Nuclear fuel optimization for molten salt fast reactor

O Ashraf1,2,*, A D Smirnov1 and G V Tikhomirov1

1Institute of Nuclear Physics and Engineering, National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe shosse, 31, Moscow, Russian Federation, 115409
2Physics Department, Faculty of Education, Ain Shams University, Cairo, Egypt, 11341

*Corresponding author’s email: osama.ashraf@edu.asu.edu.eg

Abstract. In Molten Salt Fast Reactors (MSFR), a fuel as a liquid salt circulates through the cylinder core and transport the heat to the external heat exchangers, therefore liquid salt allows carrying the fuel and transfer heat. The MSFR predicted to work in a closed Th-based fuel cycle with a full reprocessing of all actinides in the core. The aim of this paper is; modeling the primary circuit of MSFR (European model), in order to optimize the concentration of the start-up liquid fuel using code Serpent 2. According to the results, the concentrations of the start-up liquid fuel required for criticality and long life cycle were 3 mol% of $^{233}\text{UF}_4$, 6 mol% of $\text{PuF}_3$ and 6.5 mol% of $\text{TRUF}_3$ respectively. The multiplication factors as a function of burnup have been calculated for different fissile materials. In conclusion, during 500 EFPD the MSFR was self-sustained regardless of the type of fissile materials used. The $\text{PuF}_3$ and $\text{TRUF}_3$ fuels appear to be prospective fuels compared to the $^{233}\text{UF}_4$ fuel.

1. Introduction

In MSFR the fuel is expected to dissolve in a fluoride-based molten salt, which plays also the role of the coolant [1-3]. Thermal power of MSFR is 3000 MW with a fast neutron spectrum, while the average power density is 48.6 MW/t-HM [4]. The thermal efficiency about 50%. The expected inlet temperature is 650 $^\circ$C, while the outlet is 750 $^\circ$C [1, 5]. In the MSFR, a liquid fuel salt circulates through the core and transfers the heat to 16 external heat exchangers, and then through the pumps it re-enters at the bottom of the core. From a modelling point of view the axial-symmetric representation of the core can be extended to the entire primary circuit by approximating the 16 external loops with a single annular loop [6, 7]. The core geometry can be assumed as a cylindrical vessel with diameter and height of 2.25 m made of a nickel-based alloy filled with the fuel. MSFR operate under ambient pressure. Figure. 1. Shows the schematic figure of the MSFR and the input model from Monte Carlo code ‘Serpent.2.1.29’ [8]. The fuel salt volume is 18 m$^3$ in total in the primary circuit, which proposed to be composed of: 77.5%LiF-22.5%ThF$_4$-(Fissile material) $F_\Phi$. The fissile material can be enriched uranium (r $\leq$ 20%), $^{233}\text{U}$ or plutonium and minor actinides. In between, a container is located filled with a blanket salt containing thorium to increase the breeding gain. The composition of blankets is LiF-ThF$_4$. Surrounding the blanket a 20 cm thick B$_2$C layer is planned to use, which can protect the heat exchanger from the neutrons. The density of the fuel salt is 4.1 g/cm$^3$ [5].

The aim of this work is; modeling the primary circuit of MSFR, European reference, in order to optimize the concentration of the startup liquid fuel required for criticality and long life cycle.
2. Methodology and tools

Current neutron-transport tools are designed to deal with the solid-fueled reactors, therefore the modelling of liquid-fueled systems has many challenges. Two main challenges are presented by liquid-fueled systems: (1) flowing of the liquid salt and (2) online reprocessing system [10, 11]. From a modeling point of view, the axial-symmetric representation of the core can be extended to the entire primary circuit by approximating the 16 external loops with a single annular loop [6]. In this work, two dimensional (2D) axial-symmetric representation of the MSFR has been modeled with neglecting the online fuel reprocessing. The present paper investigates the MSFR neutronics by determining the concentration of fissile material required for criticality and by the burnup calculations for 25 GWd/MTU. Serpent code has been adopted, following the need for an accurate determination of these quantities. Serpent is a three-dimensional continuous energy Monte Carlo neutron transport and burnup code. The results were obtained after full-core runs of 10 million active neutron histories per burnup step with the version 2.1.29 of the code. Simulations consisted in 500 active cycles of $2 \times 10^4$ neutrons. 20 inactive cycles were used for the convergence of the fission source distribution.

3. Startup core loadings

The fuel salt is composed of LiF for 77.5 mol%, and by a mixture of AcF$_3$ and AcF$_4$ for 22.5% (Ac indicates actinides). The fuel salt volume is 18 m$^3$ in total in the primary circuit. Three fuel compositions have been considered in this work; Th-U$^{233}$, Th-Pu, and Th-TRU (TRU is TRansUranic). In the first case, i.e. using Th-U$^{233}$, it would minimize the transition time to the equilibrium cycle. In the future, the U$^{233}$ can easily produce from MSFR (i.e. breeding reactors), but in order to overcome the unavailability of U$^{233}$ different fissile material can be used instead of U$^{233}$. For example, the second case, Th-Pu startup fuel appears to be more realistic because it would employ the current PUREX reprocessing technique and facilities to recover an already available fissile resource; the MSFR would then initially operate as a Pu burner [6]. Finally, The MSFR supposed to be used as a burner of the entire TRU vector produced by the LWR fleet, while initiating a new Th cycle [5]. In this case, Th-TRU with 5-year cooled TRU from LWR used fuel would be the pursued option [6].

