

Development of Preliminary Material Balance Evaluation Methods for the Core and Off-Gas Systems during Molten Salt Reactor Operation

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1. Introduction

The inventory of the fission products generated during molten salt reactor (MSR) operation and distributed between the core and the off-gas system is a primary input for the design of the off-gas system and for the source term evaluation. In an MSR a part of the fission products is continuously removed from the fuel salt and accumulates in the off-gas components, so that the inventory has to be evaluated for the core and the off-gas system rather than for the core alone.

Such inventories can be obtained from a multiphysics framework in which a depletion solver is coupled with a thermochemistry solver [1,2]. Building such a framework, however, requires a considerable amount of time and effort. The equilibrium solver has to be called at every depletion step, and the resulting computational cost limits the number of design variants that can be examined at the conceptual design stage.

The distribution of the fission products between the core and the off-gas system is governed mainly by the noble gas stripping rate. The noble gases are the species that are removed from the fuel salt at a significant rate, and because Kr and Xe are the precursors of Rb, Sr, Y, Zr, Cs, Ba, La and Ce daughters, their removal also determines the distribution of daughter fission products. The stripping rate therefore acts as the dominant parameter of the material balance, while the remaining elements largely follow the partition established by their noble gas precursors.

In the present work a material balance evaluation method is proposed that predicts the core and off-gas inventories as a function of the noble gas stripping rate.

2. Methods and Results

In this section the physical assumptions of the material balance model, the interpolation of the fission product inventories with respect to the noble gas stripping rate, the overall calculation procedure, and the calculated results are presented.

2.1 Physical assumptions of the material balance model

Fig. 1 shows the core and the off-gas system considered in this work. The fuel salt circulates between the in-core and the ex-core volumes. A branch β of the ex-core stream is sent to a gas-liquid separator in which a fraction f of the noble gases is removed, so that the

effective stripping efficiency of the system is $\varepsilon = \beta f$. The removed gas enters the off-gas system, which consists of a decay tank and a Xe/Kr trap, and the carrier gas is returned to the core.

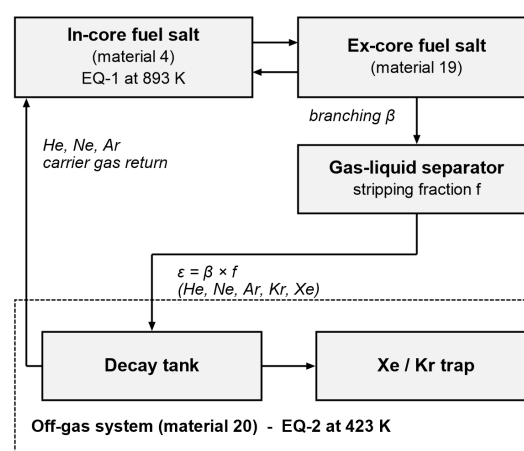


Fig. 1. Core and off-gas system considered in the material balance model.

The model is based on the following assumptions. (1) Only the noble gases (He, Ne, Ar, Kr and Xe) are removed directly from the fuel salt. The other elements leave the salt only as the decay products of the noble gases that have already been transferred to the off-gas system. (2) The fission rate and the neutron spectrum are not affected by the stripping, so that the production rate of each nuclide is the same in the two bounding depletion calculations described in Section 2.2. (3) The fuel salt is treated as two well mixed volumes, and the gas-liquid separator acts on the ex-core volume. (4) The chemistry is evaluated at the element level. Because thermodynamic equilibrium does not distinguish isotopes, the interpolated isotopic inventories are summed to elements before the equilibrium calculation. (5) Two equilibrium states are evaluated in every sub-step, the core at the fuel salt temperature (EQ-1) and the off-gas system at the trap temperature (EQ-2). The carrier gas returned to the core consists of He, Ne and Ar, and its inventory is taken from the interpolated off-gas inventory. (6) The off-gas volume is a fixed ideal gas reservoir whose reference mole number is the initial He inventory of the off-gas system in the depletion model (Table II), and the system pressure is scaled by the number of moles of the gaseous compounds that

have accumulated in the off-gas system. (7) The noble gases retained in the decay tank and in the trap and their decay daughters are outside the thermodynamic system and are followed by a simple inventory ledger instead of an equilibrium calculation.

2.2 Interpolation of the fission products with respect to the stripping rate

Two depletion calculations are carried out in advance with OpenMC [3]. In the first one the noble gases are not stripped ($\epsilon = 0$) and in the second one they are transferred to the off-gas system at the maximum rate ($\epsilon = 1$). They give, for every nuclide, the inventory of the in-core plus ex-core salt and the inventory of the off-gas system before burnup, at the end of the first year and at the end of the last year. The inventory at an arbitrary ϵ is then reconstructed nuclide by nuclide, and three groups are distinguished according to the way in which the nuclide leaves the salt. Table I compares the two limits and the interpolated result at $\epsilon = 0.9$ for representative elements in a 100 MW_{th} chloride-based MSR fueled with 43.3 NaCl–21.7 KCl–35 UCl₃ (mol %) at the end of the 30th year of operation.

