Nuclear Data Uncertainty Quantification for Reactor Physics Parameters in Fluorine-19-based Molten Salt Reactors

WONDER-2023

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Abstract. The use of the F-19 isotope in the nuclear fuel cycle is already well established for fuel enrichment, but future plans for Gen-IV reactors, such as Molten Salt Reactors, could utilize a fluorine-based salt as a basis for the fuel. It is therefore imperative that an understanding of the characteristics of F-19 is instituted, and one component of key interest is the quantification of reactor parameter uncertainties that arise from the uncertainties in the nuclear data. The results from such analyses can shed light on where experimentalists need to further improve nuclear data for F-19, as well as yielding critical information for developing and optimizing reactor designs thanks to greater knowledge of the uncertainties that result from nuclear data.

In this work, we analysed a molten salt reactor based on the designs made by Transatomic Power. We conducted uncertainty quantification on three reactor operating modes: thermal, semi-epithermal, and epithermal. In the epithermal mode, the neutron spectrum is faster than in the thermal mode because fewer moderator rods are used. We generated nuclear data that was sampled from the covariance matrices in the JEFF-3.3 nuclear data library using SANDY[1] and NJOY. By utilising the Total Monte Carlo approach, we propagated the uncertainties from the samples to uncertainties in the neutron multiplication by simulating the reactor in OpenMC, a Monte Carlo-based neutron transport code. By perturbing individual reaction channels while keeping others constant, it was possible to quantify the amount of contribution each single reaction channel has to the overall uncertainty.

For the thermal reactor, the F-19 data sampling resulted in an uncertainty in reactivity of 62 pcm. The main contributors to the reactivity uncertainty for the thermal reactor are elastic scattering, neutron capture and alpha production. The epithermal reactor, with a reactivity uncertainty of 213 pcm, is mostly affected by elastic scattering, inelastic scattering, and alpha production. The alpha production channel had an unexpectedly large contribution, and it should be investigated further. The results should be considered preliminary. Quantitatively, we observe that scattering plays a bigger role for the uncertainty in the epithermal system, a phenomenon which could be explained by the fact that with less moderation in the form of moderator rods, the role of F-19 in slowing down neutrons is greater, and hence its contribution to the uncertainty is greater.

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1 Introduction

With an increased interest for more nuclear power-based means of energy production, the hope is growing for *Generation IV nuclear reactors* in solving many of the contemporary problems with nuclear technology. Recently, there has been a growing interest in the Molten Salt Reactor (MSR) concept. This involves the use of a solution of molten salt, typically based on fluorine, to contain the nuclear fuel. This concept has gained attention from both research institutions and industry actors. Although fluorine is used in the nuclear fuel cycle in the form of UF_6 during the enrichment process, the experience with it as a part of the nuclear fuel in reactors is scarce. In a similar manner, only a small number of MSRs have ever been constructed, which limits the practical experience with the technology. Due to these reasons, it is important that investigations are conducted within the reactor physics behaviour of fluorine-based MSRs, as it can shed light on further areas of research that need to be approached before such technology can become commercially viable.

In this work, nuclear data uncertainties for the isotope F-19 are propagated to uncertainties in reactivity for a small fluorine-based MSR. In addition, the individual reaction channel contributions to the overall reactivity uncertainty are investigated in order to allow for conclusions regarding which reaction channels affect the uncertainties the most. By conducting these analyses, it is possible to identify the reaction channels that require improvements in their nuclear data. This can include both enhancing experimental measurements, improved theoretical modelling and improved evaluation techniques.

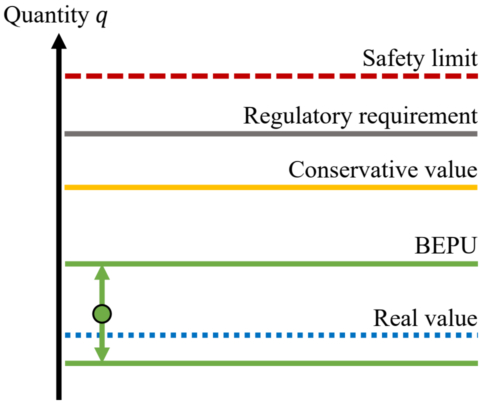


Figure 1: Illustration of the difference between estimates of a quantity q using *Best Estimate Plus Uncertainty* (BEPU) and *conservative calculations*.

