Preliminary study on the online reprocessing and refueling scheme for SD-TMS reactor

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Abstract. The Single-fluid Double-zone Thorium Molten Salt reactor (SD-TMSR) has a promising future. Inherent safety, liquid fuel, and possibility of online reprocessing and refueling are unique potentials of the SD-TMSR. In the present study, the Monte Carlo code Serpent2 has been chosen to model the full core of the SD-TMSR. In addition, online reprocessing and refueling has been modeled based on subroutine provided by Serpent2. During the burnup time (10 years), the total mass of the fuel inside the core has been checked and found to be almost constant. Furthermore, we controlled the reactivity by adjusting the feed rate of fissile and/or fertile materials. The change of K_{eff}, breeding ratio, Th and U-233 refill rates with burn-up time have been investigated. Additionally, the build-up of U-233 during 10 years has been calculated.

1. Introduction

The Molten Salt Reactor (MSR) concept was first introduced in the late 1940s to develop a nuclear-powered airplane [1]. Liquid-fueled MSR systems have promising characteristics, for instance, inherent safety, fuel cycle benefits, and economic depending on the reactor design. Therefore, MSR was selected by the Generation IV International Forum (GIF) for further studies and development [2]. Both thermal-neutron-spectrum and fast-neutron-spectrum MSR have several potentialities compared with solid-fuelled reactors [3], [4]. In the MSR, liquid fuel salt, for example, BeF_2-LiF-(Th/U)F_4 circulates in a loop-type primary circuit and allows transferring fission heat. The liquid fuel allows for online reprocessing and refueling. Therefore, MSRs can potentially operate for a long time with superior neutron economy without shutdown for refueling [4]. MSRs can operate either as a breeder reactor or as a burner reactor incinerating nuclear waste. Breeding fissile materials from fertile materials based on Th-U fuel cycle can be realized in both thermal- and fast spectrum [5]. In moderated MSR, the initial amount of fissile materials is relatively low and doubling time for production ^{233}U is short compared with unmoderated MSR.

In 2011, the Chinese Academy of Sciences (CAS) established the project "Future Advanced Nuclear Energy–Thorium-based Molten Salt Reactor System (TMSR)". The main aim of this project is to fix the technical issues of TMSR [6]. The Single-fluid Double-zone Thorium-based Molten Salt Reactor (SD-TMSR-2,250 MW$_{th}$) was introduced by the CAS [5].

In the single-fluid reactor, the fissile and fertile materials (in the present work, ^{232}Th and ^{233}U respectively) are integrated into the same salt. In contrast, the fissile and fertile materials are separated in the two-fluid reactor. Dividing the active core into two or more zones would have to enhance the breeding ratio [7]. In the Double-zone TMSR, the outer zone consists of fuel channels with larger radius relative to the fuel channels in the inner zone. This leads to an increase in the breeding ratio [5], [7].

Existing neutron transport and depletion software were designed especially for simulating solid-fueled reactors. Therefore, the lack of such codes retards progress. Liquid-fueled MSR designs depend on online reprocessing of the fuel. From a modeling point of view, there is two approaches to the online reprocessing and refueling; continuous and batch-wise approach. In the batch-wise approach, the simulation stops at a certain time and restarts after removing of poisoning isotopes and addition of fissile and/or fertile materials [8]. In contrast, the continuous approach allows the removing and addition of materials without stopping the simulation (simultaneously with burn-up).

The continuous online reprocessing requires modification of the Bateman equations. The extension of the Serpent2 code [9] takes into account the online reprocessing of the fuel and its effects on depletion calculations [10]. The Serpent2 extension directly accounts for the effects of online fuel reprocessing on the depletion calculations and features a reactivity control algorithm. The developed extended version of Serpent2 was estimated against a dedicated version of the deterministic ERANOS-based EQL3D procedure in [11] and [10] and has been adopted to analyze the MSFR fuel salt isotopic evolution.

The present work aims to utilize the Serpent2 to simulate the full-core model of the SD-TMSR with considering the online reprocessing and refueling during 10 years of reactor operation time.

