SENSITIVITY TO CHLORINE NUCLEAR DATA IN MOLTEN CHLORIDE FAST REACTORS

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Abstract

Moltex Energy is pioneering the Stable Salt Reactor – Wasteburner, a fast spectrum molten salt reactor fuelled with recycled spent fuel waste, selected to be built at Point Lepreau by NB Power. A fast neutron spectrum enables the burning of all actinides in the generation of electricity, addressing the problem of spent fuel waste. Use of chloride salts results in a particularly fast spectrum, improving neutronic performance and waste reduction when compared with the fluoride salts often considered. However, given the lack of operating data for chloride fast reactors, it is important to understand the uncertainty in the relevant data, and the effect on reactor performance. In recent years chlorine data has been revised in the major international evaluated nuclear data libraries. This paper demonstrates the level of sensitivity to these changes for the SSR-W, and to the uncertainty that still exists, in particular in the Cl-35 (n, p) reaction that dominates behaviour. Detailed full core models are used in a perturbative analysis to derive the effect on the main parameters, accounting for burnup. The resulting uncertainty is shown to be significant, which must be accounted for in design, but is also only one of many sources of uncertainty.

1. Introduction

There is currently renewed interest in innovative reactor designs that can reduce the cost of nuclear power, to make a significant contribution to net zero emissions by the displacement of fossil fuel generation. Fast molten salt reactors are particularly attractive due to their high degree of inherent safety and ability to be fuelled with trans-uranics (TRU) from spent fuel – reducing legacy waste liabilities.

The use of chloride salts gives a particularly hard neutron energy spectrum, relative even to fluoride fast reactors, due to the low slowing-down power of the non-TRU constituents. This hard spectrum maximises the TRU conversion and burning, and minimises the final TRU waste. A number of organisations worldwide are currently working on chloride fast reactors, including TerraPower and Elysium in the US and the CEA/Orano in France.

The design of any reactor relies heavily on nuclear data that describes the interaction of neutrons with the nuclides in the system. This nuclear data is the product of a large amount of experimental and theoretical data, that has been evaluated and collated by specialists into an evaluated nuclear data library. Uncertainty in this nuclear data must be accounted for in design and operation, particularly in derivation of margins to any safety limits. Reduction of uncertainties therefore in principle expands the available design and operation space and reduces costs.

No chloride molten salt reactors have been operated, and there is large uncertainty in reactivity for chloride fast reactors, due mainly to the uncertainty in the Cl-35 (n,p) cross section at high energies. This presents a difficulty for chloride fast reactor design because the required fissile loading has large uncertainty, and this uncertainty would also propagate to other parameters important to safety, such as reactivity coefficients. Production of Cl-36 via neutron capture in Cl-35 is also important for characterisation of salt waste.

This paper summarises the current state of knowledge of chlorine nuclear data and shows that Cl cross section uncertainty is a significant source of uncertainty for important integral parameters.

2. The Moltex Energy SSR-W

Moltex Energy and partners are developing advanced technologies that can produce reliable, affordable and safe carbon-free energy while reducing the stockpile of the legacy nuclear waste. The SSR-W is a small modular reactor that generates electricity by burning nuclear waste, such as used CANDU or PWR fuel [1].

The SSR-W is fueled with a molten chloride fuel salt composed of plutonium chloride, uranium chloride, other actinide chlorides, lanthanide chlorides, and potassium chloride. It is distinguished from other molten salt reactors by the use of a molten fuel salt in static tubes, cooled by a separate circulating non-radioactive coolant salt. Figure 1 shows a schematic of the reactor and Figure 2 shows the neutron energy spectrum.