Table I: Element inventories of the core and the off-gas system at the end of the 30th year (mol). The off-gas inventory vanishes at $\epsilon = 0$ and is not listed.

Element	$\epsilon = 0$		$\epsilon = 1$		$\epsilon = 0.9$	
	Core	Core	Off-gas	Core	Off-gas	Off-gas
Xe	1101	4.55×10^{-4}	1096	5.02×10^{-4}	1096	
Kr	172.0	1.28×10^{-4}	172.1	1.40×10^{-4}	172.1	
Rb	175.2	3.957	172.0	4.315	171.6	
Sr	354.0	163.7	189.6	166.4	186.9	
Y	219.7	98.30	121.5	103.6	116.2	
Zr	1563	1543	19.75	1545	18.01	
Cs	877.1	85.20	812.8	90.77	807.2	
Ba	437.8	99.19	322.5	105.3	316.5	
La	316.3	262.1	54.30	266.4	49.96	
Ce	605.3	591.0	14.11	592.3	12.80	
I	40.90	40.89	7.63×10^{-13}	40.89	6.86×10^{-13}	
Te	114.8	114.8	2.02×10^{-17}	114.8	1.81×10^{-17}	

The first group contains the nuclides that are not affected by the stripping. Their inventory is obtained by a linear blend of the two bounding inventories N_0 and N_1 ,

$$N(\epsilon) = (1 - \epsilon) N_0 + \epsilon N_1 \quad (1)$$

The second group contains the noble gases, which are removed directly. Their inventory is governed by the balance between the decay constant and the removal rate constant, $N = R / (\lambda + \epsilon \lambda_s)$, where R is the production rate. Since the reciprocal of the inventory is linear in ϵ , the interpolation is carried out harmonically,

$$N(\epsilon) = N_0 N_1 / [\epsilon N_0 + (1 - \epsilon) N_1] \quad (2)$$

Equation (2) returns N_0 at $\epsilon = 0$ and N_1 at $\epsilon = 1$, and it is exact when the nuclide reaches saturation within a

depletion step, which is the case for the noble gas isotopes that carry the fission product chains.

The third group contains the decay daughters of Kr and Xe, that is the Rb, Sr, Y and Zr chain and the Cs, Ba, La and Ce chain. These nuclides are not stripped, but the place in which they are produced follows the inventory of their precursor. For a precursor at saturation the production rate of the daughter is proportional to the precursor inventory, so that the daughter inventory is written as a linear function of the precursor inventory,

$$N_d(\epsilon) = A + B N_p(\epsilon) \quad (3)$$

The coefficients A and B are determined from the two bounding calculations, and $N_p(\epsilon)$ is the precursor inventory that has already been obtained from Eq. (1) or Eq. (2). The chains are evaluated in the order of the decay sequence so that the precursor inventory is always available. Equation (3) uses the standing inventory of the precursor as the independent variable, which is equivalent to the cumulative amount of the precursor that has decayed as long as the precursor reaches saturation within a depletion step.

2.3 Calculation procedure

Fig. 2 summarizes the calculation procedure. The two bounding depletion calculations and the interpolation are carried out once. For the required ϵ the inventory of every year is then obtained, and each year is divided into K sub-steps in which the two equilibria are evaluated with Thermochemica [4].

2.4 Result

We applied the strategy outlined in Fig. 2 to identify the chemical compounds and their inventories in the core and in the off-gas system, which consists of a decay tank and a Xe/Kr trap. The isotopic inventories were estimated with OpenMC 0.15.2 using the predictor (first-order explicit Euler) depletion integrator, and MSTDB-TC v4.0 was employed as the thermodynamic database required by Thermochemica. The calculation conditions required for calculation are listed in Table II.