The core is surrounded radially by a container filled with a blanket salt containing thorium to increase the breeding gain. The composition of blankets is LiF-ThF$_4$ (77.5 mol% LiF and 22.5 mol% ThF$_4$ for all cases considered). The NPP includes three different circuits involved in order to generate power: the fuel circuit, the intermediate circuit, and the power conversion circuit. These circuits are associated with other systems composing the whole NPP, such as online reprocessing units and the emergency draining system. It is worth noting that, for the determination of the accurate value of the multiplication factor, it is necessary to take into account the circulation of the fuel salt through the core, also the coupling between thermal-hydraulics and neutronic is needed [12-14].
4. Results and discussion

The infinite multiplication factor as a function of the concentration of fissile materials demonstrated in the figure 2. One can notice that in the case of $^{233}\text{UF}_4$ the concentration should be equal to 3 mol%, this concentration corresponding to $K_\infty=1.07132\pm0.00045$ (see figure 2). But in the case of PuF$_3$ as a fissile material, the concentration should be equal to 6 mol%, this concentration corresponding to $K_\infty=1.05407\pm0.00048$. Finally, for TRUF$_3$ the concentration should be equal to 6.5 mol%, this concentration corresponding to $K_\infty=1.02713\pm0.00043$. Although there is a good agreement between our results and results in [15] in the case of TRUF$_3$ and $^{233}\text{UF}_4$, the comparison for the PuF$_3$ case not done due to lack of data.

In burnup calculations, no poison was used. According to figure 3, the burnup calculation shows that; during the first 50 Effective Full power days (EFPD) the infinite multiplication factor drastically decreases (from $1.07219\pm0.00047$ to $1.05512\pm0.00048$), (from $1.05415\pm0.00044$ to $1.04412\pm0.00046$) and (from $1.50273\pm0.00048$ to $1.01712\pm0.00047$) in the cases of 3 mol% $^{233}\text{UF}_4$, 6 mol% PuF$_3$ and 6.5 mol% TRUF$_3$ respectively. This may be attributed to the following facts; firstly, about 50 EFPD is necessary for the concentration of the nuclear poisons to build up to an equilibrium value [4]. Secondly, it is known that the effect of $^{135}\text{Xe}$ and $^{149}\text{Sm}$ is more significant in the MSFR than in the other fast systems because the neutron spectrum is softer in the MSFR [4], Xenon will likely be removed during operation in these molten salt reactor systems, as having gas entrained within the fuel salt presents major issues. $^{135}\text{Xe}$ and $^{149}\text{Sm}$ have higher absorption cross-sections of lower neutron energy [16].

![Figure 2. The infinite multiplication factor as a function of the concentration of fissile materials.](image-url)
Finally, the multiplication factor strongly affected by the change of the $^{233}$Pa quantity (produced from Thorium). After 100 EFPD, in the case of $^{233}$UF$_4$, the value of $k_\infty$ shows continuous decreasing with a low rate and reach the value 1.02327±0.00044 by the end of the cycle (i.e. 500 EFPD). On the other hand, in the two remain cases, the value of $k_\infty$ tends to be constant with burnup time, which reflects the possibility of using them for long life cycles (see figure 3). At 500 EFPD the burnup is 25 GWd/MTU. The change in $k_\infty$ is about 4.6% (≈1.6% in the first 50 EFPD) for $^{233}$UF$_4$. For PuF$_4$ is about 1.3% (≈1% in the first 50 EFPD). For TRUF$_3$ is about 1.2% (≈1% in the first 50 EFPD). Based on the above, one can notice that during 500 EFPD the MSFR was self-sustained regardless of the type of fissile materials ($K_\infty$≥1 during whole cycle). PuF$_4$ and TRUF$_3$ fuels appear that they can be used for long life cycle compared to $^{233}$UF$_4$ fuel (see figure 3), this may be attributed to the concentration of the produced absorbers in the fuel. The optimum concentration of liquid salts has been listed in Table 1.

Table 1. The optimum concentration of liquid salts.

| Liquid | LiF  | ThF$_4$ | $^{233}$UF$_4$ | PuF$_3$ | TRUF$_3$ |
|--------|------|---------|---------------|---------|---------|
| 77.5 mol% | 19.5 mol% | 3 mol% | ----- | ----- |
| 77.5 mol% | 16.5 mol% | ----- | 6 mol% | ----- |
| 77.5 mol% | 16 mol% | ----- | ----- | 6.5 mol% |

5. Conclusion

A simplified model of a MSFR primary circuit has been elaborated. Calculations using Monte Carlo code Serpent 2.1.29 adopting the ENDF/B-VII library has been performed in order to determine the multiplication factor as a function of burnup. The optimum concentration of the start-up liquid fuel
required for criticality and long life cycle has been calculated and determined. According to the obtained results the concentrations were 3 mol% of $^{233}$UF$_4$, 6 mol% of PuF$_3$ and 6.5 mol% of TRUF$_3$ respectively. At 500 EFPD the burnup was 25 GWd/MTU. During 500 EFPD the MSFR was self-sustained regardless of the type of fissile materials used. The results showed that, the cycle length of the PuF$_3$ and TRUF$_3$ fuels are longer than the $^{233}$UF$_4$ fuel (see figure 3).

Conflict of interest
The authors declare no conflict of interest.

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