Optimization of Thorium Utilization in Fluoride Salt-Cooled High-Temperature Reactor (FHR)

Xiaoxiao Li, Jingen Chen and Yonghan Fang

Abstract Preliminary neutronics analysis of thorium utilization in Fluoride salt-cooled High-temperature Reactor (FHR) has been carried out using a similar reactor model and fuel composition as the one researched in UC Berkeley. Compared with uranium-based fuels, thorium-based fuel (i.e., Th2/U3) shows a deeper burnup but a lower conversion ratio (CR) and a smaller temperature coefficient of reactivity (TCR). To improve the TCR, the weight ratio of the fissile material (U-233) and the fertile nuclide (Th-232) and the geometry of the TRISO and/or pebble are investigated. The higher the proportion of Th-232 in the Th2/U3 fuel, the higher the CR and TCR can be obtained. On the other hand, a harder spectrum is also contributive to the improvement of the TCR through the geometry optimization of TRISO and/or pebble. These two optimization approaches can be conducted to balance the burnup, TCR, and CR to realize efficient and safe utilization of thorium in FHR.

Keywords Thorium · FHR · C/HM · TCR · Burnup

1 Introduction

Thorium–uranium fuel cycle possesses obvious advantages [1], involving better breeding capacity particularly in thermal/epithermal spectra and lower radiotoxicity of nuclear waste within 10,000 years after shutdown. In addition, to deliver on potential performance of thorium, especially its high-temperature stability, thorium was tested and taken as nuclear fuel in High-Temperature gas-cooled Reactors (HTRs) [2–4]. The Fluoride salt-cooled High-temperature Reactor (FHR) [5, 6] is essentially the same as a HTR, which contains TRISO particles moderated by graphite but is cooled by molten salt instead of helium gas. Most of the researches

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on FHRs have been concentrated in uranium-based fuel [7, 8] while that in thorium-based fuel is only in the beginning stage [9–11].

The neutronic characteristics of thorium utilization were preliminary analyzed with a whole-core model of pebble bed FHR in our previous work [9]. By comparing the thorium-based fuel to the uranium one, it can be conducted that the ^{232}Th/^{233}U fuel system (Th2/U3 for short) has the highest initial k_{eff} and deepest burnup due to the smaller resonance absorption of Th-232 and the higher effective number of fission neutrons of U-233 especially in thermal and epithermal spectrum region. Since the reactor model and the fuel composition used in our previous work [9] are similar as the one researched in UC Berkeley based on uranium fuel [8], there are two drawbacks for Th2/U3 in such FHR, i.e., low conversion ratio (CR) and small temperature coefficient of reactivity (TCR). According to the analyses in our previous work [9], approaches to optimize CR and TCR can be conducted by adjusting such as the weight ratio of fissile material and fertile nuclide (i.e., the U-233 weight fraction (w%) for Th2/U3, defined as U-233 w%) and neutron spectrum. The neutron spectrum corresponding to the ratio of moderator to heavy metal (C/HM) can be adjusted in two ways, i.e., modifying the number of TRISO coated particles and changing the fuel kernel radius. The former way using a fuel kernel radius of 0.25 mm offers an opportunity to alter C/HM without making changes to the TRISO itself.

The description of the FHR core, fuel pebble, TRISO-coated fuel particle, fuel composition, and nuclear analytical tool are presented in Sect. 2. Optimizations of the U-233 weight fraction and the TRISO geometry to improve CR and TCR are given in Sect. 3. Conclusions are summarized in Sect. 4.

2 Methodology

2.1 Geometric Properties

The present study on the use of Th2/U3 in FHR is based on a whole-core design in reference [9]. Both the diameter and the height of the active zone have the same value of 5.06 m. The active zone is filled with fuel pebbles in a regular hexagonal lattices with a packing factor of 60 % while the remaining 40 % filled with molten salt, and surrounded by a graphite reflector with thickness of 0.5–1 m [12]. The molten salt area filled with FLiBe (66 mol% ^{7}LiF–34 mol% BeF_2) is located below the active zone. The density of FLiBe is 1.90 g/cm^3 [8] (assumed operation temperature of 1050 K), and the abundance of Li-7 in FLiBe is 99.995 %.

Each pebble is composed of a fuel zone with radius of 2.5 cm and a fuel-free graphite shell with thickness of 0.5 cm. The former part is formed by a graphite matrix with density of 1.6 g/cm^3 in which several thousands of TRISO-coated fuel particles are embedded and uniformly distributed within a hypothetical six prismatic crystal lattice. The latter part is made of pure graphite with density of 1.6 g/cm^3.

