A New Reactivity Control Approach for Circulating Fuel Reactors

Giulia Merla, Antonio Cammi, Stefano Lorenzi
Politecnico di Milano
Via La Masa 34
20133, Milano, Italy
giulia.merla@mail.polimi.it, antonio.cammi@polimi.it, stefano.lorenzi@polimi.it

ABSTRACT

Molten Salt Reactors (MSRs) are nuclear power systems conceived to be working with a circulating salt-mixture, acting simultaneously as fuel and as coolant, providing safety and sustainability enhancements, other then improvements in the fuel cycle closure. The presence of a circulating fuel features not only the possibility of a continuous Fission Products (FPs) removal, but implies also the chance to perform tailored intervention on the fuel isotopic composition, adjusting the mixture as needed. These peculiarities prevent the adoption of commonly available simulation tools, which are designed for solid-fuelled reactors and lack in general the possibility to account for mass exchange processes within burn-up calculations. To overcome these limitations, a Serpent-2 extension is presented to couple depletion calculations and material transfer, thus featuring the capability to simulate FPs removal and continuous composition adjustments for both reactivity and eutectic control in MSR. The reactivity control is implemented with a new strategy allowing the independence from the chosen fuel treatment strategy and a limited impact on both the system mass and the eutectic proportions. The new implemented functionalities are verified and proven to work correctly, while the new reactivity control strategy is compared to an alternative one from previous studies, proving the superior performances of the new proposal.

1 INTRODUCTION

The concept of Molten Salt Reactors (MSRs) has raised the scientific interest for its remarkable advantages in terms of safety, sustainability and proliferation resistance, together with important improvements in the fuel cycle closure. In this scenario, the Molten Salt Fast Reactor (MSFR) has been identified as baseline concept for MSRs in the framework of Generation IV International Forum [1], and are currently under study and development in the framework of the SAMOSAFER European Project (https://samosafer.eu/). The MSFR concept is designed to work with an intermediate-fast neutron spectrum and a eutectic salt mixture of LiF-ThF$_4$-UF$_4$ (77.5-20-2.5 % mol) characterized by an excellent heat capacity and a melting point of 565 °C, thus allowing high operational temperatures at atmospheric pressure and the consequent high power efficiency. The presence of the liquid fuel also allows for a continuous removal of Fission Products (FPs) adding the possibility to monitor and adjust the salt composition directly during the salt reprocessing. This peculiarity of the MSFR features the capability to perform tailored control interventions on the mixture aimed at preserving the physical-chemical properties of the salt, its eutectic composition or even to tune the system reactivity. However, this characteristic prevents the use of conventional simulation tools for the analysis of MSFR

and calls for the development of an advanced code capable to coupling burnup calculations and material exchange processes also in a framework of continuous control interventions. An extension for the SERPENT-2 code is therefore presented in this work to feature the capability to control eutectic and reactivity via composition adjustment, thus stepping forward in the MSFR modelling.

2   SERPENT-2 CODE EXTENSION

The MSFR technology offers the possibility to feature a continuous fuel reprocessing not only as an effective method to clean the salt and tune its physico-chemical properties, but also allowing to control eutectic composition - in terms of heavy metals or actinides fraction - and long term reactivity oscillations by simply adjusting the fissile/fertile ratio in the stream. The presence of a continuous control intervention in a context of burn-up calculations prevents, from a modelling point of view, the use of conventional simulation tools which, being mainly developed for solid-fueled reactors, do not account in general for a continuous adjustment in the isotopic composition. Therefore, with the objective to accurately model the MSFR characteristic reprocessing, a new reactivity control approach for MSR and its relative implementation in the SERPENT-2 code is proposed. In particular, the tuning of reactor composition at its chemical level features, other than reactivity control itself, also the capability to impose and keep constant the eutectic proportions in the fuel. Making use of past contributions on this subject [2], the approach is therefore implemented thanks to an optimized eutectic control procedure and a new reactivity control scheme, whose performances are then tested and verified to prove the new algorithm capabilities.

