-2 package to estimate the fuel isotopic composition under different average conditions during burnup, such as enrichment, moderator density, fuel temperature and boron concentration. The possibility of replacing the deterministic calculation with a stochastic calculation using the Serpent code has been extensively explored in the past [5] and provided promising results despite significantly longer calculation times [2]. Another option would be to use the OpenMC code [6], which is released as open-source software and therefore suitable for integration into other core design packages.

The present paper presents code comparison of three cell-cluster calculations in a 3x3 grid to compare not only the differences between the results obtained with a stochastic and a deterministic method, but also between two stochastic methods. The calculations were performed using the ENDF/B-VIII.0 [7] library. A new WIMS-D library was created using methods based on the WLUP project [8]. The new WIMS-D library improves our understanding of the differences between the calculated isotopic compositions using different codes and enable us to perform further WIMS-D calculations with nuclear data obtained from a modern nuclear data library.

1.1 Problem definition

The inter-comparison focuses on a single burnup case with a 3x3 lattice of fuel elements without any in-fuel burnable absorbers and a pitch of $1.243\,80\,\mathrm{cm}$. The geometry is displayed on Figure 1.

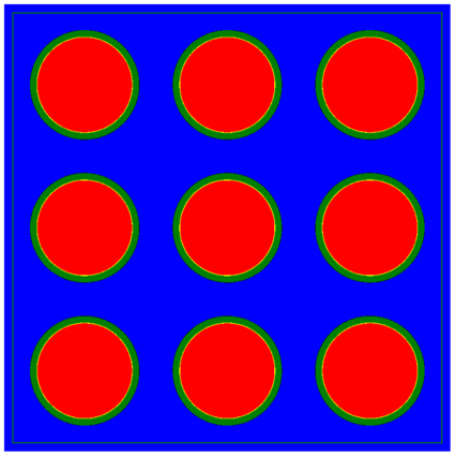


Figure 1: Geometry of the burnup calculation.The fuel is colored in red, helium in yellow, zirconium in green, water is blue and the black circle surrounding the elements is the cladding.

Parameters such as fuel and moderator temperatures, enrichment, boron concentration and power density were chosen to mimic a scenario at the Krško Nuclear Power Plant with high fuel enrichment and relatively low boron concentration. The exact starting material compositions and fuel radii were calculated using the WICORD code, which is part of the core design package CORD-2, taking into account the average core conditions during burnup, as well as fuel thermal expansion.

Table 1: Nominal burnup calculation parameters

Quantity	Value
Fuel mass (HM)	$420.0\,\mathrm{kg}$
Fuel enrichment	5.2%
Fuel temperature	$1000.0\,\mathrm{K}$
Moderator temperature	$595.0\,\mathrm{K}$
Moderator density	$0.6750\,\mathrm{g\,cm^{-3}}$
Boron concentration	$300.0\,\mathrm{ppm}$
Power density	$46.3476\,\mathrm{MW\,t^{-1}HM}$

2 PREPARATION OF NUCLEAR DATA LIBRARIES

The two stochastic codes OpenMC and Serpent track all nuclides for which nuclear data are available. For Serpent calculations the decay chains were automatically constructed from the nuclear data files using default settings. OpenMC provides decay chains in xml format for some configurations, but not for the ENDF/B-VIII.0 library. A decay chain was generated using the built-in routine, but some decay chains were missing capture reactions to excited states, which were added later. The missing capture reactions are present in the ENDF/B-VII.1 based library published on the OpenMC website, which is derived from the standard Serpent library. Ultimately, the same branching ratios were adopted for the ENDF/B-VIII.0 based OpenMC library for consistency.

