Nuclear data uncertainty challenges in Molten Salt Reactor safeguards

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INTRODUCTION

Liquid-fueled molten salt reactors (MSRs) have gained significant global interest in recent years due to the potential improvements MSRs offer (compared to current light water reactors) in terms of safety margins, fuel utilization, and economics. The Department of Energy's Office of Nuclear Energy identified through their Advanced Demonstration and Test Reactor study [1] the promise of MSRs, and the need for a demonstration reactor to advance the technology. As part of MSR development, appropriate safeguards techniques should be identified and implemented to ensure the future success of this next generation reactor.

Developing a nuclear material accounting strategy for MSRs is essential for establishing appropriate safeguards. The coupling between reactor operation, chemical processing, and continual feed and removal of nuclear material gives a unique feature of these reactors, but also presents a possible route to divert fissile material. Since the fuel and waste is liquid, there are no discrete units that can be counted in and out such as fuel rods or assemblies in a traditional reactor. A crucial challenge faced for developing an accounting strategy for nuclear material flow is the uncertainty quantification in simulations. For example, uncertainty in nuclear data libraries can be propagated throughout simulations, ultimately producing predictions which are not accurate. This work aims to quantify the uncertainty in material compositions using Sampler, a SCALE module, so that diversion of a significant quantity of fissile material can be identified with a high degree of certainty. To accomplish this, the Molten Salt Demonstration Reactor (MSDR) originally designed by Oak Ridge National Laboratory (ORNL) [2] is modeled in Serpent 2 [3] and SCALE 6.2.3 [4] codes for neutronic and isotopic analysis for actinides, fission products, and long term byproducts during the burnup of the fuel. Serpent is being used as an independent verification of the SCALE model, as well as for future use with fuel feed and removal capabilities.

THE MSDR DESIGN

The MSDR is a liquid-fueled thermal molten salt concept which consists of a cylindrical vessel filled with a matrix of graphite slabs that form flow passages for the fuel salt that circulates within the circuit [2]. The unit cell of this reactor design was described in a recent work by ORNL [5] and reproduced in Serpent 2 and SCALE. Fig. 1 shows the unit cell with dimensions in inches. The reactor core is filled with 357 unit cells and another 6 control rods cells, summing to a total of 363 cells. As for this work, some adaptations were made to simplify the geometry. Fig. 2 represents the quarter model

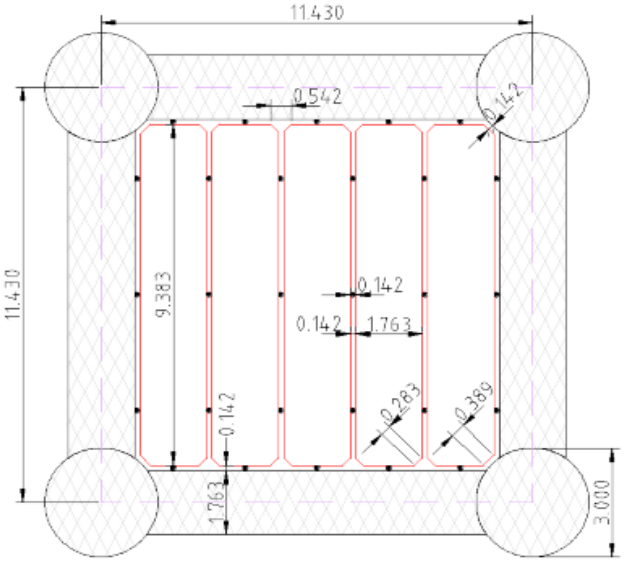

Fig. 1. Geometry of a 2D MSDR unit cell, indicated by the dashed magenta line. Dimensions are in inches. Reproduced from [5]

cell from Serpent 2. The arrows indicate the simplification of the finishing fuel salt flow passages to a flat design instead of a curved shape. This simplification is applied to all corners of the cell, both in the top and bottom. This simplification is also made in the SCALE model. The core is fueled with a low-enriched uranium (LEU) fuel in a mixture salt. The UF_4 fuel salt is dissolved in the LiF carrier salt with a molar fraction of 27.5%. The isotopic enrichment of ^{235}U and ^{7}Li are, respectively, 4.95% and 99.995%. The reason for the high enriched lithium is due to the considerable ^{6}Li absorption cross section for neutrons in the thermal energy range. With a mixture density of 4.71 g/cm^3, the total amount of salt in the core is 194.6 metric tons leading to a volume of approximately 41.3 m^3.