In this work, we used Serpent2 version 2.1.30 to obtain the results. The Serpent2 provided OpenMP and MPI parallelized memory management, which helped to perform burn-up calculations on computer clusters with many cores. This paper follows the methodology described in [10] and applies to the SD-TMSR 2,250 MW$_{th}$.

This paper is organized as follows; a brief introduction about the MSR systems is introduced in section 1. Section 2 includes a description of the SD-TMSR. Section 3 illustrates the tools and methodology implemented in this work. Section 4 demonstrates the results and discussion. Finally, the conclusion is listed in section 5.

2. Model description

The Single-fluid Double-zone Thorium-based Molten Salt Reactor (SD-TMSR-2,250 MW$_{th}$) was introduced by the Chinese Academy of Sciences (CAS) [5], [6]. The quarter-core model configuration is illustrated in figure 1.

The active core of the SD-TMSR is a cylinder filled with hexagonal graphite prisms (density of graphite is 2.3 g/cm^3). The fuel channels passed vertically through the hexagonal graphite prisms. The active core is divided into a double zone to improve the breeding ratio. The inner zone contents 486 fuel tubes with a radius of 3.5 cm and the outer zone content 522 fuel tubes with a radius of 5 cm. The active core is surrounded by graphite reflectors to decrease the leakage of neutrons. The thickness of the graphite radial reflector is 50 cm and the height of the axial reflector equal to 130 cm.

The fluoride and chloride salts are anticipated to act as a fuel carrier and coolant in the MSRs. In the graphite-moderated TMSR, the lithium and fluoride salts show promising characteristics thanks to the relatively low corrosion and more transparency to neutrons [7].

In the present study, the fuel salt (with a density of 3.3 g/cm^3) is consist of 70LiF, 17.5BeF$_2$ and 12.5(HM)F$_4$ in mol% (HM is Heavy Metal (i.e. ^{232}Th and ^{233}U)). The ratio of fissile material to fertile material is adjusted for the criticality. The total fuel volume is 52.9 m^3 and the operating temperature is 900 K under ambient pressure.

The core is surrounded by a 10-cm-thick B$_4$C cylinder for radiation protection purposes. Furthermore, a 10-cm-thick cylinder from Ni-based alloy (H-alloy) surrounds the full core as a

structural material. The optimum radius of the fuel channels, as well as the side length of the hexagonal prism, were determined in [5].
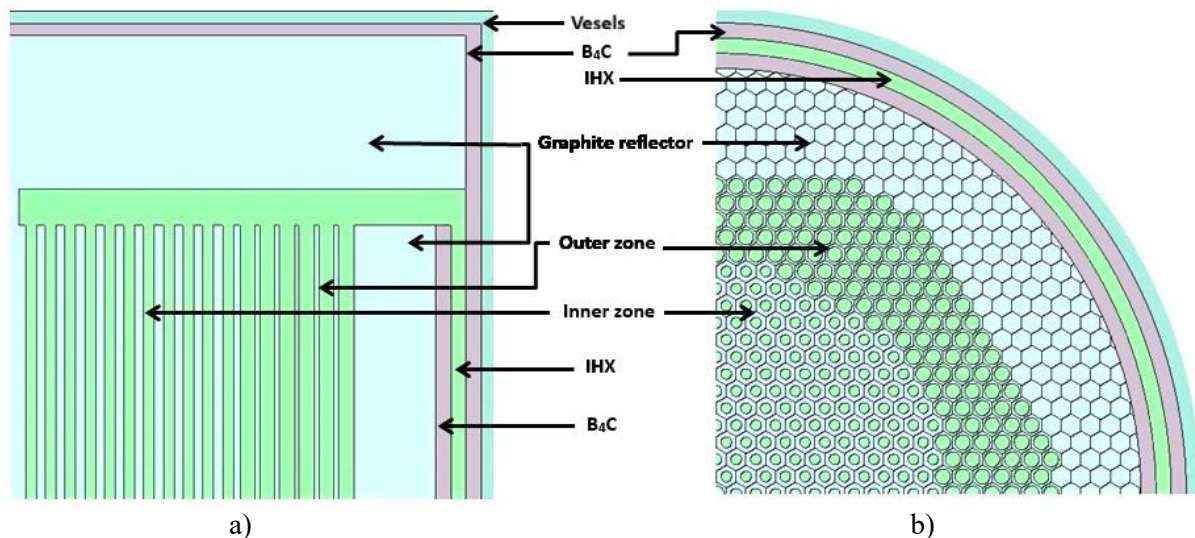

Figure 1: The quarter-core model configuration of the SD-TMSR a) Longitudinal view b) cross-sectional view at the horizontal midplane.