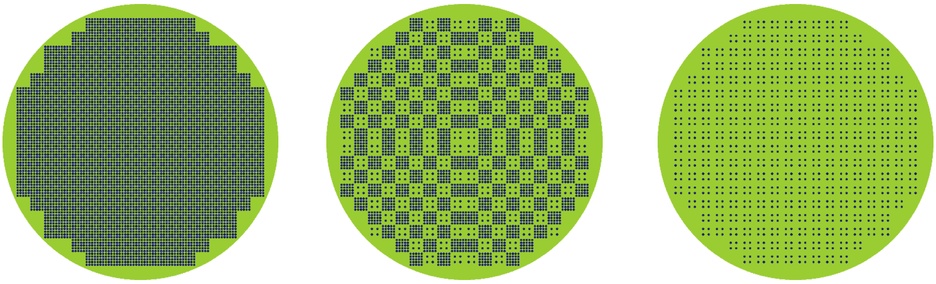
Traditionally, safety limits and regulatory requirements for a general reactor quantity, q, were determined through *conservative calculations*. These calculations assumed that the conditions in the reactor were at their worst, resulting in an estimated value for q that was closest to the safety limit. This approach aimed to provide a conservative estimate for safety purposes.. In Fig. 1, such a quantity is visualized where the safety limit is assumed to be a maximum value of q before the safety of the reactor is not guaranteed. The quantity could, for example, be the fuel temperature, where, in a solid-fuel reactor, it must not exceed the melting point of the fuel, which represents the safety limit. Regulatory authorities will demand some safety margin, and thus require that the quantity not exceed the regulatory requirements. Instead of estimating the quantity q with a conservative calculation, creating a *Best Estimate Plus Uncertainty* (BEPU) calculation in which the quantity q is estimated by a central value combined with an uncertainty can, in many cases, yield larger margins to the safety limits and regulatory requirements of the quantity while representing a more realistic estimate compared to a conservative one.

2 Method

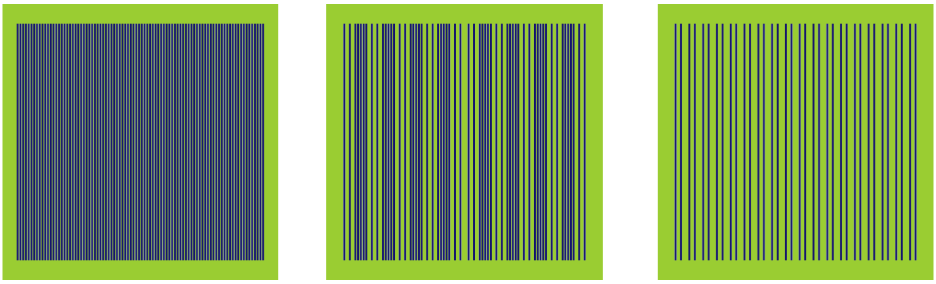
The work presented in this report is conducted with the Monte Carlo-based neutron transport code *OpenMC*[2] for simulation of the neutronics in a single-physics analysis. The *Total Monte Carlo*[3] (TMC) methodology is applied in order to quantify the uncertainty in reactivity that originates from the uncertainty in nuclear data for F-19.