DEPLETION CALCULATIONS

Depletion calculations were carried out using the Serpent 2.1.31 Monte Carlo depletion code, as well as the T-DEPL and T6-DEPL sequences in SCALE 6.2.3. T-DEPL uses the multigroup deterministic code NEWT for flux calculations, while T6-DEPL uses the Monte Carlo code KENO-VI in either multigroup or continuous energy modes. The power density was set to 25 MW per kg of heavy metal (HM). All materials, mixture salt and graphite, were processed for 900K. Nuclear cross sections from ENDF-B/VII.1 [6], as well as its decay

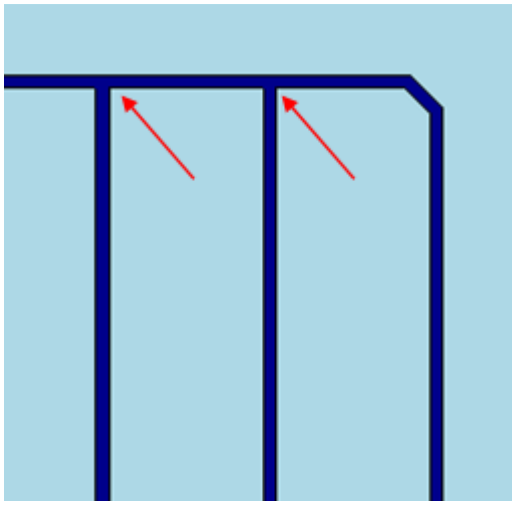

Fig. 2. Simplification of the geometry in the Serpent 2 quarter unit cell model. Arrows indicate the lack of the curved shape in the inner fuel flow passages. Fuel salt is in dark blue, and graphite is light blue.

data, were used in Serpent 2. Fission yields were from ENDF-B/VII.0 distribution [7]. This configuration of nuclear data was set in order to match the SCALE data (Although, the SCALE manual indicates that some of the fission yield and decay data are modified [4]). Other than that, values of recoverable energies (Q-values) for many actinides were defined in Serpent 2 in an attempt to match values described in the ORIGEN documentation in the SCALE manual, although the two codes use different methods to perform the power normalization. Although actual operation will involve feed of fresh fuel and online removal of fission products, these are not modeled in this initial study.

Initial criticality calculations were performed to verify the equivalency of the models in the different codes. Serpent calculations were performed with 10,000 particles and 1,000 generations while continuous energy transport solver (CE KENO) in SCALE ran using 50,000 particles and 200 generations.

Multigroup transport solvers that use Monte Carlo method (MG KENO) and deterministic method (NEWT) from SCALE were tested as well. They used the 252-group ENDF/B-VII.1 cross section library. MG cross sections are processed in SCALE through a module called CENTRM, which is responsible for self-shielding effects calculations. The description of a lattice cell model was applied using the 'symmslabcell' lattice cell option. In this unit cell calculation, the fuel width was matched to the fuel channel width (0.36068 cm), while the pitch (4.43095 cm) was selected to match the overall assembly fuel-to-moderator volume ratio. Discretization in space and angle for the NEWT quarter-assembly model were given by a 30×30 spatial mesh and a S_{10} angular quadrature order, as done previously in a similar study [5]. The Monte Carlo parameters for MG KENO were set the same as for CE KENO, mentioned before. Reflective boundary conditions were applied to all models. The Table I shows the results for k-eff from the solvers mentioned in this section a long with estimated uncertainties and differences (both in pcm) for each solver in comparison to Serpent 2.

TABLE I. Initial criticality calculation results for MSDR unit cell model in Serpent 2 and SCALE.

Solver	k_{eff}	1-σ unc. [pcm]	Δk_{eff} [pcm]
Serpent 2	1.23824	20	-
CE KENO	1.23728	27	-135
MG KENO	1.22858	24	-966
NEWT	1.22791	-	-1033

The continuous energy solvers showed agreement with around 100 pcm of difference. However, multigroup methods differ considerably from continuous energy, suggesting that the lattice cell model and/or multigroup discretization may not be sufficient for this model. This may be partly due to the difference between the lattice cell geometry (infinite slab) compared to the true assembly geometry (Fig. 1). A SCALE study of a graphite-moderated HTGR system showed a difference of approximately 300 pcm between the 252-group and continuous energy results [8]. The agreement between the deterministic and Monte Carlo multigroup results indicate that the spatial and angular discretization are adequate in NEWT.