2.1   MSFR Reprocessing

During reactor operation, the accumulation of fission products can strongly impact on neutronics, corrosion and thermal properties of the salt. In the MSFR, a selective extraction of these elements is foreseen mainly through two mechanisms, namely an on-line gas bubbling and an on-site batch reprocessing. The former consists in contacting the fuel mixture with an inert gas, typically helium, both for the removal of non-soluble fission products via dilution in the carrier gas, and the partial extraction of metallic particles through capillarity sticking to the bubbles [3]. The process results in a fast characteristic removal time of about 30 s which, however, diversifies depending on the specific extraction efficiency for each element. The on-site reprocessing instead, consists in a chemical treatment of the fuel, focused on the removal of soluble fission products, with a maximum rate of 40 l/d of salt reprocessed. The fuel is extracted, during the reactor operation, from the expansion tank and the salt is reprocessed off-line through the Fuel Treatment Unit (FTU) to control the physico-chemical properties of the mixture, to maintain constant the salt eutectic composition, and consequently its melting point, to separate FP and actinides, to eventually modify the fertile/fissile ratio, in order to handle long-term reactivity swings in the reactor. This fuel treatment process exhibits a cycle time of 450 days for the fuel, based on the maximum daily reprocessing rate and a total salt volume respectively of 18 m^3.

2.2   Eutectic Control

In order to feature the capability to simulate the peculiar reprocessing scheme of a circulating fuel reactor such as the MSFR, both standard code functionalities and new extensions are exploited. The FPs removal featured by both the Fuel Treatment Unit (FTU) and the Gas

Bubbling (GB) system is modeled through standard code capabilities as added decay terms in the Bateman equations, provided the proper decay constant $\lambda_{repro,i}$, as follows:

$$\frac{dN_i}{dt} = \sum_j N_j \phi \cdot \sigma_{j \to i} - \sum_j N_i \phi \cdot \sigma_{i \to j} + \sum_j N_j \lambda_j b_{j \to i} - N_i(\lambda_i + \lambda_{repro,i}) \qquad (1)$$

where ϕ is the neutron flux, N_i is the atomic density of nuclide i, λ_i and $\sigma_{j \to i}$ are respectively the decay constant of nuclide i and the one-group transmutation cross section from nuclide j to nuclide i; similarly, $b_{j \to i}$ expresses the branching ration of nuclide j to nuclide i. The specific reprocessing rate $\lambda_{repro,i}$ is defined as the product between the nuclide transfer coefficient, expressing the treatment efficiency on the species, and the characteristic rate of the treatment, related to the selected reprocessing strategy.

On the other hand, one of the main tasks of the on-site batch reprocessing (FTU) is to ensure the maintenance of the eutectic composition in the fuel mixture during the reactor operation. Therefore, a possible strategy to implement the eutectic control function of the FTU is to keep constant the Heavy Metal (HM) molar proportion and taking as Light Fraction (LF) all elements with $Z < 89$. The proposed control intervention is designed to replace the HM fissioned fraction with a $Th^{232}F_4$ feed, while reprocessed FPs are replaced by LiF. From a modelling point of view, this is translated in an intervention on off-diagonal terms in the burn-up matrix, modeled by adding a non proportional term in the right hand side (RHS) of Eq. (1) to express the injection of feed material proportional the fissioned heavy metals (Eq. 2):

$$\sum_{k=HM} N_k \phi \cdot \sigma_{k,f} \qquad (2)$$

The equation for the feed nuclide h becomes:

$$\frac{dN_h}{dt} = \sum_i (\phi \cdot \sigma_{i \to h} + \lambda_{i \to h})N_i - \sum_i (\lambda_{h \to i} + \phi \cdot \sigma_{h \to i})N_h$$
$$- \phi \cdot \sigma_{h,f} N_h + \sum_{k=HM} \phi \cdot \sigma_{k,f} N_k \qquad (3)$$