3. Methodology and tools

The online extraction of noble gases and Fission Products (FPs) provides many advantages for liquid nuclear systems, for example, in MSRs, it improves the breeding performance and reduces the fissile inventory required to maintain criticality [12]. The criticality can be achieved during burn-up by the online feed of ^{233}U and/or ^{232}Th [3].

The Serpent2 [9] is a three-dimensional continuous energy Monte Carlo neutron transport and burn-up code. Furthermore, Serpent2 has been extended to model the online reprocessing and refueling in MSRs [10].

In the present work, Serpent2 has been utilized for the simulation of the full-core model of SD-TMSR with online reprocessing and refueling. The results were obtained after whole-core runs of 5 million neutron history and 50 inactive cycles per burn-up step for the convergence of the fission source distribution. The reactor operation time was 10 years (84.56 MWd/kgHM) with statistical error in multiplication factor equal to ±56 pcm. During the burn-up, the gaseous, soluble and non-soluble FPs are removed in a convenient time. In the present work, the extraction time for gaseous and non-soluble FPs is 30 seconds and for lanthanides and other soluble metals is ~10.6 days [7]. In order to achieve that, we specified the flow rate of gaseous FPs and other materials from the core to the gas-tank in the input file. In particularly, Pa is removed from the core with a specific flow rate into the Pa-tank to decay into ^{233}U with $T_{1/2} \approx 27$ days.

To maintain the reactor critical and the fluctuation of the total fuel mass in the normal range, a built-in subroutine provided by the Serpent2 has been adopted. The inherent capability provides changing the transfer rates of ^{233}U and ^{232}Th throughout the burn-up [10]. The amount of fissile and fertile materials required to reach the criticality is calculated at the end of the cycle (i.e. 30 days). After that, this amount is added to the core from the beginning of the cycle by a specific flow rate.

The removal constant λ_e [s^{-1}] (the rate at which the material is removed) of gaseous and other fission products, as well as feed materials, were precisely calculated and listed in table 1.

Table 1. The reprocessing scheme.

Reprocessing group	Elements	Reprocessing time	Removal/Feed constant λ_e [s^{-1}]
FPs and non-dissolved metals	Z = 1, 2, 7, 8,10, 18, 36, 41, 42, 43, 44, 45, 46, 47, 51, 52, 54, 71, 72, 73, 74, 75, 76,77, 78, 79 and 86.	30 seconds	-3.33E-02
Chemical removal of lanthanides and other soluble FPs	Z = 30, 31,32, 33, 34, 35, 37, 38, 39, 40, 48, 49, 50, 53, 55, 56, 57, 58, 59, 60, 61,62, 63, 64, 65, 66, 67, 68, 69, 70.	10.5994 day (rate=5 m^3/d)	-1.09E-06
Pa is also extracted by the chemical reprocessing	Pa	10.5994 days (rate=5 m^3/d)	-1.09E-06
Feeds	^{232}Th	2.0875 [Kg/day], form 0 to 90 day	1.70E-09
		2.4559 [Kg/day], form 90 to 1080 day	2.00E-09
		2.1489 [Kg/day], form 1080 to 1440 day	1.75E-09
		2.4559 [Kg/day], form 1440 to 3650 day	2.00E-09
	^{233}U	2.7834 [Kg/day], form 0 to 90 day	6.80E-09
		2.0466 [Kg/day], form 90 to 1440 day	5.00E-09
		1.9443 [Kg/day], from 1440 to 1980 day	4.75E-09
		1.8828 [Kg/day], from 1980 to 3650 day	4.60E-09