2.1 Reactor Model

The reactor model used to perform the uncertainty quantification is based on a design from Transatomic Power[4], and it represents a simplified model of a small MSR with a fluorine-based fuel salt. In order to compensate for the loss in reactivity due to fuel burn-up, the level of moderation can be increased compared to the initial configuration in order to create a more thermal neutron spectrum, which is a more favourable environment for the fissile material to undergo fission in the core. Thus, three levels of moderation were analyzed in this work - a thermal, semi-epithermal, and epithermal, with the first being the most moderated (used in EOC) and the last being the least moderated (used in BOC). However, in this report, no burn-up is simulated, and the three systems will be simulated with the same fuel compositions. The reactors are visualized in Fig. 2, with the inner lattices being visualized in Fig. 3.



(a) Thermal, top-down view (b) Semiepithermal, top-down view (c) Epithermal, top-down view

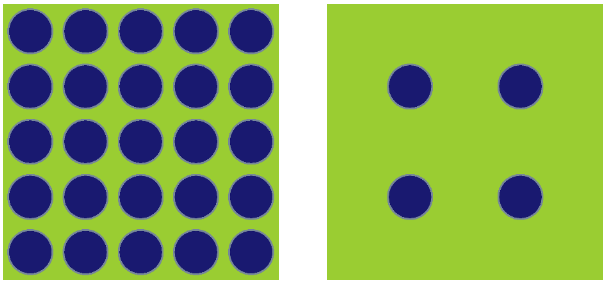


(d) Thermal, side view (e) Semiepithermal, side view (f) Epithermal, side view

Figure 2: Slice-through visualizations for top-down and side views for the thermal, semiepithermal, and epithermal systems. The fuel is green, moderator is blue, and cladding is gray - the last of which is too small to be visible in these images.

https://doi.org/10.1051/epjconf/202429405004

The fuel is based on the elements Li, F, and U with a 5 % enrichment of U-235[5]. The moderator is based on Zr and H, and the cladding is based on C and Si. The simulated reactor power is 400 MW, fuel temperature is 900 K, moderator temperature is 600 K, cladding temperature is 900 K, fuel density is 5.01 g/cm^3, moderator density is 3.21 g/cm^3, cladding density is 5.66 g/cm^3, moderator rod radius is 1.15 cm, cladding blanket radius is 1.25 cm, reactor radius is 150 cm, core height is 350 cm, plenum height is 25 cm, inner lattice pitch is 3 cm, outer lattice pitch is 15 cm, inactive batch count is 50, active batch count is 300, and the number of particle histories is 30,000.



(a) Thermal, top-down view (b) Epithermal, top-down view

Figure 3: Slice-through visualizations for top-down views for the thermal and epithermal systems. The semiepithermal system is a mix of the thermal and epithermal inner lattices. The fuel is green, moderator is blue, and cladding is gray.

2.2 Uncertainty Quantification

In order to perform uncertainty quantification on the nuclear data of F-19 using the TMC method, it is a necessity to sample data based on the uncertainties and covariances stored in the ENDF files. In this work, the nuclear data library JEFF-3.3 was used, and the nuclear data sampling code SANDY was used in conjunction with NJOY and its modules MODER, RECONR, BROADR, UNRESR, THERMR, PURR, ERRORR, COVR, VIEWR, ACER in order to create 500 perturbations of the JEFF-3.3 nuclear data for F-19. Through the use of TMC method, it is possible to propagate the nuclear data uncertainty to a quantity of interest. This is done by first perturbing the nuclear data and subsequently performing multiple reactor simulations with different realizations of the perturbed nuclear data. This process results in an uncertainty denoted as σ_{obs} for the quantity of interest, which in the case of this work is the neutron multiplication factor k_{eff} and its related reactivity. There will also be a statistical error σ_{stat} since a Monte Carlo-based transport code, OpenMC, was used to simulate the reactor. The value of σ_{stat} is estimated as the mean value of the statistical error that OpenMC reports for each reactor simulation. The uncertainty due to nuclear data, σ_{ND}, is related to the previous two uncertainties as

$$\sigma_{\text{obs}}^2 = \sigma_{\text{ND}}^2 + \sigma_{\text{stat}}^2 \tag{1}$$

It is also possible to sample each reaction channel separately in order to investigate how much each reaction channel contributes to the overall uncertainty.