Additionally, as fission products are reprocessed, lithium is progressively added to the mixture and, conversely, when FPs are produced, lithium is removed, in the mindset to maintain constant the eutectic composition of the salt. The resulting equation for the lithium density is:

$$\frac{dN_{Li}}{dt} = \sum_j (\phi\, \sigma_{j \to Li} + \lambda_j b_{j \to Li})N_j - \sum_j \phi\, \sigma_{Li \to j} N_{Li}$$
$$- \lambda_{repro,Li} N_{Li} + \sum_{l=FP} \lambda_{repro,l} N_l - \sum_{k=HM} N_k \phi\, \sigma_{k,f} \cdot \sum_{l=FP} Y_{k \to l} \qquad (4)$$

where $Y_{k \to l}$ is the yield of the product l from a fission of the heavy metal k and its sum, extended over all FPs, is usually approximated as 2. The resulting summatory extended over HM expresses the lithium removal in correspondence of FPs production, whereas the term $\sum_{l=FP} \lambda_{repro,l} N_l$ corresponds to the lithium replacement of the reprocessed fission products. In this work, a fluorine consumption rate of zero is imposed in order to limit the regulation effect to Li and Th feeds. Finally, the proposed eutectic control is implemented within the burn-up

routine, purposely modified to be capable to run sub-stepping and predictor corrector calculations even in presence of material exchange processes. Placed in a pre-existing algorithm, this control routine fits in an preexisting routine and it is consequently already optimized both for calculation time and CPU usage.

2.3 Reactivity Control Implementation

The MSFR concept is designed without any control rod, therefore the possibility to handle long term reactivity oscillations directly by adjusting the fuel composition represents a very fundamental strength pledge by the on-line reprocessing. The need to model and study the impact of this feature is fulfilled by implementing an iterative reactivity control algorithm based on a two-elements control approach using UF_4 and ThF_4 as feed nuclides. The composition adjustment is performed with a new developed strategy, injecting and extracting the feed always in equal molar quantities. The main objective is to obtain an efficient control intervention without impacting both on the total system mass and eutectic proportions, providing at the same time a flexible simulation tool capable to regulate the system reactivity independently from its starting condition.

The reactivity control algorithm is implemented at the level of the burn-up matrix construction to provide a continuous intervention and adding the sub-steps routine to obtain a finer control action. The operation of the algorithm strongly relies on the capability to track the multiplication factor evolution at each sub-step, where the k_{eff} evaluated in SERPENT through the conventional transport cycle is not available. Consequently, at each sub-step, an implicit-like estimation of the infinite multiplication factor is performed by the algorithm, each time corrected by a leakage factor depending on the real k_{eff} calculated by SERPENT. At the beginning of each main burn-up step, a transport calculation is performed and the resulting k_{eff} is used to calculate the the Beginning Of Step (BOS) and End Of Step (EOS) leakage correction factors. Such corrections are indicated as *leakage corrections* (μ) although they do not represent a real leakage, since they do not come directly from a transport calculation. At each substep, the leakage correction is then obtained by linear interpolation between the BOS and EOS ones. Afterwards, the Bateman equations are solved, providing the system composition at the end of sub-step and assessing the sub-step k_{eff}, which is accepted only if sufficiently close to the user-defined target ($|k_{eff} - k_{tgt}| < \epsilon$). Otherwise, the feed nuclides are adjusted accordingly, and the depletion matrix is built and solved once again, until convergence between the effective and the target multiplication factors is met. Once the iteration is completed, the final composition is saved and used as starting point for the next sub-step. The overall procedure can be summarized in the following:

1. Initialization burn-up step. For each step:

2. Transport calculation for k_{eff} prediction and evaluation of the transmutation cross sections

3. Initialization sub-step routine. For each sub-step:

    (a) Construction of the depletion matrix and k_{inf} estimation

    (b) Calculation of BOS and EOS leakage corrections and record the EOS one for the next burn-up step (only in the first sub step)

    (c) Interpolation of the leakage correction for the current sub-step (μ_L)

    (d) Solution of the Bateman equations (CRAM method)

(e) If $|k_{tgt} - k_{eff}| < \epsilon$

   i. Save the final composition for this sub-step, to be used as starting composition in the next

   ii. Move to the next sub-step

(f) Else ($|k_{tgt} - k_{eff}| > \epsilon$)

   i. Calculate the refill factor for composition adjustment

   ii. Add the opportune injection/extraction rates to fissile/fertile material

   iii. Repeat steps (a) to (e) (if saturation is reached within two iterations, move to the next sub-step)

4. Using the final compositions, run a second transport calculation (Monte Carlo)

5. Interpolate appropriately the homogenized cross sections, depending on the selected predictor-corrector option

6. Perform the corrector calculation: repeat step (3), from (a) to (f), then move to the next burn-up step

The feed nuclides are handled with a Semi-Diagonal control strategy, always injecting and removing an equal amount of material, with a net atomic density change equal to zero. From a modelling point of view, this effect is obtained by adding a diagonal term for one of the two feed nuclides, and an equal off-diagonal term for the other. Doing so, the total system mass (and eutectic proportions) is almost preserved, apart from a marginal mass change due to the difference in U-233 and Th-232 atomic masses. In particular, the insertion rate is calculated within each sub-step from a fixed, user-defined, maximum rate of injection (λ_{max}^{rc}) and an estimation of a k_{eff} partial derivative with respect to the refill factor itself. This refill factor (f_{rc}) in particular is assessed iteratively, bounded to the interval [-1,1], and it is multiplied, equipped with sign, by λ_{max}^{rc} to determine the final refill rate:

$$\lambda_{rc} = \pm f_{rc}\lambda_{max}^{rc} \tag{5}$$

Positive rates represent subcritical conditions, therefore correspond to an injection of ^{233}U associated with an extraction of ^{232}Th and vice versa for negative values. The resulting rates are used in the Bateman equations with a Semi-Diagonal control approach, choosing to work proportionally to the thorium share, thus with a diagonal intervention on the low feed (Th, Eq. (7)) and an off-diagonal one for the high feed (U, Eq. (6)), always providing an equal molar adjustment of the two.

$$\frac{dU^{233}}{dt} = \sum_j N_j \phi \cdot \sigma_{j,\to U^{233}} - \sum_j U^{233} \phi \cdot \sigma_{U^{233},\to j} - (\sigma_f + \lambda) \cdot U^{233} + \lambda_{rc} \cdot Th^{232} \tag{6}$$

$$\frac{dTh^{232}}{dt} = \sum_j N_j \phi \cdot \sigma_{j,\to Th^{232}} - \sum_j Th^{232} \phi \cdot \sigma_{Th^{232},\to j} - (\sigma_f + \lambda + \lambda_{rc}) \cdot Th^{232} \tag{7}$$

The choice to apply the diagonal control on thorium is driven by its higher concentration which implies larger insertion/extraction rates for U-233 and therefore a more effective control intervention, especially in terms of convergence speed to the target reactivity. The new Semi-Diagonal approach therefore allows for a continuous and effective reactivity control intervention

without interference with the eutectic point and implicitly balancing the fluoride contributions associated to the feed nuclides, which are actually handled in the forms of tetrafluorides. Furthermore, the need for a prior search of a starting composition close to the target reactivity is bypassed with the new EOS leakage correction allowing, together with the routine disentanglement the eutectic control, a higher tool flexibility.