For depletion calculations with CE KENO, 20 independent runs were performed using different starting seeds. This was done to reduce computation time as the standard SCALE distribution does not include parallel capabilities. Each run used 5,000 particles per cycle with 5 skipped cycles and 50 active cycles. This low number of skipped cycles was deemed appropriate since the small assembly size and long mean-free-path in graphite allow for very quick source convergence. Results for each run were averaged, and the central limit theorem was used to obtain the uncertainty in the mean.

Despite the fact that CE KENO and Serpent 2 agreed in the initial criticality calculation, noticeable deviations in k-eff during burnup calculations were identified. The graphic shown in Fig. 3 presents the results in k-eff for Serpent 2 and CE KENO. The absolute average difference are less than 300 pcm along the depletion interval. Uncertainties for these calculations were less than 30 pcm, and are smaller than the plot markers.

Some discrepancies in ^{235}U depletion rate were noted between CE KENO and Serpent, which suggested a difference in the effective recoverable energy per fission (Q-value) in the system. The standard Serpent calculation mode assumes a fixed energy per fission for normalization, but a new methodology using problem-dependent KERMA coefficients was recently developed [9]. SCALE uses a different methodology, a fixed library of recoverable energies for fission and capture for each isotope. The average difference between SCALE and Serpent in the change in ^{235}U atom density over the depletion interval drops from -0.28%, using the standard Serpent calculation mode, to 0.04% when using the new methodology. A comparison of the change in ^{235}U concentration from its initial value were performed and can be visualized in Fig. 4.

Plutonium isotopic concentrations from the Serpent 2, CE KENO, and NEWT models are shown in Fig. 5. The ^{239}Pu concentration in CE KENO has an average difference

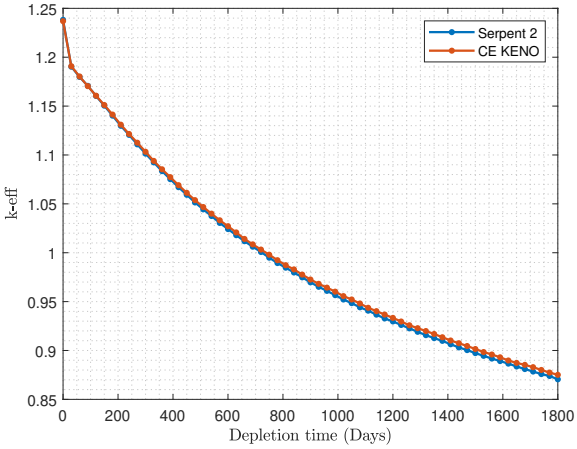

Fig. 3. k-effective for the MSDR model during a depletion interval of 5 years.

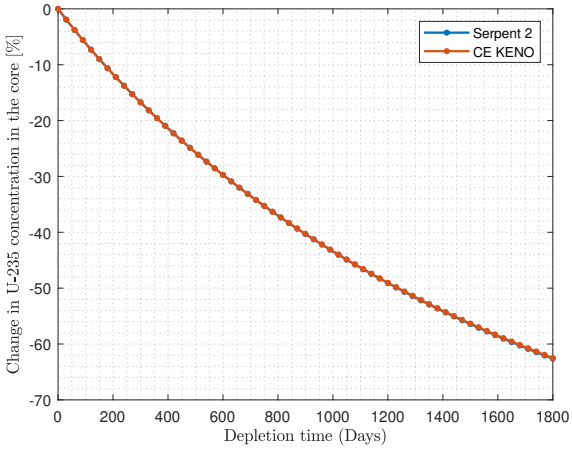

Fig. 4. Change in ^{235}U concentration from its initial value comparison between Serpent 2 and SCALE.

of 0.65% in comparison to Serpent 2. It starts 1.68% higher than Serpent after the first timestep, which decreases to 0.44% at the end of the depletion interval. These differences are currently under investigation, and may also be related to the power normalization between the codes.