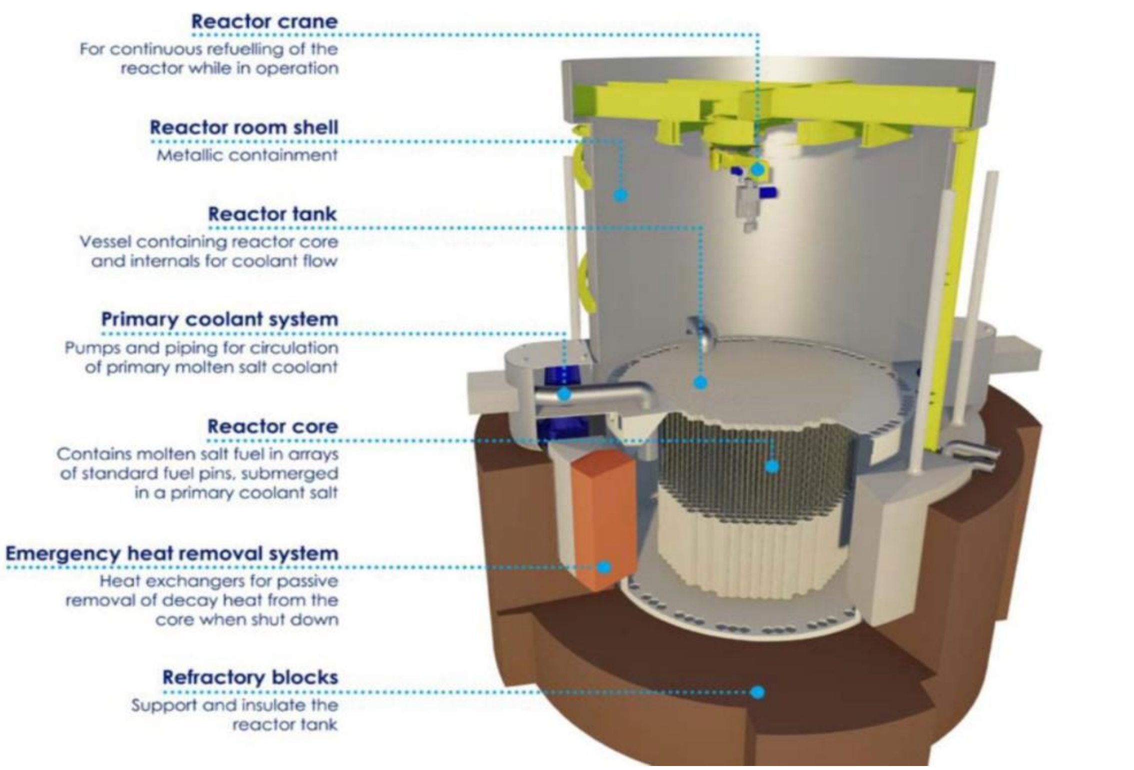

Figure 1 – SSR-W reactor tank and primary loop.

