

Tightly Coupling of Thermochemistry Code to OpenMC for Modeling Noble Gas Stripping and Off-Gas System in Chloride-based Molten Salt Reactor

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1. Introduction

Among Generation IV reactors, the Molten Salt Reactor (MSR) is distinguished by the complex chemical behavior of the fission products dissolved in the fuel salt. This behavior governs the redox state of the salt, the release of the volatile species and, consequently, the design of the off-gas system, so that neutronic codes have to be integrated with thermochemistry codes for reactor analysis and design. The integration can be achieved either in a loosely or in a tightly coupled fashion, the former offering greater flexibility and modularity for addressing other complex multiphysics problems [1, 2].

However, in the loosely coupled approach [2, 3] the core inventories of Cs, Rb, Y, Ba and Sr — the decay daughters of Xe and Kr — are overestimated when the depletion step is sufficiently large. In that scheme the two codes are executed alternately: OpenMC advances the depletion over a full time step, Thermochemica then partitions the resulting elemental inventory into phases by solving the Gibbs energy minimization, and the amount that belongs to the gas phase is subtracted from the inventory that starts the next step. Because noble gases such as Xe and Kr are removed only at the end of each depletion step, they remain in the core throughout the step and their decay daughters are produced and retained there, which is physically inconsistent.

In the present study a tightly coupled approach [1] is applied, in which the chemical removal is embedded directly in the depletion (Bateman) equation through element-wise transfer rates so that the release proceeds continuously during burnup. The transfer rates are determined so as to reproduce the in-core noble gas inventory given by the Henry constant for the noble gases, and the thermodynamic vapor-to-feed ratio evaluated by Thermochemica for the non-noble gas elements. The approach is applied to a chloride-based MSR core connected to a simple off-gas system consisting of a decay tank and a Xe/Kr trap, and 25 years of operation are simulated.

2. Methods and Results

In this section the transfer rate formulation of the tightly coupled approach, the procedure by which the transfer rates are determined, the core and off-gas system model, and the calculated results are presented.

2.1 Transfer Rate in the Bateman Equation

OpenMC allows the continuous removal of feed nuclides from, or their feed to, one or more depletable materials by defining a transfer rate L in the Bateman equation [4]. With the chemical removal and the stream transfer terms included, the depletion equation of nuclide (Z, A) is written as

$$\begin{aligned} \frac{dN_Z^A(t)}{dt} = & \sum_{Z', A'=1}^M \left(\gamma_Z^A \sum_{g=1}^G \sigma_{f(Z', A') \rightarrow (Z, A), g}(t) \varphi_g(t) \right. \\ & + \sum_{g=1}^G \sigma_{(Z', A') \rightarrow (Z, A), g}(t) \varphi_g(t) + r_{(Z', A') \rightarrow (Z, A)} \lambda_{Z'}^A \left. \right) N_{Z'}^A(t) \\ & - \lambda_Z^A N_Z^A(t) - \sum_{g=1}^G \sigma_{a(Z, A), g}(t) \varphi_g(t) N_Z^A(t) \\ & - f_{\text{chem}, Z}^A(t) - f_{\text{stream}, Z}^A(t) \end{aligned} \quad (1)$$

where the last two terms, $f_{\text{chem}, Z}^A$ and $f_{\text{stream}, Z}^A$, represent the chemical removal from the fuel salt and the transfer between the components of the system, respectively. Both are expressed in the form $L N_Z^A(t)$, where L [1/s] is the transfer rate of the corresponding element. Because L acts inside the depletion equation, the removal is applied continuously over the time step, and the daughter nuclides produced in the off-gas components are followed by the same depletion solver.

2.2 Determination of the Transfer Rates

The essential idea of the present work is that L is not an arbitrary user input, but is determined so that the calculated inventory reproduces the equilibrium state predicted by thermochemistry. Two criteria are used, depending on the chemical nature of the element.