Table 1 Description of fuel pebble and TRISO coatings

Item	Size	Material
Fuel pebble		
Fuel zone radius	2.5 cm	Graphite matrix
Fuel-free zone thickness	0.5 cm	Graphite
Pebble packing factor	60 %	
TRISO coatings		
Fuel kernel radius	0.25 mm	UO_2 + ThO_2 (variable: U-233 w%)
Porous carbon buffer layer thickness	95 μm	Graphite
Inner pyrolytic carbon layer thickness	40 μm	Graphite
Silicon carbide layer thickness	35 μm	SiC
Outer pyrolytic carbon layer thickness	40 μm	Graphite
TRISO packing factor	Variable	

Each TRISO-coated fuel particle consists of a fuel kernel surrounded by four coating layers [13, 14], i.e., a Porous Carbon Buffer layer, an Inner Pyrolytic Carbon layer, a silicon carbide layer, and an Outer Pyrolytic Carbon layer. The fuel kernel contains a mixture of UO_2 and ThO_2, and U-233 is the only isotope in the uranium of UO_2. The thickness of the TRISO coating layers has a linear relation with the fuel kernel radius to ensure the retention of all the fission products and make very little influence upon neutronics [15].

The geometrical parameters used for the fuel pebble and the TRISO-coated fuel particles are summarized in Table 1. The purpose of this work is to improve CR and TCR by altering neutron spectrum through changing C/HM without modifying the core geometry. Therefore, possible variations in the fuel design are fuel kernel radius, number of fuel particles corresponding to C/HM, graphite matrix diameter (the fuel zone of pebble). For simplicity, only the second case is chosen in this work without making changes to TRISO and pebble themselves. Furthermore, U-233 w % in fuel kernel is adjusted to optimize CR and TCR through the competition between the fission ability of U-233 and the capture ability of Th-232.

2.2 Neutronics Simulation Tool

The calculation including criticality and burnup for a whole FHR core is performed using the SCALE code package [16] which is an inclusive modeling and simulating tool applicable for nuclear criticality and safety analysis. Considering the FHR-specific phenomenon, a DOUBLEHET module is used to treat the resonance self-shielding effect and the double heterogeneous nature [17, 18], which is capable of handling all the layers of the TRISO-coated fuel particles and the whole pebble at once [12]. Time-dependent cross-section processing reveals the fuel composition variation during burnup based on the ENDF/B-VII library with 238-group. Moreover, the burnup calculation is carried out under a constant power of 1000 MWth and a large enough number of nuclides (388 nuclides) are monitored in trace quantities. Considering the calculation accuracy, the burnup steps with a smaller time are required particularly at the original irradiation to achieve an equilibrium concentration of much of fission products. Furthermore, raising the order of burnup steps as the increasing depletion is also needed to save computation time [19]. For each burnup step, 50 cycles are skipped and a total of 200 cycles with 2000 neutrons per cycle are applied. The average computation time of one burnup step is around 15 min with a single-core computer.

3 Results and Discussions

Three-dimensional Monte Carlo neutronics calculations are performed to study the characteristics of the FHR core with Th2/U3. The influences of the U-233 weight fraction (U-233 w%) and C/HM upon TCR and burnup are investigated for six different U-233 weight fractions, between 7.5 and 20 %, and for four different C/HM ranging from 100 to 400.

3.1 The Influence of U-233 w% and C/HM upon Spectrum

Spectrum factor (unit: eV) is defined as the energy of average lethargy of fission, which can be used to characterize the neutron energy spectrum. The larger the spectrum factor, the harder the spectrum becomes. Figure 1 gives the variation of the spectrum factor with the U-233 weight fraction for different C/HM. Under the same U-233 weight fraction, the spectrum factor becomes larger with the decreasing C/HM, and it is clear that the U-233 weight fraction is of great importance for the spectrum factor especially for a lower C/HM. To find out the reason for this trend, the difference of the neutron fractions between the highest C/HM (i.e., 400) and the lowest C/HM (i.e., 100), defined as Δ, is presented in Fig. 2. The three values (Δ1, Δ2, and Δ3) correspond to thermal, intermediate, and fast neutron regions, respectively, and the positive (negative) value represents