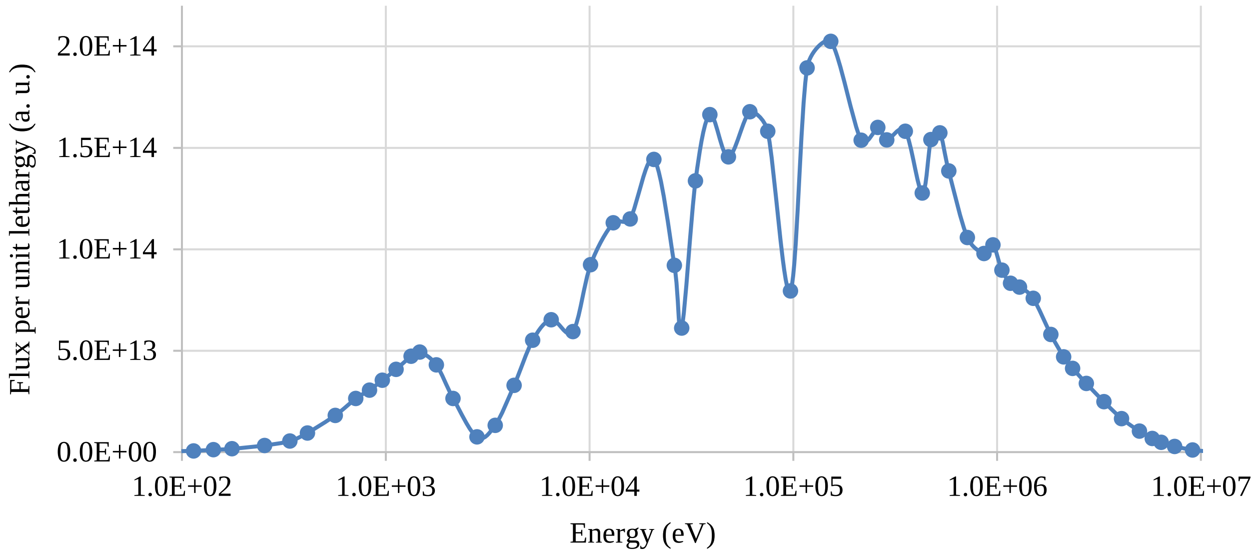

Figure 2 – The neutron energy spectrum in the fuel salt of the SSR-W.

3. Current status of chlorine nuclear data

3.1 Chlorine cross sections

While low in absolute terms, the neutron absorption cross sections of Cl-35 are significant at fast energies. There are also multiple reaction pathways available, with gamma, proton or alpha emission all possible with significant probability, and with complex resonant structure up to high energies – see Figure 3.

The Cl-35 (n,p) cross section has changed significantly at fast energies in recent evaluated nuclear data libraries. The JEFF-3.3 [2], ENDF/B-VII.1 and ENDF/B-VIII.0 [3] evaluated nuclear data libraries contain an evaluation of the Cl-35 (n,p) cross section which is orders of magnitude lower than that in previous libraries in the range ~ 0.1 – 1.3 MeV, due to the use of a resolved resonance approach rather than a statistical approach [4]. This difference is visible in Figure 3 in the difference between the ENDF/B-VII.0 and ENDF/B-VIII.0 libraries. The (n,γ) cross section is also reduced at MeV energies relative to ENDF/B-VII.0, while the (n,α) cross section is unchanged. It will be shown below that these differences result in a large difference in reactivity as calculated with different nuclear data libraries.

The Cl-35 (n,γ) cross section is much lower than the (n,p) cross section at energies above ~1 MeV, however between 0.1 and 1 MeV it lies between recent evaluations of the (n,p) cross section, as seen in Figure 3. In addition to the reactivity effect, this reaction is significant due to the resulting production of Cl-36, a long-lived nuclide which is important for waste disposal considerations.

Other chlorine cross sections are also relevant for prediction of chloride fast reactor performance: the Cl-36 capture cross sections for prediction of Cl-36 destruction, and the scattering cross sections for prediction of leakage.