3 Results

Using the TMC approach as outlined in Sec. 2.2, the uncertainty in reactivity can be extracted using Eq. 1 and the relation

$$\rho = \frac{k_{\mathrm{eff}} - 1}{k_{\mathrm{eff}}} = 1 - \frac{1}{k_{\mathrm{eff}}} \tag{2}$$

Using the concept of error propagation, the uncertainty in reactivity is

$$\sigma_\rho = \sqrt{\left(\frac{\partial \rho}{\partial k_{\mathrm{eff}}}\right)^2 \sigma^2_{k_{\mathrm{eff}}}} = \frac{1}{k^2_{\mathrm{eff}}}\sigma_{k_{\mathrm{eff}}} \tag{3}$$

Hence, the uncertainty in reactivity can be calculated when sampling all the reaction channels for F-19, as presented in Sec. 3.1, as well as each individual reaction channel for F-19, as presented in Sec. 3.2.

Table 1: Values of k_{eff}, its associated uncertainty in relative and absolute terms, along with the reactivity and ρ and its absolute uncertainty for the thermal, semi-epithermal, and epithermal systems. The uncertainty reported here comes from the propagation of F-19 nuclear data uncertainties using the TMC analysis. The JEFF-3.3 nuclear data library was sampled with 500 perturbations using the associated nuclear data uncertainties and covariances.

Reactor model	k_{eff}	$\sigma_{k_{\mathrm{eff}}}$ [pcm]	$\sigma_{k_{\mathrm{eff}}}/k_{\mathrm{eff}}$ [%]	ρ	σ_ρ [pcm]
Thermal	1.3670	117.0	0.09	0.2684	62.6
Semi-epithermal	1.2767	167.2	0.13	0.2167	102.6
Epithermal	0.9728	205.8	0.21	-0.0279	217.5

3.1 Reactivity Uncertainties

In Tab. 1, it can be observed that the reactivity uncertainty increases as the level of moderation decreases. Thus, the thermal system with the most amount of moderation has the lowest reactivity uncertainty, while the epithermal system, which has the least amount of moderation, has the highest reactivity uncertainty. Possible explanations for this effect are twofold:

- The volumetric amount of fuel is larger in the epithermal inner lattice compared to the thermal inner lattice, as seen in Fig. 3. As the total amount of fuel is higher in the less moderated systems, the system would be more susceptible to the uncertainties in the fuel, which means that the uncertainties from the nuclear data of F-19 have a larger footprint.
- In general, it was observed that the relative uncertainties of the cross sections for F-19 increased with energy for many of the reaction channels. As the neutron spectrum is faster for the less moderated systems, this means that a larger part of the spectrum is exposed to the regions that have a higher relative cross section uncertainty.

Table 2: Reactivity uncertainty contributions in terms of absolute uncertainties and relative variances for the thermal and epithermal systems, when different reaction channels are perturbed individually. The filled proportion of the circles corresponds to the proportion that each reaction channel contributes to the total uncertainty. At the bottom, a summation of the relative variances for each reaction channel is presented. The uncertainty reported here comes from the propagation of F-19 nuclear data uncertainties using the TMC analysis. The JEFF-3.3 nuclear data library was sampled with 500 perturbations using the associated nuclear data uncertainties and covariances.