Preparing nuclear data libraries for deterministic codes is usually a more involved process, and the WIMS-D libraries are no exception. As a starting point, the NJOY inputs for ENDF/B-VII.0 from the WLUP update project [8] were updated to work with the ENDF/B-VIII.0 library. These inputs contain a wider range of materials and temperatures up to $2000\,\mathrm{K}$ compared to the original set described in the WLUP documentation. The first change was the addition of the isotopes ^{50}V, ^{51}V, ^{12}C, and ^{13}C, since natural compositions for vanadium and carbon are no longer available in the ENDF/B-VIII.0 library.

All libraries were used to create ISOlib files, which are text-based databases of burnup-dependent isotopic compositions for the 3x3 test case. The resulting ISOlib databases were compared with the ISOPLT program of CORD-2 that produces plots of burnup-dependent isotopic compositions for all tracked nuclides and a comparison of multiplication factors. Differences in isotopic compositions were examined on a case-by-case basis. Some arise from the fact that the WIMS-D library does not track as many nuclides as the stochastic codes. In some cases it was necessary to add nuclides to complete the missing reaction pathways. In this way, the number of nuclides in the WIMS-D library was increased from 185 to 191. The complete list of nuclides added to the WIMS-D library is given in the Table 2. ^{159}Gd has no capture product because ^{160}Gd is not important and is implicitly included in the pseudo-fission product material. ^{158}Gd was defined as a capture product of ^{157}Eu because ^{158}Eu has a relatively short half-life of 49.5 min and decays into ^{158}Gd.

Table 2: New isotopes added to the WIMS-D library with their respective capture and decay products

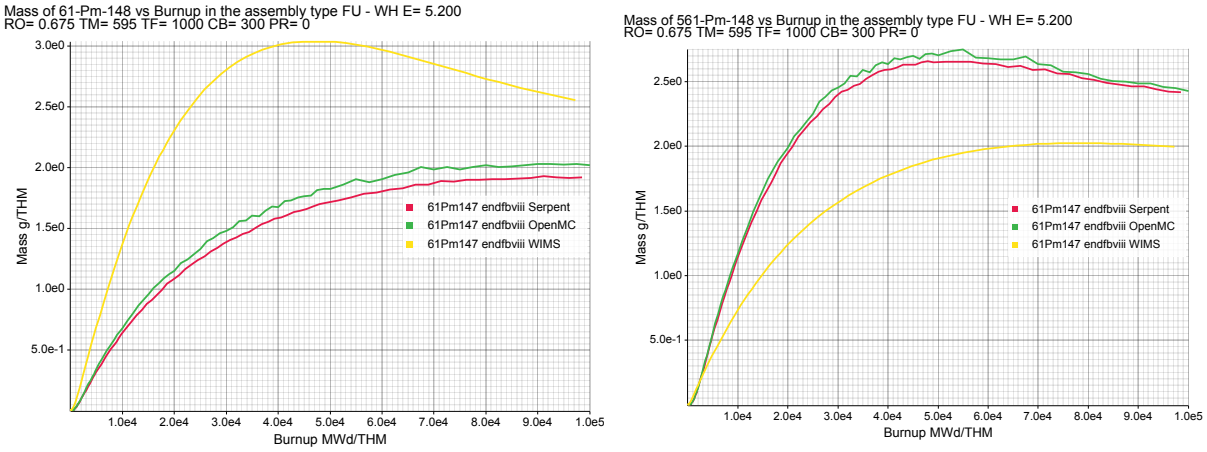
New nuclide	Capture product	Decay product
^{156}Eu	^{157}Eu	^{156}Gd
^{157}Eu	^{158}Gd	^{157}Gd
^{159}Gd	Void	^{158}Gd
^{159}Dy	^{160}Dy	^{158}Gd
^{159}Tb	^{160}Tb	Stable
^{160}Tb	^{161}Dy	^{160}Dy