4. Results and discussion

4.1. Steady-state calculation

In order to achieve the criticality, the concentration of the heavy metal in the initial fuel salt is set to 12.5 mol%. Then, the ratio between ^{232}Th and ^{233}U is adjusted until we reach the criticality. This leads to an initial inventory of ^{233}U and ^{232}Th equal to 1.3 and 76.87 tons respectively. The Breeding Ratio (BR) at the start-up (the ratio between the total ^{232}Th capture rate and total ^{233}U absorption rate) was calculated and found to be equal 1.11598±0.00096. These findings are in good agreement with [5]

4.2. Neutronic parameters

The chemical reprocessing rate of lanthanides and other soluble FPs (see table 1) is set to 5 m^3/d and the operation time is 10 years (122-time step). Figure 2 demonstrates the effective multiplication at every time step. The K_{eff} is decreased by only 110 pcm at the end of the 10-years operation time. Figure 2 shows that the K_{eff} fluctuates in a very narrow region (less than ±1063 pcm) thanks to the

online reprocessing and refueling technique (removing neutron poisons and addition of fissile materials). This illustrates the possibility of controlling the TMSR by adjusting the refill rate of fissile and/or fertile materials.
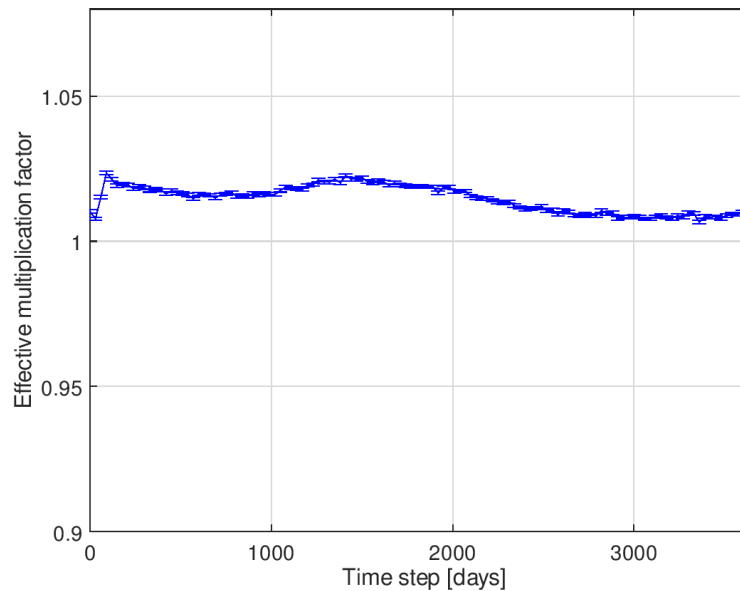

Figure 2: Variation of the effective multiplication factor with time step (burn-up) statistical error ±56 pcm.

The change of ^{233}U and ^{232}Th refill rates with time is presented in figure 3. As shown in figure 3, the maximum ^{233}U refill rate is 2.78 kg/d during the first 90 d, as well the mean value is 1.73 kg/d throughout 10 years of operation. Meanwhile, the maximum ^{232}Th refill rate is 2.45 kg/d during the last 6 years, as well the mean value is 2.28 kg/d throughout 10 years of operation. These findings are in good agreement with [8].
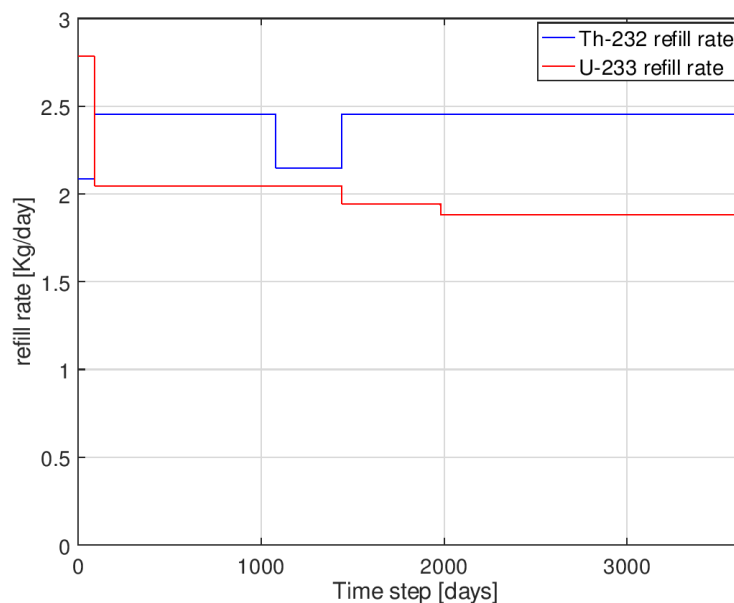

