The Effect of Nuclear Data Discrepancies on Criticality Simulations of Molten Salt Fast Reactors

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INTRODUCTION

The purpose of this paper is to study the effects of nuclear data discrepancies on the effective multiplication factor (k_{eff}) calculations and to understand the reactor physics contributing to k_{eff} differences. The Molten Salt Fast Reactor (MSFR) was chosen for this study due to its diverse considerations of fuel compositions with fluoride and chloride for its unique molten salt. By considering many isotopes, it will allow for a more in-depth investigation of data differences. Another reason chloride salt was chosen in the study was because chloride is known to have a lack of well-studied cross section data needed for reactor simulations in TerraPower's Molten Chloride Fast Reactor (MCFR).

To understand the changes in criticality simulations due to data differences, it is important to understand which isotopes have the most difference between libraries and which isotopes the system is most sensitive to. A further investigation on these relevant isotopes into their neutron flux, energy spectra, reaction rates, and sensitivity between libraries will provide a more complete understanding of the physics effecting differences in k_{eff} calculations

NJOY2016 was used to investigate discrepancies in nuclear data libraries. These libraries were used in the Serpent modeling of the reactor and fuel to perform an in-depth analysis on the cause of k_{eff} variations .

NUCLEAR DATA

ENDF Libraries

The Evaluated Nuclear Data Files (ENDF) library is the main library of nuclear data used in computational reactor modeling [1]. Their data is the collective compilation result of reported nuclear data from different scientists and institutions. Every few years updated versions of these libraries will be released with new nuclear data identified and included. Some isotopes have small variability between libraires while some have high differences. This is due to the isotope's relevance in nuclear and the demand for data on a given isotope.

Data Discrepancies

The two latest releases of the ENDF libraries, namely ENDF/B-VII.1 and ENDF/B-VIII.0, are the focus for data selected to perform the comparison in this study. In these libraries the most prominent and relevant isotopes to MSFR were investigated. This includes salt bases, fluoride and chloride, along with the fissile and other prominent fissionable isotopes.

To visualize the discrepancy in cross sections among different libraries, the data was condensed into the groupwise format. Using NJOY2016 and its GROUPR module, ENDF data files can be processed into a groupwise format. The GROUPR module calculates averages of pointwise cross sections over multigroup matrices describing the transfer of neutrons from one group to the next. Fig. 1 shows the groupwise total cross section of ^{235}U yielded from both libraries. Here the 239-group LANL energy group structure was used in this view [2].

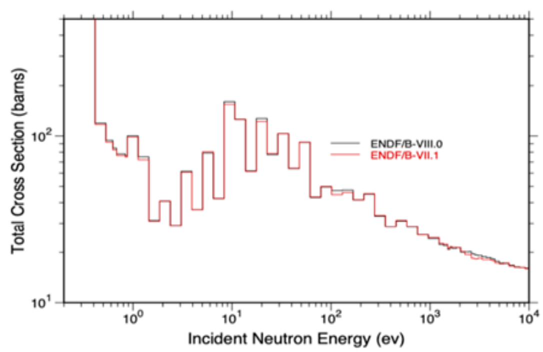

Fig. 1. Comparison of ENDF/B-VIII.0 and ENDF/B-VII.1 data libraries of ^{235}U zoomed in on the resonance range.

As shown in Fig. 1, noticeable difference between libraries was seen in the fissionable isotopes like ^{235}U. It is noteworthy that the fluoride and chloride isotopes showed no difference between the libraires. This is expected since they are not highly investigated isotopes in the nuclear field.

MSFR REACTOR MODEL

The MARS and EVOL projects offer an in-depth collaborative study between Russia and several European countries on the MSFR concept. Their reactor design laid the basis of this study [3]. A simplified MSFR design was created

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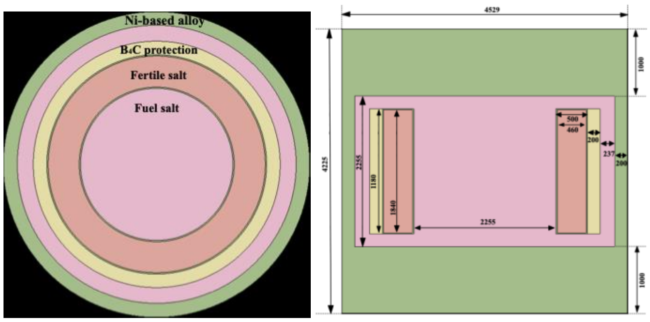
in Serpent, a Monte Carlo based transport code [4] for reactor analysis. Only the reflector, blanket, and B_4C shield were considered in the model. Fig. 2 sketches the reactor model.