The isotopic composition of the fuel in the central element of the lattice was followed up to 10^5 MWd/tHM (megawatt-days per tonne of heavy metal) and compared both on an absolute scale and as a ratio. Special attention was paid to isotopes that have two capture products, since the WIMS-D library format does not provide a way to enter more than one capture product and approximations are used to simulate them. Two of these products are ^{147}Pm and ^{241}Am. Fission yield of ^{147}Pm is split into two fractions according to the branching ratio forming two nuclides with identical cross sections, one of which transforms through capture to ^{148g}Pm and the other

to ^{148m}Pm. Figure 2 shows the burnup-dependent concentration of the two ^{148}Pm nuclides. It can be seen that the WIMS-D calculation leads to a higher concentration of ^{148g}Pm and a lower concentration of ^{148m}Pm than either of the stochastic codes, mainly due to differences in the assumed branching ratios. The branching ratios are in principle energy-dependent. The values at thermal and at 2 MeV from different evaluated data libraries are listed in Table 3. None of the major libraries provides the branching ratios, except TENDL-2019 [9]. The value adopted in the WIMS-D library is from the Atlas of Neutron Resonances [10].

Table 3: Branching ratios on capture in ^{147}Pm for the production of ^{148g}Pm in different evaluated nuclear data libraries.

Thermal	2 MeV	Library
0.43	-	Atlas [10]
0.533	0.742	EAF-2010 [11]
0.533	0.545	JEFF-3.1/A [12]
0.570	0.633	TENDL-2019 [9]



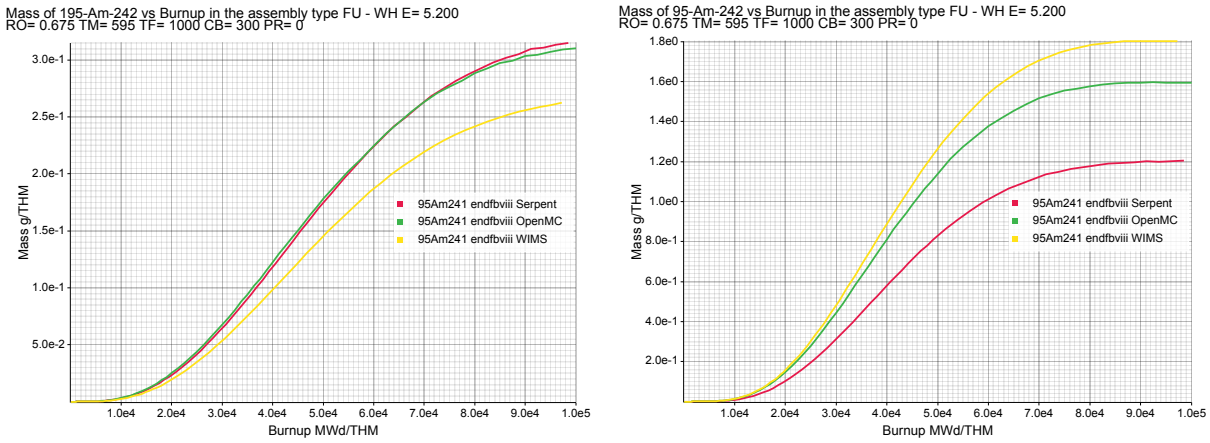
(a) Ground state

(b) Excited state

Figure 2: The WIMS calculation produced more ^{148}Pm in the ground state and less ^{148}Pm in the excited state

A different approximation is used in the WIMS-D library to generate the ground and the excited states of ^{242}Am. Since WIMS-D does not accept two capture products for one nuclide, the less-important ^{242}Am is treated as a fission product. The fission cross section of ^{241}Am is replaced by the scaled capture cross section so that the capture to fission ratio of a typical thermal reactor is preserved. The effective "fission yield" is the capture to fission ratio so as to approximately reproduce the ^{242g}Am production.

