

SIMULATION TOOLS AND NEW DEVELOPMENTS OF THE MOLTEN SALT FAST REACTOR

E. MERLE-LUCOTTE*, X. DOLIGEZ, D. HEUER, M. ALLIBERT, V. GHETTA
LPSC-IN2P3-CNRS / UJF / Grenoble INP
53 avenue des Martyrs, F-38026 Grenoble Cedex - France

ABSTRACT

Starting from the Molten Salt Breeder Reactor project of Oak-Ridge, we have performed parametric studies in terms of safety coefficients, reprocessing requirements and breeding capabilities. In the frame of this major re-evaluation of the molten salt reactor (MSR), we have developed a new concept called Molten Salt Fast Reactor or MSFR, based on the Thorium fuel cycle and a fast neutron spectrum. This concept has been selected for further studies by the MSR steering committee of the Generation IV International Forum in 2009.

Our reactor's studies of the MSFR concept rely on numerical simulations making use of the MCNP neutron transport code coupled with a code for materials evolution which resolves the Bateman's equations giving the population of each nucleus inside each part of the reactor at each moment. Because of MSR's fundamental characteristics compared to classical solid-fuelled reactors, the classical Bateman equations have to be modified by adding two terms representing the reprocessing capacities and the fertile or fissile alimentation. We have thus coupled neutronic and reprocessing simulation codes in a numerical tool used to calculate the extraction efficiencies of fission products, their location in the whole system (reactor and reprocessing unit) and radioprotection issues.

1. Introduction

The Generation-IV International Forum (GIF) for the development of new nuclear energy systems has established a set of goals as research directions for nuclear systems: enhanced safety and reliability, reduced waste generation, effective use of uranium or thorium ores, resistance to proliferation, improved economic competitiveness. Molten Salt Reactors (MSRs) are one of the systems retained in 2002 by this forum. Starting from the Molten Salt Breeder Reactor project of Oak-Ridge [1], we have performed parametric studies in terms of safety coefficients, reprocessing requirements and breeding capabilities. In the frame of this major re-evaluation of the molten salt reactor (MSR), we have developed an innovative concept based on the Thorium fuel cycle and called Molten Salt Fast Reactor or MSFR with no moderator in the core, leading to a fast neutron spectrum while ensuring excellent safety characteristics [2,3,4]. This reactor is associated to a chemical reprocessing unit located on-site. This MSFR configuration devised, developed and conducted by the Reactor Physics Group of LPSC in Grenoble has been officially selected by the Generation IV International Forum in 2009 for further studies. This concept of Molten Salt Fast Reactor together with its reprocessing unit will be presented in the next section.

Our reactor's studies of the MSFR concept rely on numerical simulations making use of the MCNP neutron transport code [5] coupled with a code for materials evolution which resolves the Bateman's equations giving the population of each nucleus inside each part of the reactor at each moment. Because of MSR's fundamental characteristics compared to classical solid-fuel reactors, the classical Bateman equations have to be modified by adding two terms representing the reprocessing capacities and the fertile and fissile alimentations. This code, whose calculation scheme will be described in section 3, includes recent developments in order to take into account the fast spectrum of the MSFR and the coupling of the reactor with a salt control and on-site reprocessing unit. Finally some preliminary results of this neutronics-reprocessing coupling will be presented in section 4 in terms of fission products extraction efficiencies and location in the system and radioprotection issues.

2. The concept of Molten Salt Fast Reactor

2.1 Reactor Geometry

The standard MSFR is a 3000 MWth reactor with a total fuel salt volume of 18 m³, operated between 650 and 850°C. As shown in Fig. 1, the core of the MSFR looks like a single cylinder (with diameter equal to the height) where the nuclear reactions occur within the flowing fuel salt. MSFR simulations have been performed using a binary fluoride salt, composed of LiF enriched in ⁷Li to 99.995 % and a heavy nuclei (HN) mixture initially composed of fertile thorium and either fissile, ²³³U or Pu. The (HN)F₄ proportion is set at 22.5 mole % (eutectic point), corresponding to a melting temperature of 565°C.

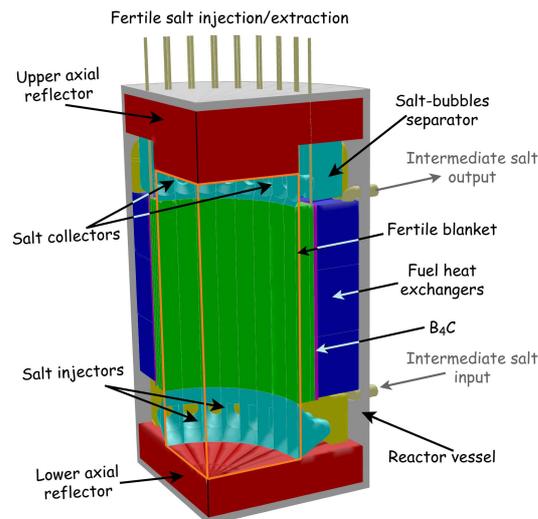


Fig. 1: Schematic view of a quarter of the MSFR. The fuel salt (not represented here) would be located within the orange lines