(a) (b)

Fig. 2. Radial (a) and axial (b) view of the reactor core.

Material Composition

The fertile blanket in this reactor is designed to be a radial reflector, along with improving breeding capabilities. The neutron shielding is on the outer wall of the fertile blanket and is composed of B_4C. The top and bottom walls of the core are made up of NiCrW Hastelloy material. These walls act as neutron reflectors. The B_4C layer is the protection on the exterior of the fertile blanket and acts as the neutron shielding and protection for the heat exchangers. The Ni-based alloy walls act as neutron reflectors. All materials are assumed with a temperature of 973K in all calculations. The compositions for the shielding and reflectors are summarized in Table I.

Table I. Molar composition and density of core materials.

Material	Molar Composition	Density [g/cm^3]
Shield	B (80%), C (20%)	2.52
Reflector	Ni (79.432%), W (9.976%), Cr (8.040%), Mo (0.736%), Fe (0.632%), Ti (0.295%), C (0.294%), Mn (0.257%), Si (0.252%), Al (0.052%), B (0.033%), P (0.023%), S (0.004%)	10.0

The two fuels that we are considering are chloride-based fuel, similar to what TerraPower proposed, and the fluoride-based fuel, which is an iteration of the MARS and EVOL project [5]. Both salt bases are considered with three different fissionable fuel compositions. The iteration from the MARS EVOL project for the fluoride-based fuel is shown in Table II, and the chloride-based fuel is shown in Table III.

Table II. Molar composition and density of fluoride fuels.

Material	Molar Composition	Density [g/cm^3]
^{233}U -Fluoride	LiF (77.5%) ThF_4 (19.895%) $^{233}UF_4$ (2.515%)	4.16
TRU-Fluoride	LiF (77.5%) ThF_4 (16.068%) $TRUF_3$ (6.432%)	4.27
TRU/ U^{enr} -Fluoride	LiF (77.5%) ThF_4 (6.6%) U^{enr} F_4 (12.3%) $TRUF_3$ (3.6%)	4.30
Fertile salt	LiF (77.5%) ThF_4 (22.5%)	4.15

Table III. Molar composition and density of chloride fuels.

Material	Molar Composition	Density [g/cm^3]
^{233}U -Chloride	NaCl (50%) $ThCl_4$ (42.3%) $^{233}UCl_3$ (7.7%)	3.01
TRU-Chloride	NaCl (50%) $ThCl_4$ (37.5%) $TRUCl_3$ (12.5%)	3.16
TRU/ U^{enr} -Chloride	NaCl (50%) $ThCl_4$ (20.8%) U^{enr} Cl_3 (21%) $TRUCl_3$(8.2%)	3.26
Fertile salt	NaCl (70%) $ThCl_4$ (30%)	2.65

RESULTS

Criticality Analysis

The most important neutronics parameter in gauging the effects of data discrepancies is changes in k_{eff}, an integral factor demonstrating many physics reactions, so changes in k_{eff} would also indicate changes in other physics parameters.

Using Serpent, the MSFR was modelled with the six fuel compositions respectively and the k_{eff} of each model was calculated and summarized in Table IV. It is important to note that there was no comparison done against the model that this paper references. However, our results from Serpent have been cross verified by OpenMC using the same reactor model. It is also noteworthy that not all k_{eff} values are in ideal proximity to criticality, but this is irrelevant since we are only looking at the difference in k_{eff}.

Of the six fuels that were considered, the TRU/U^{enr}-Chloride fuel (referred to as Fuel 6 hereafter) showed the largest difference in k_{eff} when simulated with the different libraries. The remainder of this study will dive into the reactor physics parameters or specific isotopes that contribute to the difference in k_{eff}, focusing on the reactor loaded with Fuel 6.

Table IV. Comparison of the k_{eff} of six fuel compositions.

Fuel	ENDF/B-VII.1	ENDF/B-VIII.0	Absolute Difference in[pcm]
^{233}U -Fluoride	0.927006 ±0.00004	0.94256 ±0.00004	-1555.4
TRU-Fluoride	0.994779 ±0.00004	0.993006 ±0.00004	177.3
TRU/ U^{enr} -Fluoride	1.04199 ±0.00004	1.0439 ±0.00004	-191.0
^{233}U -Chloride	0.976869 ±0.00008	0.981182 ±0.00008	-431.3
TRU-Chloride	1.08409 ±0.00007	1.08876 ±0.00008	-467.0
TRU/ U^{enr} -Chloride	0.974182 ±0.00007	1.01372 ±0.00007	-3953.8

Neutron Energy Spectra

The neutron energy spectra of the reactor were first examined following the k_{eff} comparisons. Neutron energy spectra give good indications on what type of events occur in the reactor such as fission, capture, and so on. Therefore, the neutron spectra have an influence on k_{eff}, and therefore it is relative to understand its difference between libraries. Fig. 3 shows the 252 group neutron energy spectra of the Fuel 6 fueled reactor with two different cross section libraries used.