For the noble gases the amount retained in the fuel salt is limited by their solubility, which is evaluated from the Henry constant,

$$n_{i, \text{salt}} = K_{H, i}(T) \times p_i \times V_{\text{salt}} \quad (2)$$

where $K_{H, i}$ is the Henry constant of noble gas i at the core temperature T , p_i is its partial pressure and V_{salt} is the salt volume in the core. The transfer rate of a noble gas is adjusted until the in-core inventory obtained from

the depletion calculation coincides with the solubility limit of Eq. (2), the surplus being stripped into the off-gas system.

For the non-noble gas elements the equilibrium partitioning is provided by Thermochemica. For the given temperature, system pressure and elemental inventory, a Gibbs energy minimization is performed and the thermodynamic volatility, defined as the vapor-to-feed ratio

$$K_{t,i} = n_{i,gas} / n_{i,feed} \quad (3)$$

is obtained for every element i . The transfer rate is then adjusted until the volatility actually realized by the coupled calculation, $K_{c,i}$, defined as the fraction of element i that is removed by the transfer term during the depletion step, becomes equal to $K_{t,i}$.

The transfer rates are obtained by the following iteration. (i) The transport equation is solved by OpenMC to obtain the neutron flux and the one-group cross sections at the beginning of the step. (ii) A transfer rate is assumed for each fission element and the depletion equation including the transfer term is solved. (iii) Thermochemica is called with the resulting inventory, the core temperature and the current system pressure, and the thermodynamic volatility K_t is evaluated. (iv) The volatility K_c realized by the depletion calculation is evaluated. (v) If K_c differs from K_t , or if the solubility condition of Eq. (2) is not satisfied for the noble gases, the transfer rates are updated and the steps (ii) to (iv) are repeated. (vi) Once the criteria are met, the calculation proceeds to the next depletion step.

2.3 Core and Off-Gas System Model

The reactor model is a cylindrical chloride-salt MSR of 100 MWth whose fuel salt is 43.3 NaCl – 21.7 KCl – 35.0 UCl₃ in mole percent. The core average temperature is 580 °C and the initial system pressure is 1.5 bar. As shown in Fig. 1, the active core is surrounded by a riser/plenum region, rotating control drums and an MgO reflector, which extend to a radius of 142.6 cm, and an excore region occupies the annulus between 142.6 cm and 180 cm. The fuel salt is distributed between the in-core and the out-of-core regions with a volume ratio of 50 : 50.

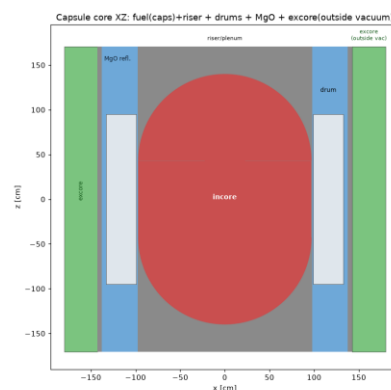


Fig. 1. OpenMC model of the chloride-based MSR core (X-Z view)

The off-gas system, shown in Fig. 2, consists of a decay tank and a Xe/Kr trap, which are modeled as additional depletable materials that are neutronically isolated from the core. The species stripped from the fuel salt are transferred to the decay tank, in which the residence time is 2 hours. Ten percent of the stream leaving the decay tank is directed to the Xe/Kr trap, whose residence time is 1 hour and in which the noble gases are retained, while the remaining ninety percent is returned to the out-of-core (excore) region of the fuel loop.

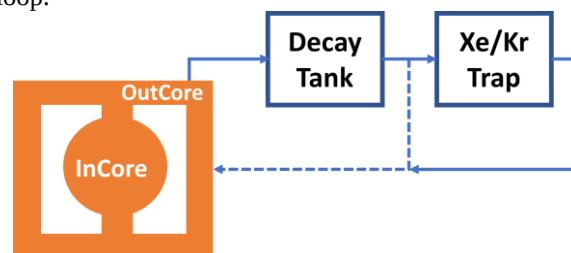


Fig. 2. Core and off-gas system model

OpenMC v0.15.2 with the ENDF/B-VIII.0 library was employed. Each transport calculation uses 50,000 particles with 250 batches, of which 100 are inactive. The operation period is 25 years and the depletion step is 1 year. MSTDB-TC v4.0 together with FactPS, supplemented by a small number of additional species, was used as the thermodynamic database.