The production of ^{242m}Am is defined in the usual way by capture in ^{241}Am, taking the branching ratio into account. The Approximation is validated by correct production of ^{243}Am. The concentration of ^{242g}Am predicted by OpenMC, Serpent and WIMS are similar, but Serpent calculation results in a lower concentration of ^{242m}Am because the default branching ratio for Serpent was taken from the JEFF-3.1 data library [12]. The branching ratio in JEFF-3.1 at thermal is 0.081, while the ENDF/B-VIII.0 gives 0.10 at thermal and 0.43 at 2 MeV. The spectrum-averaged branching ratio is thus close to 0.11, adopted in the WIMS-D library. In the OpenMC calculation the branching ratio of 0.13 was used.



(a) Ground state

(b) Excited state

Figure 3: Burnup dependant concentrations of ^{242}Am nuclides

Some differences exist also among nuclides with one capture product, ^{95}Mo has around 10% higher concentration when computed with WIMS-D compared to the other two codes and the concentrations of ^{109}Ag computed with WIMS-d are around 10% lower. With ^{115}In, ^{125}Sb, ^{127}I and ^{113}Cd the OpenMC and WIMS-D calculated concentrations match within a few percent, but the Serpent calculated concentrations differ by more than 50%. The ^{135}I concentration matches between WIMS-D and Serpent mid-cycle, but OpenMC derived concentration is off by about 5%. The concentrations of ^{147}Pm and ^{147}Sm match between all codes up to the middle of the fuel life, but diverge afterwards due to slower build-up in WIMS-D. The WIMS-D derived concentrations of ^{151}Sm, ^{152}Sm, ^{151}Eu, ^{152}Eu differ from the ones calculated with OpenMC or Serpent by approximately 25%. The lighter actinides ^{232}Th, ^{233}Pa, ^{232}U and ^{233}U are currently not calculated in WIMS-D because they are produced through (n,2n) reactions, which are not explicitly present in a WIMS-D library.

3 MULTIPLICATION FACTOR COMPARISON

In addition to isotopic compositions, ISOlib libraries include a burnup-dependent multiplication factor. The multiplication factor is a useful integral quantity because it depends on several cross sections, although a suitable multiplication factor does not guarantee the correctness of the cross sections. Since the lattice problem was computed with periodic boundary conditions, the multiplication factor k is equal to k_{inf}. Despite its simple definition as the ratio between two successive neutron generations, not all transport programs calculate the multiplication factor in the same way. Differences in the way the neutron multiplication factor is accounted for can result in differences of up to 0.4% in k_{eff} when the same problem is computed using different Monte-Carlo codes [13]. Another problem when comparing the multiplication factor is that only the multiplication factor of the central element of the 3x3 grid needs to be extracted. This is easily accomplished in WIMS-D and Serpent, but OpenMC does not provide a well-documented way to do this. For these two reasons, we derived a common method for computing the multiplication factor based on the homogenized effective cross sections computed by stochastic codes.

$$
\begin{aligned}
-D_1\nabla^2\phi_1 + \Sigma_1\phi_1 &= \ 1/k\,(\nu_1\Sigma_f 1\phi_1 + \nu_2\Sigma_f 2\phi_2) + \ \ \Sigma_{\ (2\to1)}\phi_2 \\
-D_2\nabla^2\phi_2 + \Sigma_2\phi_2 &= \Sigma_{\ 1\to2}\phi_1 \\
\Sigma_1 &= \Sigma_{a1} + \Sigma_{1\to2} \\
\Sigma_2 &= \Sigma_{a2} + \Sigma_{2\to1}
\end{aligned}
\tag{1}
$$

The multiplication factor was derived from the two-group diffusion equation with both up-scattering and down-scattering and assuming that all neutrons are born in the fast energy group. They are written out in eq. (1), where D is the diffusion coefficient, Σ_a, Σ_f and $\Sigma_{\ 1\to2}$ are absorption, fission and scattering cross sections and ϕ is the neutron flux. The equations for an infinite medium can be written out in matrix form (2) and solved for k when the matrix determinant equals zero (3).