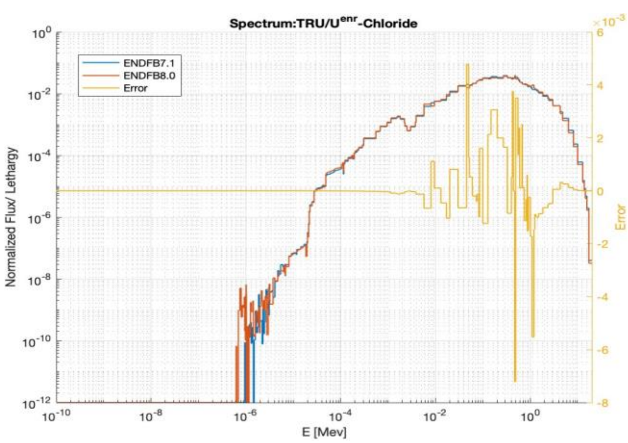


Fig. 3. Flux spectra of ENDF/B-VII.1 vs. ENDF/B-VIII.0.

Integral Reaction Rates

As noticeable differences are shown in the neutron spectra between libraries, it is necessary to see how this affects the integral reaction rates in the core. Since the main difference appears at the higher end of the spectrum, the difference in fission and capture cross section will be of special interest as these reactions are highly correlated to those energy ranges. Table V shows the key reaction rates calculated by Serpent. Note the values are normalized to a single neutron particle.

Table V. Integral reactor rates of the core.

Integral Reaction Rate	ENDF/B-VII.1	ENDF/B-VIII.0	Relative % Difference
Fission	0.34468	0.358892	-4.12
Absorption	0.998896	0.998558	0.03
Capture	0.654216	0.639667	2.22
Leakage	0.001103	0.001440	-30.5

Sensitivity Analysis

The last part in understanding the physics is to examine the k_{eff} sensitivity of cross section reactions of isotopes. Serpent performs sensitivity calculations using generalize perturbation theory based on neutron collision-history tracking. Summing the accepted events (what happens) and the rejected events (what would have happened if the cross sections were changed) the sensitivity of a response to small perturbations can be calculated. In this case our response is k_{eff} and our perturbations are cross section reactions [4].

This sensitivity analysis informs which isotopes, and their respective cross section reaction, have the largest impact on changing k_{eff}. For our case all isotopes in Fuel 6 were simulated. A further sensitivity analysis was done the determine how the energy spectra (using an 8-group structure) for each reaction contributed to the sensitivity. Fig. 4 shows the sensitivity indices of k_{eff} on different reaction types, which reveals the fission and capture reaction rates are the main contributor to changes in the k_{eff} value.

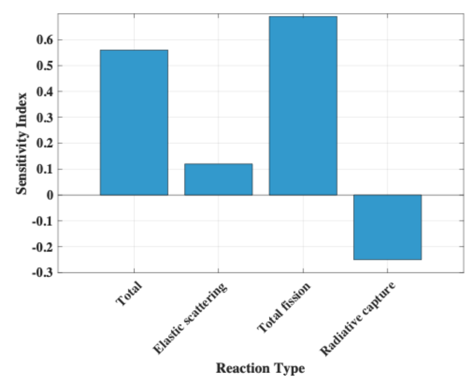


Fig. 4. Sensitivity indices of k_{eff} on different reaction types.

As ^{235}U is important for reactivity control and fuel depletion accuracy, a further examination on this specific nuclide is performed in the sensitivity analysis. ^{235}U plays a large role in the fission sensitivity and therefore the overall sensitivity of k_{eff} of the reactor. This is shown in Fig. 5 where there is a high sensitivity of k_{eff} on the ^{235}U fission reaction.

Since earlier analysis showed non-negligible differences in the neutron energy spectra, a sensitivity analysis on energy dependent reactions was calculated. Fig. 6 shows the k_{eff} is most sensitive to changes in the high energy groups of the reactions for ^{235}U. The total reaction rate energy dependent sensitivity analysis, corresponding to Fig. 4, also indicates the same high energy sensitivity.

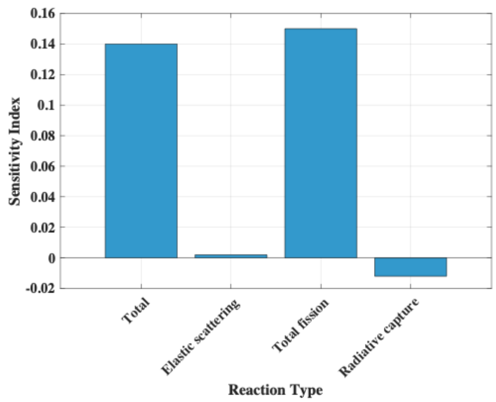


Fig. 5. ^{235}U sensitivity by reaction types.