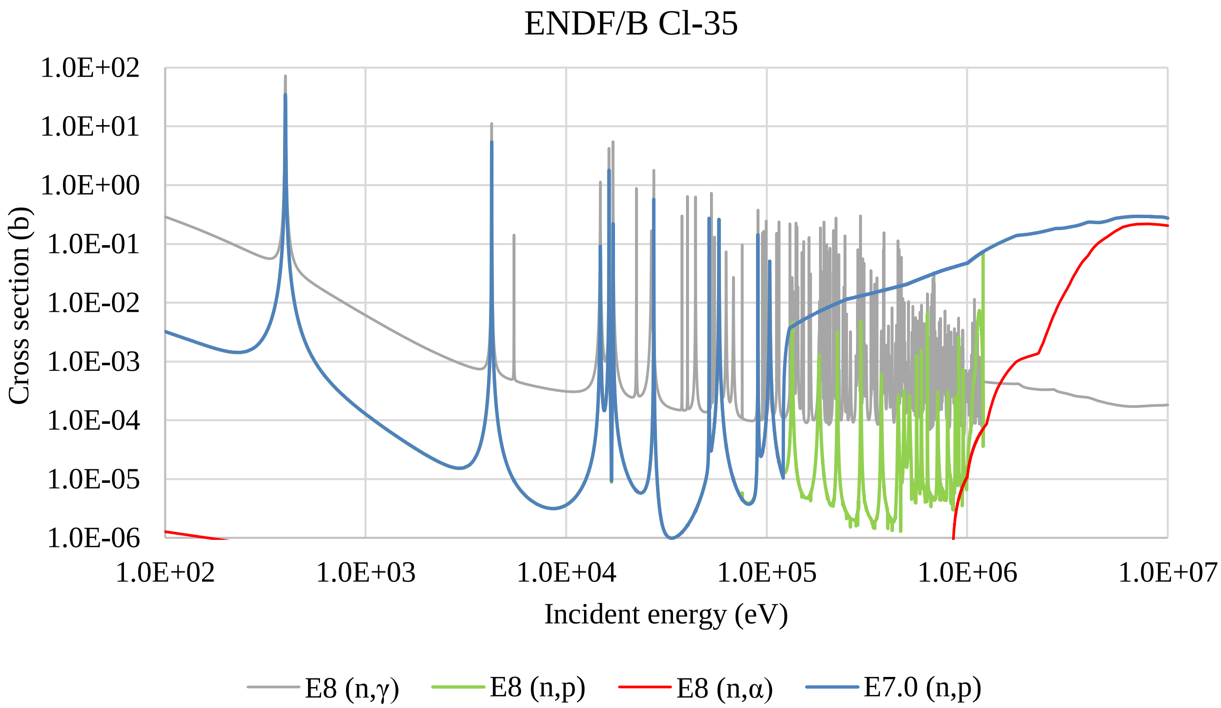

Figure 3 – Cl-35 cross sections in the energy range of interest, from the ENDF/B-VII.0 and ENDF/B-VIII.0 evaluated nuclear data libraries.

3.2 Recent experimental data

There is little experimental data available for chlorine cross sections at fast energies (above about 100 keV). The presence of narrow resonances at the energies of interest means that high resolution is required in measurement of neutron energy, which is experimentally difficult. Direct measurement of the (n,p) cross section is also complicated by the need to distinguish multiple charged particle output channels, due to the significant (n,α) cross section above ~1 MeV.

However two US groups have performed new measurements in recent years [5], [6]. Both sets of measurements of the Cl-35 (n,p) cross section provide new data at high energies. Reference [5] reports measurements around 2.5 MeV, finding a cross section significantly lower than that in all evaluated libraries and evidence of resonant structure even at these high energies, where all evaluations to date have used a statistical treatment.

Reference [6] reports measurements over a wider energy range which are also significantly lower than those in evaluated libraries between 1 and 5 MeV, although the data is not fully consistent with that from [5].

On the other hand, [6] suggests that the most recent evaluations (e.g. ENDF/B-VIII, JEFF-3.3) have *underestimated* the Cl-35 (n,p) cross section between 600 keV and 1 MeV. In this energy range, these measurements appear closer to the previous evaluations, in e.g. JEFF-3.2 or ENDF/B-VII.0.

To summarise, recent differential measurements suggest a higher cross section below 1 MeV, but a lower cross section above 1 MeV, with resonant structure up to at least 3 MeV. However, these two sets of measurements, [5] and [6], do not fully agree. There are also new cross section measurements for the Cl-35 (n,α) cross section [7].

3.3 Cross section uncertainties

Covariance data are important for the quantification of uncertainties in output integral parameters due to (input) nuclear data. These covariance data are presented as an energy-dependent matrix, with the uncertainty (variance) in the cross section on the diagonal and correlations between energy groups off-diagonal.

These covariance data can be combined with sensitivity coefficients for an output of interest, *R*, to estimate the uncertainty in this output due to the nuclear data, using generalised perturbation theory [8]:

$$\Delta R^2 = S_R^+ D S_R, \tag{1}$$

where *S* is the matrix of sensitivity coefficients of R to a cross section and *D* the covariance matrix for this cross section.

Uncertainties contained in different evaluated data libraries for Cl-35 (n,p) vary significantly. For example, in the TENDL-2021 library [9] the uncertainty is around 10 % at energies of interest, however in other libraries the uncertainty is much higher.

Above 1.2 MeV there is no uncertainty available for Cl-35 (n,p) in any evaluated library, except the theoretical TENDL libraries and BROND-3.1 [10]. This makes it difficult to justify a reasonable total uncertainty.