$$
\begin{bmatrix} \Sigma_1 - \frac{1}{k}\left(\nu_1\Sigma_{f1}\right) & \frac{1}{k}\left(\nu_2\Sigma_{f2}\right) + \Sigma_{2\to1} \\ \Sigma_{1\to2} & \Sigma_2 \end{bmatrix} \begin{bmatrix} \phi_1 \\ \phi_2 \end{bmatrix} \tag{2}
$$

$$
k = \frac{\Sigma_2\nu_1\Sigma_{f1}\phi_1 + \nu_2\Sigma_{f2}\phi_2\Sigma_{1\to2}}{\Sigma_1\Sigma_2 - \Sigma_{1\to2}\Sigma_{2\to1}} \tag{3}
$$

The resulting multiplication factors match within their statistical uncertainty for all three compared codes when normalized to 500 MWd/tHM burnup. When compared on the absolute scale, the WIMS calculation shows a consistent offset of 4% compared to Serpent and OpenMC results, which is about 1100 pcm (parts per 100 000) at zero burnup. The multiplication factors from OpenMC and Serpent calculations are within 100 pcm. (Figure 4). The results in [2] report a smaller difference in neutron multiplication factor calculated with WIMS and Serpent for a problem with similar geometry with fresh fuel, but the calculations are not directly comparable because of a slightly tuned model for self-shielding.

4 CONCLUSION

Thanks to its open source nature, a free license and an active developer community, OpenMC offers an attractive option for integration into core design packages such as CORD-2. We used OpenMC to generate burnup-dependent isotopic compositions of fuel in a 3x3 grid with periodic boundary conditions, and compared it to similar libraries generated using Serpent and WIMS-D. In addition to the isotopic composition, the multiplication factor of the central fuel cell was also compared. This quantity is calculated directly by WIMS-D, but had to be estimated for both stochastic calculations from homogenized cross sections for the central cell using a two-group formula for k_{inf} derived from the two-group diffusion approximation. Since agreement between three transport and burnup codes is not a guarantee of correctness, the goal of the comparison was not to achieve complete agreement of the results but to identify and understand the differences. Based on the results, a new WIMS-D library was created based on the ENDF/B-VIII.0 library with 196 nuclides. Burnup calculations with the new WIMS-D library still lead to some differences that need to be investigated in the future. There are many reasons for such differences, such as transport approximations in the WIMS-D code, missing reaction paths, a smaller number of nuclides tracked in WIMS-D, differences in the treatment of neutron mutliplication reactions, and the treatment of branching reactions. The work presented in this paper will enable us to use the new WIMS-D library based on the latest nuclear data from the ENDF/B-VIII.0 library in nuclear design calculations and provide us with a new alternative for generating homogenized effective cross sections with OpenMC.