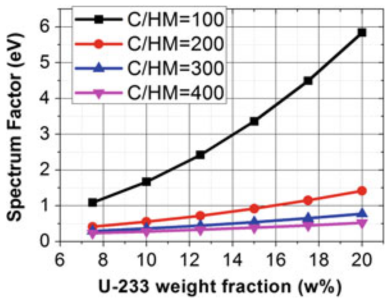

Fig. 1 Spectrum factor for the FHR core as a function of U-233 weight fraction for different C/HM

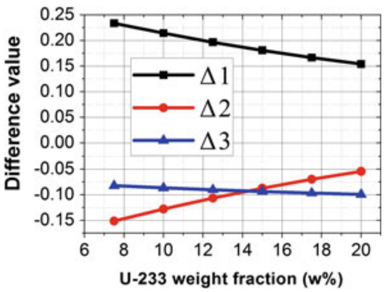

Fig. 2 Difference of neutron fraction between highest C/HM (i.e., 400) and lowest one (i.e., 100) versus U-233 w%

raising (reducing). It can be seen that the differences of the thermal and intermediate neutron ranges are more evident while that of the fast one is less significant along with the increase of the U-233 weight fraction. That is to say, the evolution of the spectrum factor as a function of the U-233 weight fraction mainly depends on the competition of Δ1 and Δ2. For the higher U-233 weight fraction, the higher the difference between the absolute value of Δ1 and Δ2, the more significant change of the spectrum factor is.

Besides, with the same C/HM, the spectrum factor tends to be larger with the increase of the U-233 weight fraction, and the U-233 weight fraction is more significant to the spectrum factor for a higher U-233 weight fraction. The reason for such trends is same as described above.

3.2 The Influence of U-233 w% and C/HM upon k_{eff}

The initial k_{eff} and the conversion ratio (CR) from Th-232 to U-233 are two competitive factors during burnup, which determine the depth of burnup. A deeper burnup can be obtained with a higher initial k_{eff} and a larger CR. The k_{eff} is plotted as a function of C/HM for different U-233 weight fractions in Fig. 3. For all weight fractions of U-233, the increase of C/HM from 100 to 400 initially leads to an increase of k_{eff} when the fuel pebbles is for the under-moderated condition and then to a decrease as the fuel pebbles become over-moderated due to a larger C/HM.

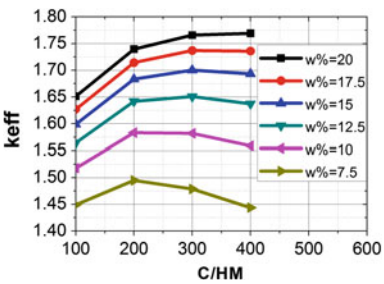

Fig. 3 k_{eff} for the FHR core as a function of C/HM for different U-233 weight fractions

The value of C/HM at which the transition from under-moderated to over-moderated occurs also increases as the U-233 weight fraction rises.

It is worth mentioning that the value of C/HM with 200, denoted as C/HM200, is an interesting point in Fig. 3. C/HM200 almost corresponds to the transition point for the lower U-233 weight fraction (7.5 %). In addition, the evolutions of k_{eff} with C/HM for the higher U-233 weight fractions (≥ 10 %) are almost parallel before C/HM200 and show different evolution trends after this point. It can be analyzed from the perspective of six-factor formula, but it will not be discussed here. Furthermore, C/HM200 is also a dividing point in the trend of TCR with the increase of the U-233 weight fraction, which will be discussed in detail in Sect. 3.3.

3.3 The Influence of U-233 w% and C/HM upon TCR

The TCR has been determined for the Th2/U3 fueled pebbles in a whole FHR core. Therefore, the separate k_{eff} value can be obtained by changing the temperature of the pebbles, the moderator, the coolant, and the reflector, respectively. A negative TCR is necessary for safe operation of a reactor. The total TCR with negative values for the whole FHR core is plotted as function of the U-233 weight fraction for different C/HM in Fig. 4. It can be seen that a wider choice of the U-233 weight fraction is available for a lower C/HM to obtain a negative TCR.

Under the same U-233 weight fraction, a lower C/HM leads to a stronger negative TCR which is chiefly determined by the fuel TCR (FTCR) and coolant TCR (CTCR). In all cases, FTCR is negative and reduces as C/HM rises. Likewise, the same law can be found in the trend of the coolant coefficient of reactivity (CTCR). FTCR and CTCR for two different U-233 weight fractions (10 % and 20 %) and for different C/HM (100, 200, 300, and 400) are given in Table 2. Moreover, FTCR decreases and CTCR increases with the increase of the U-233 weight fraction. TCR decreases with the increasing U-233 w% for lower C/HM (i.e., 100) due to the greater reduction of FTCR than the smaller enhancement of CTCR. Instead, TCR increases with the increase of U-233 w% for higher C/HM (i.e., ≥ 200) due to the inverted variation of FTCR and CTCR.