2.4 Neutronic Results

Figure 3 compares the effective multiplication factor obtained with the tightly coupled calculation and with a reference calculation in which no chemical removal is considered. The tightly coupled result remains above the reference over the whole operation period, because the neutron absorbers that are continuously stripped from the fuel salt no longer contribute to the parasitic absorption in the core. At the end of the 25-year cycle k_{eff} is 1.00154 for the tightly coupled case and 0.99769

for the case without coupling, which corresponds to an excess reactivity gain of 385 pcm.

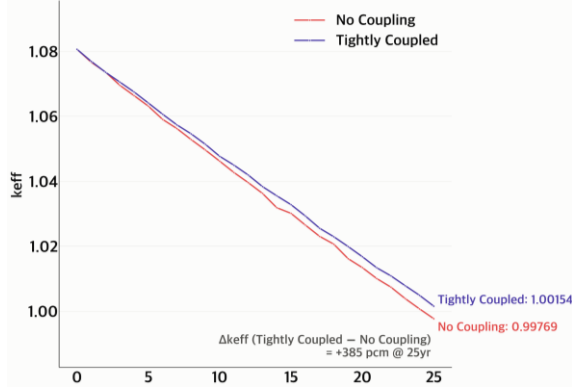


Fig. 3. Comparison of k_{eff} between the calculations without coupling and with tight coupling

2.5 Inventory of the Core and the Off-Gas System

Table I lists the ten most abundant chemical species of the molten salt solution phase in the core at the 1st and at the 25th year of operation. UCl_4 and $ZrCl_4$ are the dominant species over the whole period, followed by the trichlorides of the lanthanides and by the plutonium chlorides. The alkali and alkaline-earth chlorides, $CsCl$ and $BaCl_2$, remain comparatively small because a part of their precursors, Xe and Kr, is stripped from the core before they decay.

Table I: Ten most abundant chemical species in the core (mol)

Chemical species	1st year	25th year
UCl_4	95.5	2,538.3
$ZrCl_4$	50.6	1,280.9
$Pu[CN=VI]Cl_3$	38.0	317.2
$NdCl_3$	28.2	845.9
$CeCl_3$	26.3	494.7
$Pu[Dimer]Cl_6$	17.0	704.3
$SrCl_2$	9.5	159.2
$LaCl_3$	8.9	218.6
$CsCl$	4.1	79.8
$BaCl_2$	2.9	82.9

Table II gives the elemental inventories accumulated in the two off-gas components. The decay tank is dominated by Cs, Ba, Sr, Rb and Y, which are not gaseous themselves but are produced by the decay of the Xe and Kr isotopes while these reside in the tank. Since they are not carried away by the gas stream, they accumulate there. The Xe/Kr trap holds 912.5 mol of Xe and 140.5 mol of Kr at the 25th year, together with 364.3 mol of Cs produced by the decay of the trapped Xe. Such a build-up of daughter nuclides in the off-gas components cannot be reproduced by a loosely coupled approach, in which the noble gases are removed as a lump at the end of each depletion step and their subsequent decay outside the core is not followed.

Table II: Elemental inventory of the off-gas components (mol)

Component	Element	1st year	25th year
Decay tank	Cs	13.9	312.9
	Ba	8.8	254.8
	Sr	5.9	123.3
	Rb	4.9	117.2
	Ar	4.3	111.7
	Y	2.6	76.6
Xe/Kr trap	Xe	36.1	912.5
	Cs	14.2	364.3
	Kr	6.0	140.5
	Rb	0.76	24.5
	Sr	0.91	21.9

The continuous release of gas into the off-gas system raises the system pressure. As shown in Fig. 4, the core pressure increases almost linearly from 1.59 bar after the first year of operation to 3.72 bar at the end of the 25th year, which is a direct consequence of treating the release as a continuous process rather than as an end-of-step correction.