The external core structures and the fuel heat exchangers are protected by thick reflectors made of nickel-based alloys, which have been designed to absorb more than 80% of the escaping neutron flux. These reflectors are themselves surrounded by a 10cm thick layer of B₄C, which provides neutronic protection from the remaining neutrons. The radial reflector includes a fertile blanket (50 cm thick - green area in Fig. 1) to increase the breeding ratio. This blanket is filled with a fertile salt of LiF-ThF₄ with 22.5%- mole of ²³²Th initially.

2.2 Reprocessing Scheme

The on-site salt management of the MSFR combines a salt control unit, an online gaseous extraction system and an offline lanthanide extraction component by pyrochemistry [6]. This salt reprocessing scheme is presented in Fig. 2. The only continuous salt chemistry process is the gaseous extraction system. It consists first in injecting helium bubbles at the lower part of the core to trap the non-soluble fission products (noble metals) dispersed in the flowing liquid and also gaseous fission products. A liquid/gas phase separation is then performed on the salt flowing out of the core to extract gaseous species and dragged condensed particles.

Following this “physical” process of purification, a small part of the gas is withdrawn in order to let the fission products decay and the remaining part of gas is sent back to the lower part of the core. An experimental forced flow loop of fluoride salt* is under construction to evaluate the efficiency of this bubbling process in a fluoride salt.

The salt properties and composition are monitored through the online chemistry control and adjustment unit. A fraction of salt is periodically withdrawn and reprocessed offline in order to

* “FFFER project” in progress at LPSC Grenoble

with A_i the fertile or fissile alimentation of nucleus i . Our simulations consider several hundreds of nuclei (heavy nuclei, fission products, structural materials...) with their interactions and radioactive decays.

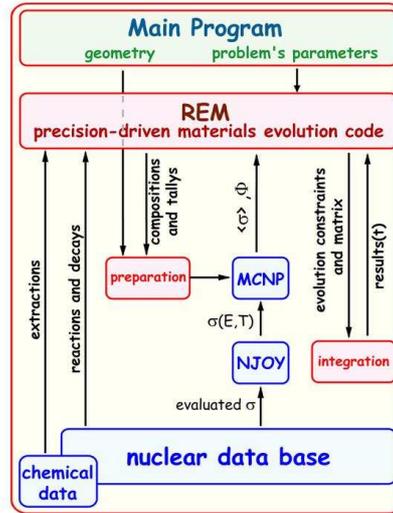


Fig.3. Coupling of the MCNP neutron transport code with the in-house materials evolution

The simulations of reactor evolution take into account the input parameters (power released, criticality level, chemistry...), by continuously adjusting the materials composition and thus the neutron flux of the system, via multiple interactions between the neutronic and the evolution tools. The REM code is indeed a precision-driven code, i.e. it has been designed to determine the reactor evolution while controlling the precision of the results at each step of this evolution. The resolution of the Bateman equations is constrained by several variables to keep the reactor's simulated physical parameters constant during the evolution. These include the total power (with a one percent or so precision) and the reactivity (with a huge precision of some ten pcm, much smaller than the computational uncertainty of this parameter under MCNP). The numerical integration of the Bateman equations is finally done using a Runge-Kutta method.

Simulation of the whole system: coupling of neutronics and reprocessing

The method developed in the previous paragraph calculates each nucleus population only inside the core. In order to calculate the nuclei populations inside the whole process, we partitioned the whole system (reactor, reprocessing unit...) into elementary sub-systems characterized by transfer functions from one sub-system to another. Just like the core extraction, those transfer functions have to characterize the kinetic of the considered operations and the thermodynamic equilibrium. There is thus at each step a competition between nuclear decays and chemical extraction. For instance, let's consider uranium just before the fluorination. There are three possibilities: nuclear decay, extraction by fluorine, or it could stay in the salt and thus could go to the next step which is the reductive extraction.

To couple the reprocessing and the core evolution, we add a dimension in equation 1. We have to add the location of each nucleus in the system as a new parameter. Consequently, the size of the matrix which was N (the number of nucleus) becomes $N \times x$, where x is the number of elementary operations done in the reprocessing unit. Equation 1 becomes equation 2 where ' B ' symbolizes the location of nucleus i in the sub-system B and ' $B \rightarrow C$ ' the transfer from sub-system B to sub-system C :

$$\frac{\partial N_i^B}{\partial t} = \sum_{j \neq i} \left\{ \left(\lambda_{j \rightarrow i} + X_j \langle \sigma_{j \rightarrow i} \phi \rangle \right) N_j^B \right\} + \sum_{C \neq B} \lambda_{Chem}^{C \rightarrow B} N_i^C - \lambda_i N_i^B - \langle \sigma_i \phi \rangle N_i^B - \sum_{C \neq B} \lambda_{Chem}^{B \rightarrow C} N_i^B$$

(Eq. 2)

We are thus able to calculate the evolution of matter in each process of the system and to know isotopes concentrations, gamma or neutron flux or the residual heat (fundamental data for radioprotection) everywhere as presented below in section 4.