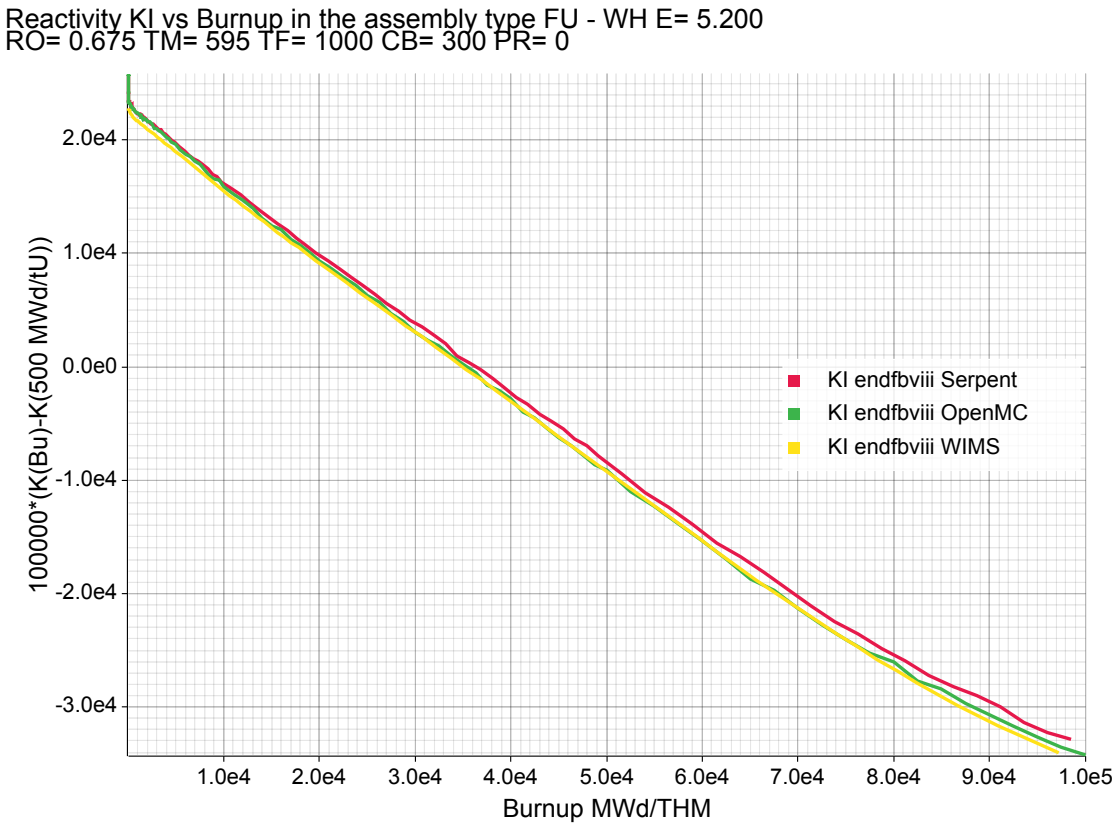


Figure 4: Comparison of neutron multiplication factor in units of pcm $(10^5(k-1))$.

ACKNOWLEDGMENTS

The authors acknowledge the financial support from the Slovenian Research Agency (research core funding No.P2–0073 Reactor physics and the project L2-2612 Stability of nuclear reactors in load follow mode of operation)

REFERENCES

[1] Andrej Trkov. *CORD-2 package user's guide*, volume 6948 of *IJS delovno poročilo*. "Jožef Stefan" Institute, 1997.

[2] Dusan Calic, Andrej Trkov, Marjan Kromar, and Luka Snoj. Use of effective diffusion homogenization method with the monte carlo code for light water reactor. *Annals of Nuclear Energy*, 94:392–398, 2016.

[3] A. Trkov and M. Ravnik. Effective diffusion homogenization of cross sections for pressurized water reactor core calculations. *Nuclear Science and Engineering*, 116(2):86–95, 1994. Publisher: Taylor & Francis.

[4] J R Askew, F J Fayers, and P B Kemshell. GENERAL DESCRIPTION OF THE LATTICE CODE WIMS. *The Journal of British Nuclear Energy society*, 1966.

[5] Jaakko Leppänen, Maria Pusa, Tuomas Viitanen, Ville Valtavirta, and Toni Kaltiaisenaho. The serpent monte carlo code: Status, development and applications in 2013. *Annals of Nuclear Energy*, 82:142–150, 2015.

[6] Paul Romano and Sterling Harper. Nuclear data processing capabilities in OpenMC. *EPJ Web of Conferences*, 146:06011, 2017.