2.4 Functionalities Verification

The implemented features and code performances are tested and verified on the toroidal reactor configuration in Figure 1. Simulations are performed starting from the initial fuel composition in Table 1, with the LELI predictor corrector option [4], employing 50 sub-steps and 16 processors in the OMP parallelization mode. The simulated time covers a twenty years of burn-up normalized to a full power of 3 GW and employing a value of $2.5720 \cdot 10^{-8} \ s^{-1}$ for the reactivity control maximum refill rate λ_{max}^{rc} .

Table 1: Initial fuel composition

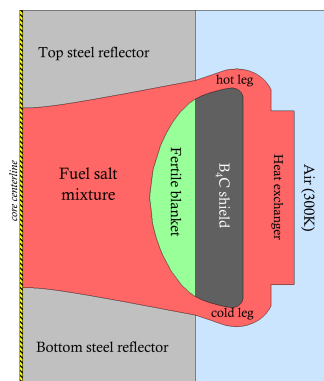| Element	^{6}Li	^{7}Li	^{19}F	^{232}Th	^{233}U
% mol	0.008%	28.964%	62.617%	7.421%	0.991%

Figure 1: MSFR axial-symmetric model developed in the framework of the EVOL Benchmark.

The functionalities are tested both independently and mixed one-another, referring in particular to Full Reprocessing Scheme (FRS) as a fuel treatment comprehensive of batch reprocessing (FTU), gas bubbling (GB) and Eutectic Control (EC). The new implementations have proved good capabilities to bring and maintain the system to a user defined reactivity condition limiting long term reactivity swings to less than 150 pcm (Figure 2a). Oscillations are unavoidably present due, on one hand, to the step nature of the burn-up calculation, and on the other to the approximation on to the leakage correction in the algorithm, resulting from both the stochastic nature of the calculation itself and the associated statistic error of the implicit k_{eff} calculated in Serpent. However, it is also worth to point out that, in reactor operation, small reactivity fluctuations around the criticality value translate in changes of the system average temperature, in the sense that positive variations of k_{eff} lower the criticality temperature and vice versa. The accuracy of the k_{eff} estimation performed by the reactivity control algorithm is also tested, proving a good agreement with the SERPENT calculated one as shown in Figure (2b), with a relative percentage difference between the two always bound to less than 0.15 %. Furthermore, the capability to control the eutectic composition is also verified, even in presence of reactivity control, proving a limited interference and a good interplay between the two implemented routines, as shown in Table 2.
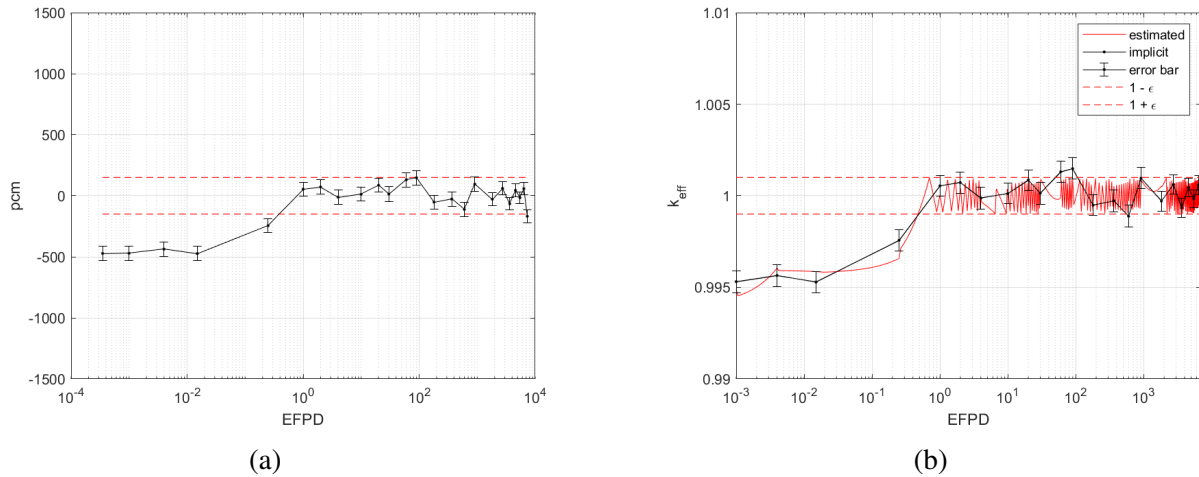
(a)