The main issue in the reprocessing unit simulation is to determine the kinetic of each step of the process and consequently the transfer constants $\lambda_{chem}^{B \rightarrow C}$. As technological choices have not been fixed yet, only the available thermodynamic data have been used [8].

We assume that the reductive extraction will be done thanks to counterflow exchangers. In such exchanges, the reaction is limited by diffusion issues in the salt. For example, as described in [8], the chemical transfer constant corresponding to transition from the salt to the metal is given by equation 3:

$$\lambda_{salt \rightarrow metal} = \frac{k}{V_{salt} / S} \quad (\text{Eq. 3})$$

With S the exchange surface, k the transfer coefficient being equal to around 10^{-5} m.s^{-1} and V_{salt} the concerned salt volume.

4. Preliminary results of the neutronics-reprocessing coupling

Extraction efficiencies

The extraction efficiencies are evaluated from the ratio between the input flow of matter in the reprocessing unit and the output flow sent to storage. The difference between the input and the output corresponds to the matter reinjected in core. For the actinides (also called transuranian elements or TRU in the following), this ratio is rather viewed as reprocessing loss, while it really corresponds to an extraction efficiency for lanthanides.

Actinides		Lanthanides	
U	$<10^{-7}$	La	0.83
Pa	$1.5 \cdot 10^{-6}$	Ce	0.82
Np	$1.0 \cdot 10^{-7}$	Nd	0.91
Pu	$3.8 \cdot 10^{-5}$	Sm	0.90

Tab 1: Extraction efficiencies

The calculations have been performed with an extraction step for the actinides carried out through two cycles, while it requires around 20 cycles for the lanthanides. The fuel salt volume reprocessed is equal to 40 litres, corresponding to 200l of Bismuth and 400l of chloride.

The results obtained for the lanthanide extraction efficiencies and the actinide reprocessing losses are given in Tab. 1. The actinides losses during the reprocessing are lower compared to what is measured in current processes (PUREX...) due to the pyrochemical processes involved. However the results obtained are based only on thermodynamic considerations, neglecting possible technological limitations. They are thus optimistic.

Residual heat and neutron flux in the reprocessing unit

The coupling of the neutronic and reprocessing simulation tools allows the evaluation of the residual heat and the neutron flux present at each step of the reprocessing unit. The calculations have been performed with a 40 litres volume of fuel salt. The results are shown in Tab. 2.

The neutron flux is largely dominated by the spontaneous fissions of ^{244}Cm , ^{252}Cf , ^{246}Cm , ^{242}Cm , ^{250}Cf , ^{248}Pu , ^{248}Cm and ^{240}Pu . A total amount of 182 kW of residual heat has to be extracted from the reprocessing unit.

Location in the reprocessing unit	Residual heat (kW)	Neutron flux (neutrons emitted per second)
Extraction	81	$7.21 \cdot 10^7$
Fluorination (gaseous phase)	0.9	$1.70 \cdot 10^6$
Fuel salt after fluorination	80	$7.10 \cdot 10^7$
Bismuth bath (TRU extraction)	4	$6.46 \cdot 10^7$
Fuel salt after TRU extraction	9	$5.87 \cdot 10^5$
Bismuth bath (Ln extraction)	0.3	$5.34 \cdot 10^5$
Fuel salt after Ln extraction	5	$5.87 \cdot 10^5$
Chloride bath (hydrolysis)	2	$5.86 \cdot 10^2$
Total	182.2	$2.11 \cdot 10^8$

Tab 2: Residual heat and neutron flux in the reprocessing unit for 40 litres of fuel salt

5. Conclusions

Our simulations of the Molten Salt Fast Reactor concept rely on numerical tools making use of the MCNP neutron transport code coupled with a code for materials evolution which resolves the Bateman's equations which give the population of each nucleus inside each part of the reactor at each moment.

Because of MSR's fundamental characteristics compared to classical solid-fuel reactors, the classical Bateman equations have been modified by adding two terms representing the reprocessing capacities and an online alimentation. We have thus coupled neutronic and reprocessing simulation codes in a numerical tool used to calculate the extraction efficiencies of fission products, their location in the whole system (reactor and reprocessing unit) and radioprotection issues.

The very preliminary results presented in the paper, even if based on rough data of the pyrochemical processes involved, illustrate the potential of the neutronic-reprocessing coupling we have developed. We also show that these studies are limited by the uncertainties on the design and knowledge of the chemical reprocessing processes.

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