[7] D. A. Brown, M. B. Chadwick, R. Capote, A. C. Kahler, A. Trkov, M. W. Herman, A. A. Sonzogni, Y. Danon, A. D. Carlson, M. Dunn, D. L. Smith, G. M. Hale, G. Arbanas, R. Arcilla, C. R. Bates, B. Beck, B. Becker, F. Brown, R. J. Casperson, J. Conlin, D. E. Cullen, M. A. Descalle, R. Firestone, T. Gaines, K. H. Guber, A. I. Hawari, J. Holmes, T. D. Johnson, T. Kawano, B. C. Kiedrowski, A. J. Koning, S. Kopecky, L. Leal, J. P. Lestone, C. Lubitz, J. I. Márquez Damián, C. M. Mattoon, E. A. McCutchan, S. Mughabghab, P. Navratil, D. Neudecker, G. P. A. Nobre, G. Noguere, M. Paris, M. T. Pigni, A. J. Plompen, B. Pritychenko, V. G. Pronyaev, D. Roubtsov, D. Rochman, P. Romano, P. Schillebeeckx, S. Simakov, M. Sin, I. Sirakov, B. Sleaford, V. Sobes, E. S. Soukhovitskii, I. Stetcu, P. Talou, I. Thompson, S. van der Marck, L. Welser-Sherrill, D. Wiarda, M. White, J. L. Wormald, R. Q. Wright, M. Zerkle, G. Žerovnik, and Y. Zhu. ENDF/b-VIII.0: The 8th major release of the nuclear reaction data library with CIELO-project cross sections, new standards and thermal scattering data. *Nuclear Data Sheets*, 148:1–142, 2018.

[8] Kemshell P B Askew J R, Fayers F J. *WIMS-D Library Update*. Non-serial Publications. INTERNATIONAL ATOMIC ENERGY AGENCY, 2007.

[9] A. J. Koning, D. Rochman, J. Ch. Sublet, N. Dzysiuk, M. Fleming, and S. van der Marck. TENDL: Complete nuclear data library for innovative nuclear science and technology. *Nuclear Data Sheets*, 155:1–55, 2019.

[10] Said F. Mughabghab Ph D. MSc BSc. *Atlas of Neutron Resonances: Volume 1: Resonance Properties and Thermal Cross Sections Z= 1-60*. Elsevier Science, 6th edition edition, 2018.

[11] EASY Documentation Series, LW Packer, and J-Ch Sublet. The european activation file: EAF-2010 decay data library. Technical report, EURATOM/CCFE Fusion Association, Culham Science Centre, 2010. https://t2.lanl.gov/nis/data/jeff/EAF2010.pdf.

[12] Arjan Koning, Robin Forrest, Mark Kellett, Robert Mills, Hans Henriksson, Yolanda Rugama, O. Bersillon, O. Bouland, A. Courcelle, M.C. Duijvestijn, E. Dupont, J. Kopecky, D. Leichtle, F. Marie, M. Mattes, E. Menapace, B. Morillon, C. Mounier, G. Noguerre, P. Pereslavtsev, P. Romain, O. Serot, S. Simakov, S. Tagesen, H. Vonach, P. Batistoni, P. Bem, F. Gunsing, M. Pillon, A. Plompen, P. Rullhusen, K. Seidel, M. Avrigeanu, V. Avrigeanu, E. Bauge, H. Leeb, M.J. Lopez Jimenez, D. Bernard, A. Bidaud, R. Dagan, C. Dean, P. Dos-Santos-Uzarralde, U. Fischer, A. Hogenbirk, R. Jacqmin, C. Jouanne, I. Kodeli, J. Leppanen, S.C. van der Marck, R. Perel, R. Perry, M. Pescarini, A. Santamarina, J.C. Sublet, A. Trkov, M.M. Be, T.D. Huynh, M.A. Kellett, R. Mills, A. Nichols, H. Henriksson, C. Nordborg, A. Nouri, Y. Rugama, and E. Sartori. The JEFF-31 nuclear data library - JEFF report 21. Technical Report 92-64-02314-3, Nuclear Energy Agency of the OECD (NEA), 2006. NEA–6190.

[13] Dermott E. Cullen, Roger N. Blomquist, Chris Dean, Dave Heinrichs, Mikhail A. Kalugin, Mark Lee, Yi-Kang Lee, Robert MacFarlane, asunobu Nagaya, and Andrej Trkov. How accurately can we calculate thermal systems? Technical Report UCRL-TR-203892, IAEA, 2004.