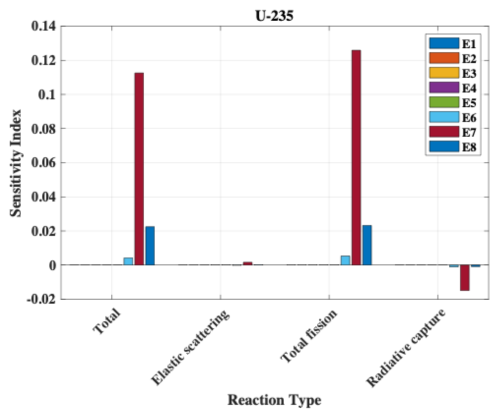


Fig. 6. Energy-dependent ^{235}U sensitivity by reaction types.

Additionally, sensitivity analysis on isotopes such as ^{241}Pu, ^{240}Pu and ^{37}Cl were performed but results are not shown in the summary for concise purposes. It appears that ^{241}Pu also has high fission and moderate capture sensitivity. The results for these isotopes indicate they may play an important role in long-term reactivity and fuel breeding. On another side, ^{37}Cl shows less sensitivity than fissionable isotopes, but its results are still important. Its radiative capture sensitivity is worth noting, which is important for assessing neutron economy and leakage.

DISCUSSION AND CONCLUSIONS

The results from this study support the conclusion that data discrepancies significantly impact k_{eff} reactor criticality calculations. The neutron energy spectra analysis showed that the energy spectra have noticeable difference between libraries, especially in the high energy region. Spectral changes in this region are significant as they influence the chances of competing neutron reactions, especially fission and capture which are highly correlated in these regions. These spectra difference can be linked to the difference in the integral reaction rates by the sensitivity analysis.

The sensitivity analysis results showed that k_{eff} is very sensitive to changes in fission and capture cross sections reactions, with the highest sensitivity seen in ^{235}U, ^{240}Pu, ^{241}Pu, and ^{37}Cl. Breaking down the two reactions into an energy dependent sensitivity analysis, the total reactions of the system and for the respective isotopes showed to be most sensitive to the high energy groups. This, in combination with the results from the neutron energy spectra analysis, shows that where the most uncertainty and difference in the hightail of the energy spectra is seen is where k_{eff} is most sensitive to changes in energy dependent reactions.

Between libraries it can be concluded that the fluctuation in high energy spectra has an impact on the energy dependent reaction types (capture and fission) that the reactor is most sensitive to. This relation explains that the difference in the integral reaction rates is a result from difference in high energy spectrum across the two libraries and therefore provides evidence on changes in $k_{eff.}$ Connecting the neutron spectra difference and sensitivity to high energy dependent reactions to the integral reaction rates, there is a full picture for how different physics parameters play into reactor criticality simulations. Since ^{235}U, ^{240}Pu, ^{241}Pu, and ^{37}Cl had the most capture and fission sensitivity, it can be concluded they carry the most impact on the k_{eff} estimate.

These findings underscore the importance of accurate energy-resolved nuclear data for key fissionable isotopes and salt constituents. Given that fuel TRU/U^{enr}-Chloride showed the most difference, future work should investigate refinement and further nuclear data analysis on ^{235}U, plutonium nuclides, and the chloride nuclides to ensure the accuracy of the chloride fuel simulation in MCFRs.

ACKNOWLEDGEMENTS

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REFERENCES

[1]. National Nuclear Data Center. Evaluated Nuclear Data File (ENDF) nndc.bnl.gov.

[2]. R MACFARLANE et al., "The NJOY Nuclear Data Processing System, Version 2016," LA-UR--17-20093, LANL, United States (2017).

[3]. M. BROVCHENKO et al., "Neutronic benchmark of the molten salt fast reactor in the frame of the EVOL and MARS collaborative projects," *EPJ Nuclear Science and Technology*, **5**, pp.2 (2019).

[4]. J LEPPÄNEN et al., "The Serpent Monte Carlo code: Status, development and applications in 2013," VTT Technical Research Centre of Finland, Finland, Tietotie 3, Espoo, FI-02044 VTT (2014).

[5]. M. A. NASR et al., "Neutronic and fuel cycle performance analysis of fluoride and chloride fuels in Molten Salt Fast Reactor (MSFR)," *Nuclear Engineering and Design*, **413**, p.112506 (2023).