4. Sensitivity to chlorine cross sections

Reactor physics analysis has been performed for the SSR-W using ENDF/B-VII.0 and the Argonne Reactor Computation (ARC) suite of codes including MC^2-3 [11], DIF3D-VARIANT [12] and PERSENT [13]. MC^2-3 is a lattice physics code which is well suited to generate multi-group cross-sections for fast reactor physics analysis. DIF3D-VARIANT solves the diffusion or transport problem and provides full core forward and adjoint flux solutions for perturbation and sensitivity analysis. PERSENT is the physics code which implements generalized perturbation theory and allows the user to perform perturbation theory and sensitivity calculations to a variety of reactor physics parameters ranging from reactivity coefficients, reaction ratios to kinetics parameters, etc. The US DOE Nuclear Energy Advanced Modeling and Simulation (NEAMS) program Workbench facilitates the use of the ARC codes [14].

Due to the lack of complete covariance data for Cl-35 in the data libraries available, the uncertainties of the calculated output physics parameters due to nuclear data uncertainties were quantified via two types of uncertainty analysis:

- Direct comparison of the reactor physics parameters by deploying the ENDF/B-VII.0, ENDF/B-VII.1 and ENDF/B-VIII.0 libraries. The impacts of the large uncertainties of Cl-35 (n, p) cross sections around 2.5 MeV were also analyzed by manually adjusting the cross sections around that energy.
- Sensitivity analysis using the PERSENT code to calculate the relative importance of the uncertainties among the different cross section data and isotopes. Covariance data based on the ENDF/B-VII.0 library was used [15]. The lack of covariance data for the chlorine

isotopes means this excludes what is expected to be a large contribution to the total uncertainty.

4.1 Sensitivity to choice of nuclear data library

An MCNP6 [16] model with homogenized assemblies was used to study the impacts of adopting different nuclear data libraries in modeling the SSR-W core. Calculations used the ENDF/B-VII.0, ENDF/B-VII.1 and ENDF/B-VIII.0 evaluated nuclear data libraries. The simulations were also performed varying *only* the Cl-35 nuclear data, with ENDF/B-VII.0 used for all other isotopes. This isolates the effect of Cl-35 data. Table 1 shows that there is a large difference in the k-eff calculated using different data libraries, and furthermore that this difference is almost entirely due to the Cl-35 cross sections alone.

Table 1 – Calculated eigenvalues of the SSR-W with different data libraries. Statistical uncertainty as estimated by MNCP is < 10 pcm.

	ENDF/B-VII.0		ENDF/B-VII.1		ENDF/B-VIII.0	
	k-eff	Diff (pcm)	k-eff	Diff (pcm)	k-eff	Diff (pcm)
All isotope	1.00862	-	1.04996	4134	1.04763	3901
Only ^{35}Cl	1.00862	-	1.04801	3939	1.04843	3981

The impact on the spectrum has also been checked, however the difference between cases with different libraries is seen to be small.

Deterministic reactor physics are better suited to investigate the impact of uncertainty in the Cl-35 (n,p) cross sections around 2.5 MeV. The ANL ARC reactor physics codes allow manual adjustment of the neutron cross sections for each isotope at any energy group for sensitivity studies. Therefore, for this purpose, the (n,p) macroscopic cross section in the SSR-W model in the energy range from 2.23 to 3.68 MeV was reduced to 50 %, 33 % and 20 % of its nominal value. This is an artificial change to the cross section, but demonstrates sensitivity to the cross section at energies where recent measurements have been made. DIF3D and PERSENT were then used to calculate k-eff, kinetics parameters and reactivity coefficients with these reduced (n,p) cross sections.

Table 2 shows a large increase in the eigenvalue due to the reduction of the neutron loss to the (n,p) reaction in this energy range. Because of the increased neutron population in this energy range, the prompt neutron lifetime, Λ, and delayed neutron fraction, β, become slightly smaller. In addition, Table 2 also shows that the reactivity coefficients have low sensitivity to the large differences of the (n,p) cross section in this energy range. The reactivity coefficients are only reduced by a few percent, with the maximum change in the coolant density coefficient, which is reduced by around 5 – 6 % for a 5-fold reduction in the (n,p) cross section in this energy range.