Figure 3: The change of ^{233}U and ^{232}Th refill rates with time steps (burn-up).

The fluctuation of the fuel mass throughout the 10-years of operation is demonstrated in figure 4. It is worth noting that the variation of the total fuel mass in both cases (5 m^3/d chemical reprocessing and without reprocessing) is less than 0.1% this result in good agreement with [8]. In order to restrict the variation of the total fuel mass throughout the burn-up time, the refill rate of ^{232}Th is adjusted as listed in Table 1.
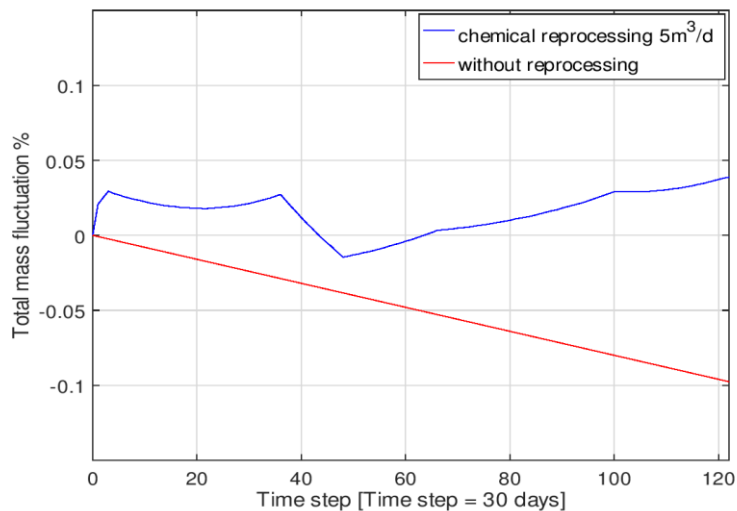

Figure 4: The change of the total fuel mass throughout the 10-years of reactor operation time.

The BR as a function of the time step is represented in figure 5. As shown in figure 5, the BR fluctuates around 1.1 (±96 pcm) during the 10-years of operation time. This would reflect the ability of the SD-TMSR to breed more fissile materials.

After 10-year of the reactor operation, the total mass of the elements in the Pa-tank and gas-tank was 7.75 t and 8.5 t respectively. The total mass of ^{232}Th and ^{233}U is compensating the extracted elements. Thus, the fuel mass was almost constant during the 10-years of reactor operation (see figure 4).
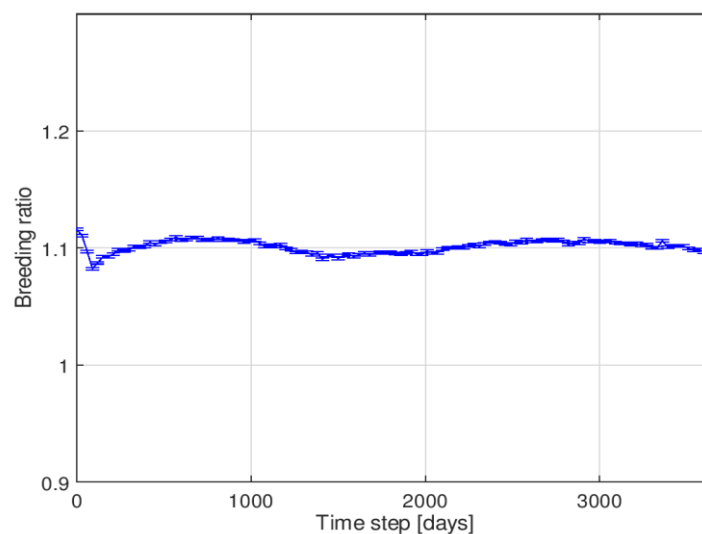

Figure 5: The breeding ratio (BR) as a function of the time step (burn-up).