		Thermal			**Epithermal**		
MT	**Label**	$\sigma_{\rho,\mathrm{MT}}$ [pcm]	$\frac{\sigma^2_{\rho,\mathrm{MT}}}{\sigma^2_{\rho,ALL}}$ [%]		$\sigma_{\rho,\mathrm{MT}}$ [pcm]	$\frac{\sigma^2_{\rho,\mathrm{MT}}}{\sigma^2_{\rho,ALL}}$ [%]	
ALL	-	61.5	100.0	●	213.4	100.0	●
2	(n,elastic)	28.6	21.6	◔	155.8	53.3	◑
4	(n,inelastic)	9.3	2.3	○	113.9	28.5	◔
16	(n,2n)	5.5	0.8	○	16.7	0.6	○
22	(n,na)	10.1	2.7	○	18.6	0.8	○
28	(n,np)	9.9	2.6	○	15.1	0.5	○
102	(n,γ)	34.1	30.7	◔	53.4	6.2	○
103	(n,p)	11.0	3.2	○	19.9	0.9	○
104	(n,d)	5.7	0.9	○	15.5	0.5	○
105	(n,t)	10.1	2.7	○	16.7	0.6	○
107	(n,α)	42.7	48.2	◑	138.5	42.1	◑
		$\sum_{\mathrm{MT}\neq ALL} \frac{\sigma^2_{\rho,\mathrm{MT}}}{\sigma^2_{\rho,ALL}}$	$= 115.7$		$\sum_{\mathrm{MT}\neq ALL} \frac{\sigma^2_{\rho,\mathrm{MT}}}{\sigma^2_{\rho,ALL}}$	$= 134.0$	

3.2 Individual Reaction Channel Uncertainty Contributions

From the results in Tab. 2, it is evident that the different levels of moderation affect which reaction channels that play the most important roles for the reactivity uncertainty. Note that the semi-epithermal system was not investigated in terms of individual reaction channels, as it was assumed that it would be possible to interpolate the results between the thermal and epithermal systems, as was the case in Sec. 3.1. Tab. 2 shows that the thermal system is mostly affected by uncertainties in elastic scattering (MT2), neutron capture (MT102) and alpha production (MT107). The epithermal system is mostly affected by uncertainties in elastic scattering (MT2), inelastic scattering (MT4) and alpha production (MT107).

The fact that the proportional contributions from scattering increase when decreasing the moderation can be explained by the fact that when there is less moderation in the core, the role of F-19 in providing the thermalization of neutrons increases. Thus, the scattering components, both elastic (MT2) and inelastic (MT4), have a greater effect in the epithermal system compared to the thermal system.

The variances for the individual reaction channels, when normalized to the total variance when all reaction channels are sampled, sum up to approximately 100 %.

4 Discussion and Outlook

The purpose of this work was to investigate the reactivity uncertainty that originates from nuclear data uncertainties of F-19 in single-physics simulations of fluorine-based MSRs. Using the Monte Carlo-based neutron transport code OpenMC, the reactivity uncertainties of three different levels of moderation have been deduced and the contribution from individual reaction channels have been determined for the most and least moderated systems.

The results that have emerged from this analysis shed light on which parts of the nuclear data of F-19 that need to be improved. The large contribution from alpha production (MT107) was rather unexpected, and needs to be investigated further. The results should hence be seen as preliminary. The MT107 reaction only occurs at high energies, while other reactions, such as neutron capture (MT102), have the largest cross section in the resonance range, which could be a contributing factor as to why MT107 has a larger uncertainty contribution compared to MT102 even though MT102 has a higher reaction rate. Additionally, the results also show which other reaction channels that have the largest impact on the reactivity uncertainty at different levels of moderation.

Further investigations within uncertainty propagation of F-19 nuclear data uncertainties are imperative for the further development of fluorine-based reactor concepts. To build on the analyses performed in this work, it would be valuable to investigate other nuclear data libraries than JEFF-3.3. The ENDF/B-VIII.0 nuclear data library was in fact investigated, and the uncertainties were found to be much smaller than for JEFF-3. Here further analysis is needed. In conclusion, it was found that the nuclear data uncertainties of F-19 do contribute to some significant uncertainties in the reactivity of fluorine-based MSRs. Improvements in the nuclear data, its uncertainties and covariances should be made in order to allow for higher levels of confidence around the designs and optimizations of the previously mentioned reactor concepts.

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Acknowledgements

This work has been partially supported by the ENEN2plus project (HORIZON-EURATOM-2021-NRT-01-13 101061677) founded by the European Union.

Co-funded by
the European Union

The work has partly been performed under the Swedish competence centre ANItA, https://www3.uu.se/forskning/anita/.