Table 2 – Calculated parameters with adjustment by a factor *f* of the Cl-35 (n, p) cross section in the energy range [2.23, 3.68] MeV.

f	1.0	0.5	0.33	0.2
k-eff	1.00416	1.01336	1.01662	1.01917
β	---	0.992	0.990	0.988
Λ	---	0.990	0.986	0.984
α_r (core radial expansion)	---	0.987	0.982	0.979
α_{ρ_f} (fuel density)	---	0.993	0.990	0.988
α_{ρ_c} (coolant density)	---	0.965	0.953	0.942

4.2 Sensitivity coefficients

The sensitivity coefficient is defined as the subsequent change in an integral parameter R due to a constant variation (generally 1 %) of the cross section [17]:

$$S_R = \frac{\partial R}{\partial \sigma} \times \frac{\sigma}{R} \tag{2}$$

The PERSENT code [13] was used to calculate the sensitivity coefficient matrix for the SSR-W300 core. The integral parameter R considered in these sensitivity analyses are the kinetics parameters and reactivity coefficients applied in the reactor safety transient simulations: eigenvalue, Λ, β, Doppler coefficient, $\alpha_{\rho_{f,doppler}}$, fuel expansion coefficient, α_{ρ_f}, coolant expansion coefficient, α_{ρ_c}, and core radial expansion coefficient, α_r.

Figure 5 shows the calculated sensitivity coefficients, using ENDF/B-VII.0 data, for the four isotopes that the integral parameter *k*-eff is most sensitive to. In general, the integral parameters are all seen to be most sensitive to the uncertainties in the fissile and fertile materials. For instance the number of neutrons released per fission, ν, the prompt fission spectrum, χ, and the fission cross section of Pu-239 are always observed to have the largest sensitivity coefficients.

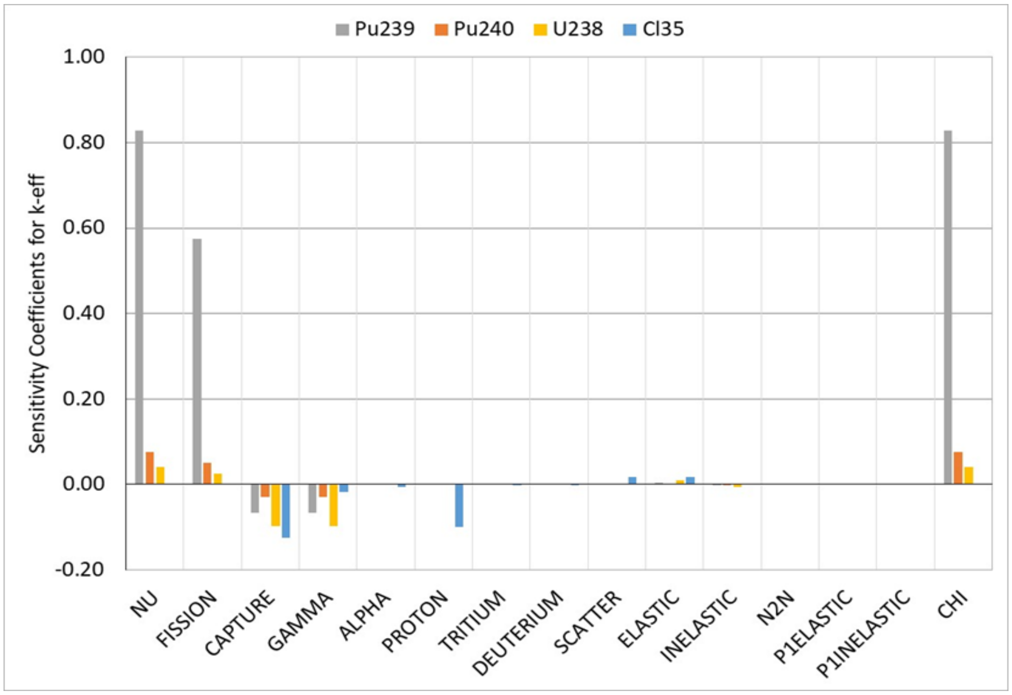

Figure 4 – The calculated sensitivity coefficients for k-eff to four isotopes, using ENDF/B-VII.0 data.