The net production of ^{233}U throughout the considered operational time (10-years) is illustrated in figure 6. The net production of ^{233}U increases with depletion time and reached 654 kg at the end of 10 years. As shown in figure 6, the 10-years of reactor operation not enough to produce a sufficient amount of ^{233}U to trigger another TMSR (i.e. 1.3 t). Meanwhile, one can see that the net production of ^{233}U during the first 90 days is decreased, therefore about 129 kg of ^{233}U must be added in the core during this period. This may be attributed to the fact that the Pa-tank does not contain any ^{233}U at the start-up of the reactor operation.
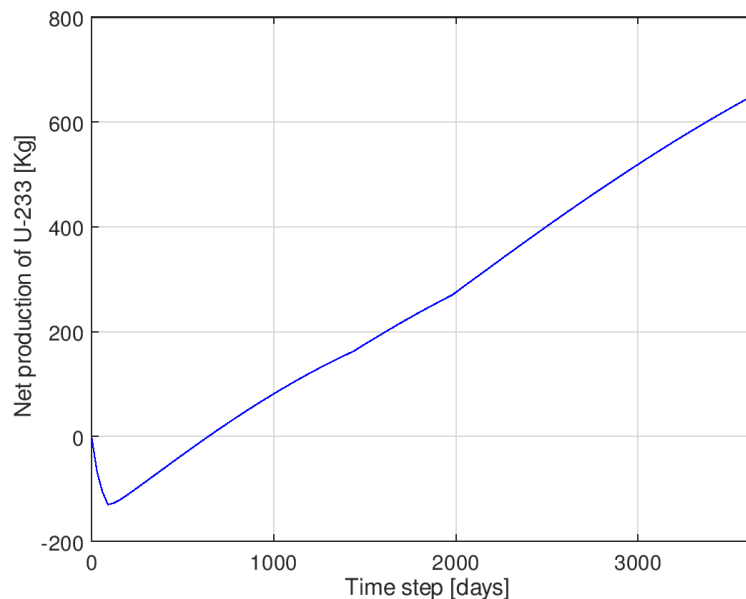

Figure 6: Net production of ^{233}U as a function of time step (burn-up).

5. Conclusion

In the present work, Serpent2 has been adopted to simulate the SD-TMSR with online reprocessing and refueling. The results were obtained after whole-core runs of 5 million neutron history and 50 inactive cycles per burn-up step. The reactor operation time was 10 years (84.56 MWd/kgHM) with a statistical uncertainty of ±56 pcm in K_{eff}.

In the steady-state calculation, the amount of fissile material is adjusted in the start-up molten salt fuel. Hence, the initial inventory of ^{233}U and ^{232}Th is 1.3 and 76.87 tons respectively, this leads to K_{eff} equal 1.01025±56 pcm.

The Breeding Ratio (BR) at the start-up was calculated and found to be equal 1.11598±0.00096. Moreover, the variation of BR with burn-up time emphasizes the possibility of operating the TMSR as a breeder reactor for a long lifetime. Under 5 m^3/d chemical reprocessing rate, the K_{eff} fluctuates in a very narrow region (less than ±1063 pcm).

During normal operation, the reactor is maintained critical by regulating the feed rate of ^{233}U and ^{232}Th as listed in Table 1. Meanwhile, the change in the total fuel mass is less than 0.1%.
The net production of ^{233}U increases with depletion time and reached 654 kg at the end of 10 years operating time. As shown in figure 6, the 10-years of reactor operation not enough to produce a sufficient amount of ^{233}U to trigger another TMSR.

Future work

The authors intend to extend the study, for example, increase the burn-up time to 60 years, investigate the build-up of important isotopes affecting the core of the reactor and the temperature coefficient of reactivity. In addition, the authors intend to investigate the neutron spectrum and flux.

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