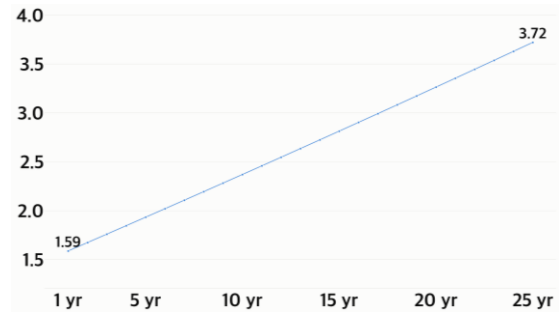


Fig. 4. Evolution of the core system pressure during 25 years of operation

Table III summarizes the state of the fuel salt at the end of the cycle. The total fuel mass decreases from 41,633 kg to 41,021 kg, that is, 612 kg are transferred from the fuel salt to the off-gas system during 25 years of operation. The UCl_4/UCl_3 mole ratio, a representative measure of the redox state of MSR salts, reaches 0.0312, indicating that the continuous stripping of the volatile species drives the fuel salt toward an oxidizing state far more strongly than the loosely coupled evaluation predicted.

Table III: State of the fuel salt at the end of the 25-year cycle

Quantity	Value
Total fuel mass (BOC)	41,633 kg
Total fuel mass (EOC)	41,021 kg (-612 kg)
$PuCl_3$ mass	604.7 kg (1.47 %)
UCl_4 mass	964.2 kg (2.35 %)
UCl_4/UCl_3 mole ratio	0.0312

In Table III the $PuCl_3$ mass is the sum of $PuCl_3$ and Pu_2Cl_6 , and the UCl_3 amount used for the mole ratio is the sum of UCl_3 and twice the amount of U_2Cl_6 .

3. Conclusions

A tightly coupled approach between OpenMC and the Python-based Thermochemica was developed for a chloride-based MSR. The chemical removal is represented by the transfer term of the Bateman equation, and the transfer rate of each element is determined so that the calculated in-core inventory reproduces the solubility given by the Henry constant for the noble gases and the thermodynamic vapor-to-feed ratio evaluated by Thermochemica for the non-noble gas elements. In contrast to the loosely coupled approach, the release proceeds continuously during burnup and the decay daughters produced in the off-gas components are tracked by the same depletion solver.

The approach was applied to 25 years of operation of a chloride-based MSR connected to a simple off-gas system composed of a decay tank and a Xe/Kr trap. The continuous stripping yields an excess reactivity gain of 385 pcm at the end of the cycle, 612 kg of material are transferred from the fuel salt to the off-gas system, the core pressure rises from 1.59 bar to 3.72 bar, and the $\text{UCl}_4/\text{UCl}_3$ mole ratio of the fuel salt reaches 0.0312. Further work will address the sensitivity of the results to the depletion step size, to the Henry constants and to the thermodynamic database, as well as a more detailed model of the off-gas system.

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REFERENCES

- [1] S. A. Walker, M. E. Tano, A. Abou-Jaoude and O. Calvin, Depletion-driven thermochemistry of molten salt reactors: review, method, and analysis, *Frontiers in Nuclear Engineering*, Vol.2, p. 01, 2023.
- [2] M. E. Tano, S. A. Walker, A. Abou-Jaoude, R. Roper, T. Karlsson, M. C.F. Karlsson, P. Bajapi, R.d. Oliveira, R. Freile, V. Kyriakopoulos, M. K. Jaradat, P. Sabharwall, Coupled neutronics, thermochemistry, corrosion modeling and sensitivity analyses for isotopic evolution in molten salt reactors, *Progress in Nuclear Energy*, Vol.178, p.105503, 2025.
- [3] J. H. Lee, C. -H. Lee, Loosely Coupling of Thermochemistry code to OpenMC for Chloride-based Molten Salt Reactor, In *Transactions of the Korean Nuclear Society Autumn Meeting*, Korean Nuclear Society.
- [4] P. K. Romano, N. E. Horelik, B. R. Herman, A. G. Nelson, B. Forget, K. Smith, OpenMC: A state-of-the-art Monte Carlo code for research and development, *Annals of Nuclear Energy*, Vol.82, p. 90, 2015.