The coefficients for sensitivity to the Cl-35 cross sections are relatively small except for the coolant density reactivity coefficient. However, if large uncertainties exist in the chlorine cross sections, their contributions to the overall uncertainties will not be small. In particular, for Cl-35, Figure 3 suggests the (n,p) reaction will have the dominant impact, when using ENDF/B-VII.0 data. Figure 5 shows the calculated sensitivity coefficient as a function of energy for the Cl-35 (n,p) reaction, compared with another important reaction for the most absorbing actinide, Pu-239 (n,γ) – the importance of Cl-35 (n,p) at high neutron energy is clear.

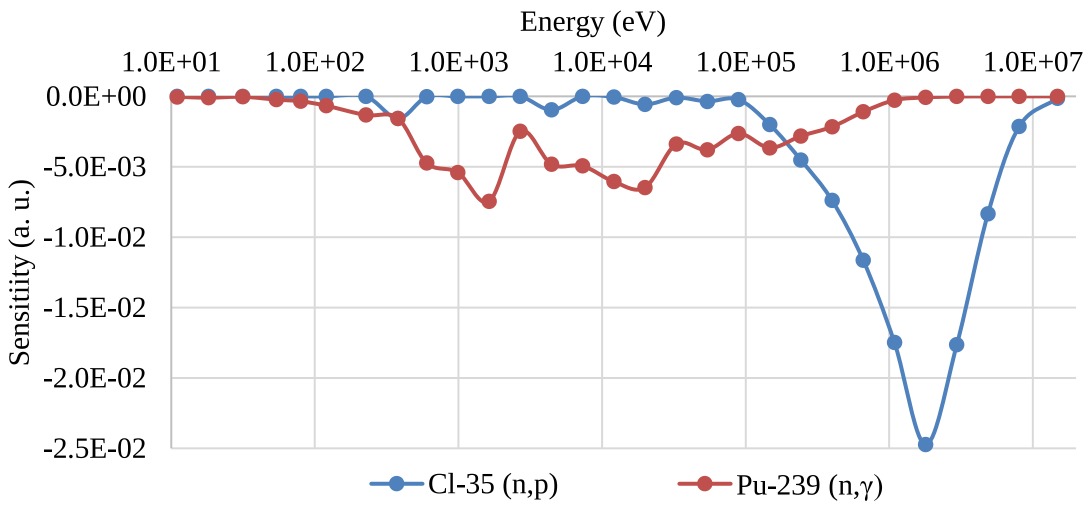

Figure 5 – The energy-dependent sensitivity profile for k-eff to two important reactions, using ENDF/B-VII.0 data.

5. Uncertainty on integral parameters due to nuclear data

Using the available covariance data, the total uncertainties from the uncertainties of those isotopes included in the covariance matrix file can be estimated. Their values are shown in Table 3. The overall uncertainty in k-eff is estimated to be only 630 pcm. However, the impact of Cl-35 cross section uncertainties is not included.

It has already been seen that updating chlorine cross sections can result in several thousand pcm differences in k-eff. Therefore the uncertainties in the chlorine isotopes are clearly important in order to provide accurate estimations of the uncertainties in all the integral values required by the reactor safety analyses.

An independent estimate of the total uncertainty for the SSR-W, with an attempt to account for Cl nuclear data uncertainty, gives a total of around 1000 pcm, and this using Cl cross section uncertainties from TENDL-2021, which may be too low (given the difference between recent measurements and evaluated data is much larger than 10 %).

Table 3 – The estimated standard deviation in outputs due to nuclear data and salt density, with 1 % uncertainty assumed for salt densities.