Table II: Input Parameters required for material balance model

Parameters	Values
Initial Pressure	1.5 bar
Initial inventory of He	115.91 mol
Number of K sub-steps per year	20
ϵ	0.90

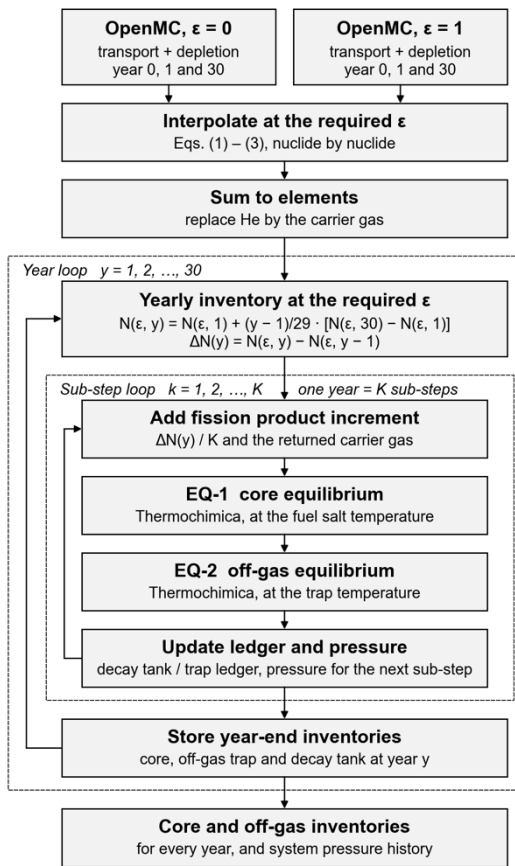


Fig. 2. Flowchart of the material balance calculation. The inventories of every year are reconstructed at the required ϵ from the two bounding runs, and each year is divided into K sub-steps in which the two equilibria are evaluated.

Tables III to V summarize the composition obtained at the end of the 30th year for NaCl-KCl- UCl_3 based MSR with 20 sub-steps per year. Table III lists the major species of the molten salt phase in the core with the three carrier salt components NaCl, KCl and UCl_3 excluded. UCl_4 is the largest of the remaining species, which reflects the oxidation of the fuel salt as the fission products take up chlorine, and it is followed by the tetrachloride of Zr and by the trichlorides and the dichlorides of the lanthanides and of the alkaline earth elements. Table IV lists the condensed phases that accumulate in the off-gas trap. ZrCl_4 dominates, and the remaining solids are the Cd, Sn and Ge compounds that condense at the trap temperature. Table V lists the element inventory of the decay tank and Xe/Kr Trap, which is followed as a simple ledger instead of an equilibrium calculation and is therefore dominated by the stripped noble gases and by their decay daughters Cs, Rb, Sr and Ba.

Table III: Major species of the core molten salt phase at the end of the 30th year (mol). The carrier salt components NaCl, KCl and UCl_3 are excluded.

Species	Inventory (mol)
UCl_4	3046.6
PuCl_3	1481.7
ZrCl_4	1329.3
NdCl_3	1013.1
CeCl_3	592.3
BaCl_2	105.3
LaCl_3	266.4
CsCl	90.7

Table IV: Condensed phases accumulated in the off-gas trap at the end of the 30th year (mol). The phase names are those of the thermodynamic database.

Species	Inventory (mol)
ZrCl_4	200.4
CdCl_2	4.012
Sn(wh)	3.432
CdSb	1.958
CdTe	0.681
CdSe	0.525
Ge	0.175
Sb	0.116

Table V: Element inventory of the decay tank at the end of the 30th year (mol).

Element	Inventory (mol)
Xe	1096.4
Cs	807.2
Ba	316.5
Sr	186.9
Kr	172.1
Rb	171.6
Y	116.2
La	50.0

3. Conclusions

A material balance evaluation method has been developed that predicts the core and off-gas inventories of a molten salt reactor as a function of the noble gas stripping rate. Two bounding depletion calculations, one without stripping and one with complete stripping, are performed once, and the inventory at an arbitrary stripping rate is reconstructed nuclide by nuclide with three interpolation rules that follow the way in which each nuclide leaves the salt. The interpolated inventories are summed to elements and advanced through a sub-step loop in which the core and the off-gas equilibria are evaluated with Thermochemica, so that no further depletion calculation is required for each stripping rate.

The method was applied to a chloride-based molten salt reactor connected to a decay tank and a Xe/Kr trap. The noble gases are transferred to the off-gas system almost completely, and their decay daughters follow the same partition with an effect that weakens along each decay chain, so that Cs, Ba and Sr are strongly

redistributed while Zr and Ce remain in the salt. In the fuel salt UCl_4 is the largest species after the carrier salt components, ZrCl_4 dominates the solids retained in the trap, and the decay tank inventory is dominated by Xe, Cs and Kr. Further work will compare the interpolated inventories with a fully coupled depletion and thermochemistry calculation, and will examine the linear chain interpolation for precursors that do not reach saturation within a depletion step.

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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] 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.
- [4] M. H. A. Piro, S. Simunovic, T. M. Besmann, B. J. Lewis, W. T. Thompson, The thermochemistry library *Thermochimica, Computational Materials Science*, Vol.67, p. 266, 2013.