(b)

Figure 2: (a) Reactivity worth for twenty years of depletion under reactivity and eutectic control, in presence of FPs removal, with bandwidth of ± 150 pcm in red. (b) Comparison of the estimated (red) and implicit (black) multiplication factors.

Table 2: Molar fractions of light and heavy components over the mixture density, excluding the fluorine share. Final results are referred to twenty years of depletion.

Test Case	Light Fraction (%)	Heavy Fraction (%)
Initial	77.5	22.5
SD-RC + FRS	77.2	22.8
FRS only	77.1	22.9
Free Evolution	89.6	10.4

It is significant to stress the importance of the routines independence because it allows to separate the contribution of reactivity control from the reprocessing one and, most importantly from the eutectic control, which can be in general performed with different approaches with respect to the one proposed in the present work. The limited impact on the total system mass of the chosen reactivity control strategy is also proven both in presence and absence of eutectic control, although the very different cumulative amounts of required material to inject in the two cases reflects the importance of the FRS treatment, as presented in Table 3.

Table 3: Total inserted and extracted quantities after twenty years of depletion in case of RC only and RC coupled with FRS.

Switch	RC	RC+FRS
Total extracted ^{232}Th (kg/m^3)	-248.93	-50.95
Total injected ^{233}U (kg/m^3)	250.01	51.17

3 CONCLUSIONS

In this paper, a new strategy for reactivity control is presented as a SERPENT-2 code extension, featuring the capability to exploit the coupling of burn-up and material exchange processes to perform a tailored composition adjustment toward a target reactivity condition for Molten Salt Reactors. A new Semi-Diagonal control approach is devised and implemented in

order to perform a reactivity control intervention with a negligible interference with both the eutectic composition in term of heavy metal fraction and the system mass. Furthermore, the composition adjustment can also be exploited to control the evolution of relevant species in the fuel mixture. In particular, in the present work, the code is implemented with the additional capability to perform a continuous control of the eutectic proportions in the salt through an optimized approach, specifically developed for the MSFR reprocessing scheme, but in principle extendable to different alternative control approaches. The implementations are tested and verified, proving the capabilities of the new functionalities to bring and maintain the system to a user defined target reactivity, limiting long term reactivity swings with a minimal impact on both the total system mass and the eutectic proportions. This last characteristic in particular permits a mixable action with the implemented eutectic control strategy, with a good interplay of the two routines.

RESEARCH DATA

The data that support the findings of this study are openly available in Zenodo at http://doi.org/10.5281/zenodo.5172247

FUNDING

DISCLAIMER

The context of this paper does not reflect the official opinion of the European Union. Responsibility for the information and/or views expressed therein lies entirely with the authors.

REFERENCES

[1] Generation-IV International Forum, Technology Roadmap Update for Generation IV Nuclear Energy Systems, OECD-NEA, 2014.

[2] M. Aufiero, A. Cammi, C. Fiorina, J. Leppanen, L. Luzzi, M.E. Ricotti, "An extended version of the SERPENT-2 code to investigate fuel burn-up and core material evolution of the Molten Salt Fast Reactor", J. Nucl. Mater., 441, 2013, 473–486.

[3] S. Delpech, E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le-Brun, X. Doligez, G. Picard, "Reactor physic and reprocessing scheme for innovative molten salt reactor system", J. Fluor. Chem., 130, 2009, 11–17.

[4] A. Isotalo and P. Aarnio, "Higher order methods for burnup calculations with Bateman solutions", Ann. Nucl. Energy, 38, 2011, 1987–199.