Integral R	Estimated relative SD from cross sections	Estimated relative SD from fuel and coolant salt densities
k-eff	0.63%	0.097%
Λ	1.55%	0.455%
β	0.54%	0.0026%
$\alpha_{\rho_{f,doppler}}$	2.68%	0.393%
α_{ρ_f}	1.25%	0.279%
α_{ρ_c}	3.93%	0.269%
α_r	0.87%	0.234%

6. Discussion

Reference [6] recommends that a full revaluation of the Cl-35 (n,p) cross section is performed, supported by new measurements over a wider energy range and to resolve discrepancies seen with measurements reported in [5]. Previous work on the REBUS-3700 fast chloride reactor also concluded that more accurate nuclear data for Cl was needed [18].

Covariance data is also needed for the highest energy range, which is still very relevant for fast chloride systems, due to the hard neutron spectrum, and as visible in Figure 5. With realistic covariances over the relevant energy range, the uncertainty due to nuclear data can be accurately quantified and this uncertainty accounted for in design of the SSR-W.

Moltex have prepared an entry for Cl-35 (n,p) for the NEA's High Priority Request List for nuclear data [19]. This list forms an agreed set of reactions that are priorities for improvement, via new measurement and/or evaluation. The proposed entry suggests a target accuracy of 8 %, on the basis of the feasibility of differential measurement via standard neutron time of flight techniques. Integral experiments would also be valuable for further uncertainty reduction, if necessary.

6.1 Other uncertainties

This paper focusses on uncertainties due to nuclear data, however a full uncertainty analysis needs to account for all significant sources of uncertainty. For a molten salt reactor, knowledge of the thermophysical properties of the salts is also key to design and future operation of the reactor. An analysis of the effect of uncertainty in salt densities shows that, for the parameters above, the output uncertainty is significant but likely smaller than that due to nuclear data. Table 3 shows estimated uncertainties assuming a 1 % uncertainty on both fuel and coolant salt density.

6.2 Design implications

At this stage of the design, the aim is to build in large margins to safety limits, to simplify design substantiation and licencing and to provide design flexibility. It must be demonstrated that uncertainties are compatible with these margins and that they do not place any undue constraints on operation of the SSR-W.

For example, commissioning of the initial core clearly must account for the uncertainty in reactivity, compensating for any calculation bias via direct adjustment of core reactivity, by adjusting the core size (i.e., the number of fuel assemblies) or by utilising other reactivity control measures. A commissioning test program should also be developed carefully to determine temperature feedback coefficients accurately by minimizing the calculation bias due to cross section uncertainties.

Reduced uncertainties would simplify commissioning, increase confidence in predictions of core behaviour and enable more optimised operation. Even without reduced uncertainty, high quality covariance data is key in being able to accurately quantify the uncertainty, reducing the extra margin needed to cover the effect of incomplete knowledge.

It should be noted that all the analysis presented here has assumed natural chlorine. Enrichment of chlorine in Cl-37 is not assumed in the SSR-W design, however it has been considered for other chloride fast reactor designs – see e.g. [20] for a discussion of chlorine enrichment – and would of course reduce the sensitivity to Cl-35 nuclear data.

6.3 Summary

The key physics parameters of the SSR-W have been shown to exhibit high sensitivity to chlorine nuclear data, particularly the Cl-35 (n,p) cross section. This sensitivity will be a feature for any fast chloride reactor. The sensitivity, together with large uncertainty in the fast region, results in significant uncertainty in important output reactor physics parameters, such as reactivity, and hence the required fuel loading for criticality.

A full sensitivity and uncertainty analysis is complicated by the lack of knowledge of Cl cross sections and their uncertainty. An uncertainty analysis has been performed without Cl cross section uncertainty data, and used to inform uncertainty analysis in safety studies.

This uncertainty analysis will be updated in future using updated covariance data, however the data in the most recent evaluated nuclear data libraries is still incomplete and could be improved.

A proposal has been prepared for addition of the Cl-35 (n,p) cross section to the NEA's High Priority Request List for nuclear data, thus highlighting it as a key application need.

Future improvement of cross sections and covariance data would be a significant benefit for the design and future operation of molten chloride salt fast reactors, which are extremely promising for future deployment, providing low cost, carbon-free electricity and addressing the issue of nuclear waste.

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