NUCLEAR DATA UNCERTAINTY

Evaluation of uncertainties in modeled results is essential for nuclear material accountancy. Detection of diversion is only possible if the detected signature is outside of any experimental or modeling uncertainty. In addition to uncertainties such as material composition, dimensions, temperatures, one key driver of modeling uncertainty is due to the nuclear data (cross-sections, fission yields, and decay data). For this system, this uncertainty was estimated using Sampler [10]. In this methodology, a number of samples are taken using different sets of nuclear data perturbed according to known covariance information. Here, we used 60 samples, with perturbations on

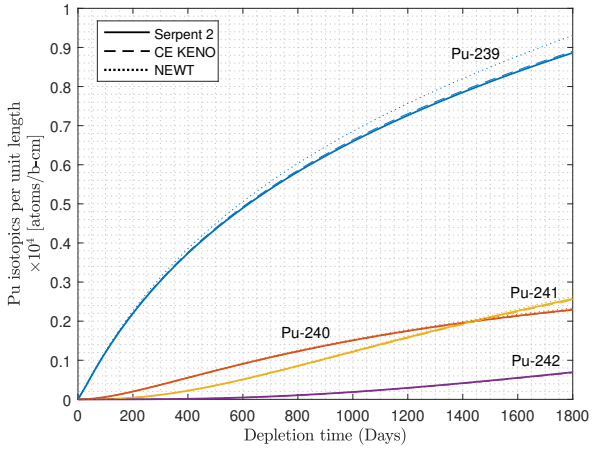

Fig. 5. Pu isotopics concentration for Serpent 2, CE KENO and NEWT during a depletion interval of 5 years.

cross-sections, decay data, and fission yields, and propagated their effect throughout the burnup cycle using the T-DEPL sequence (with NEWT as the transport solver). The uncertainties in the resulting plutonium isotopics are shown in Fig. 6. The uncertainty in ^{239}Pu content approaches 2% over the 5 year depletion cycle. Considering the entire fuel inventory, this is an uncertainty of 29 kg of total ^{239}Pu mass. Considering that a Significant Quantity as defined by the IAEA is 8kg of plutonium [11], this implies that even with no measurement or other uncertainties, a basic material accounting could have an uncertainty of several significant quantities from nuclear data uncertainty alone.

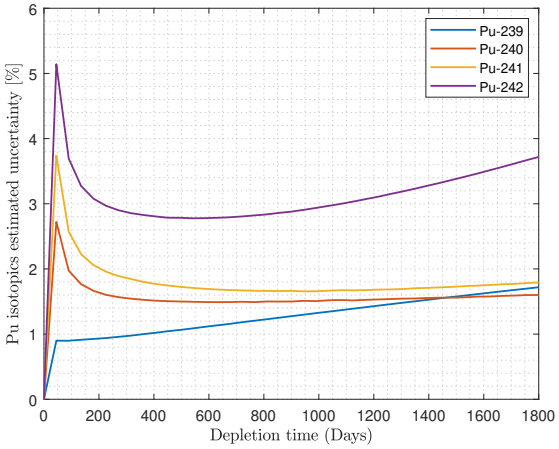

Fig. 6. Plutonium isotopic estimated uncertainty over depletion time.

CONCLUSIONS

This work presented a depletion analysis of an LEU-fueled molten salt reactor based on the MSDR design using the Monte Carlo code Serpent 2 and several sequences within the SCALE package. Multi-group results showed differences up

to 1000 pcm in k-eff compared to continuous energy, indicating limitations in the 252-group cross sections or in the lattice cell treatment in the SCALE model. Although initial criticality results between continuous energy KENO and Serpent were in agreement, the methods showed some increasing differences with burnup in k-eff, ^{235}U, and Pu concentations. The effect of nuclear data uncertainties on plutonium concentration was estimated using Sampler, which showed uncertainties up to 2% in ^{239}Pu content, resulting to an amount of 29 kg when applied to the core inventory. From a safeguards perspective, an uncertainty of 0.55% for ^{239}Pu in the MSDR model would be equivalent to 1 significant quantity. This presents a challenge for any nuclear material accounting strategy for MSRs, and alternative metrics should be sought to identify potential diversion. Future studies will also investigate the effect of online fuel reprocessing using the new feed and removal capabilities of both SCALE and Serpent.

ACKNOWLEDGMENTS

This material is based upon work supported by a Department of Energy Nuclear Energy University Program under award CFA-19-17395.

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