Fig. 4 TCR for FHR core as a function of the U-233 weight fraction for different C/HM

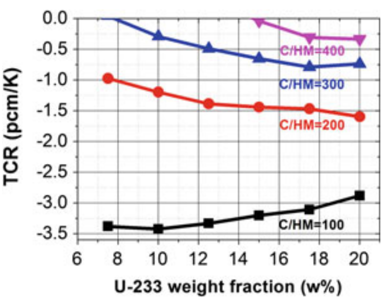

Table 2 FTCR and CTCR for the FHR core

C/HM	100	200	300	400
U-233 w% = 10				
FTCR	−3.22	−1.58	−0.97	−0.8
CTCR	−1.03	0.3	0.52	1.27
U-233 w% = 20				
FTCR	−2.26	−1.47	−0.97	−0.97
CTCR	−1.2	−0.23	−0.15	0.17

3.4 The Influence of U-233 w% and C/HM upon Burnup

The burnup calculations are performed for four different C/HM by varying the number of TRISO coated particles, and for six different U-233 weight fractions, from 7.5 to 20.0 % and presented in Fig. 5. It is found that the burnup deepens with the increasing U-233 weight fraction and the decreasing C/HM. For all C/HM, the burnup increases gradually and obtains a saturated value at the point with the U-233 weight fraction of about 12.5 %.

Two dimensions are used to explain the trends in Fig. 5. First, Fig. 6 shows the evolution of k_{eff} and CR for the FHR core with the U-233 w% of 12.5 for four different C/HM. It can be seen that the FHR core with a lower C/HM possesses a higher CR during depletion and thus the k_{eff} decreases more slowly with burnup and then a deeper burnup can be obtained even with a smaller initial k_{eff}. Second, Fig. 7 gives the evolution of k_{eff} and CR for the FHR core with C/HM of 200 for four different U-233 weight fractions. It is evident that the FHR core with a higher U-233 weight fraction has a lower CR and then the k_{eff} displays a rapider decline with burnup but a deeper burnup is achieved due to a much larger initial k_{eff}. In short, a deeper burnup can be realized through a higher CR with a small difference of initial k_{eff} (see Fig. 6), but it does not work for a large difference of initial k_{eff} (see Fig. 7).

Based on the above discussions, a deeper burnup can be obtained with appropriate initial k_{eff} and CR by varying either C/HM or the U-233 weight fraction. Furthermore, a similar burnup can be achieved for the FHR core with a higher

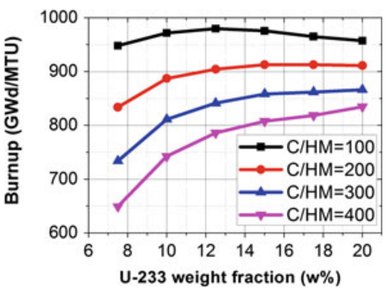

Fig. 5 Burnup for FHR core as a function of the U-233 weight fraction (w%) for different C/HM

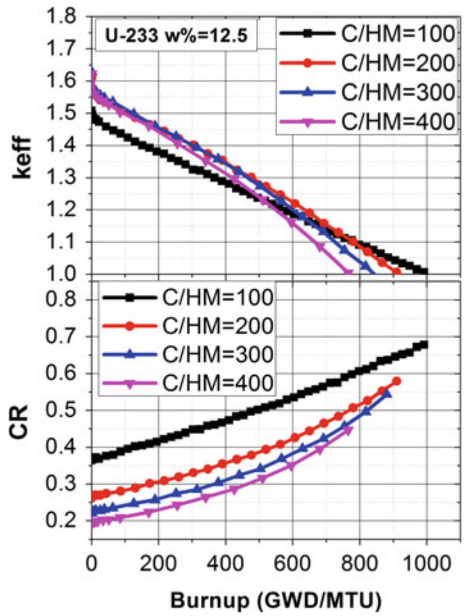

Fig. 6 Evolution of k_{eff} (*top*) and CR (*bottom*) with the burnup of U-233 w% = 12.5 for different C/HM

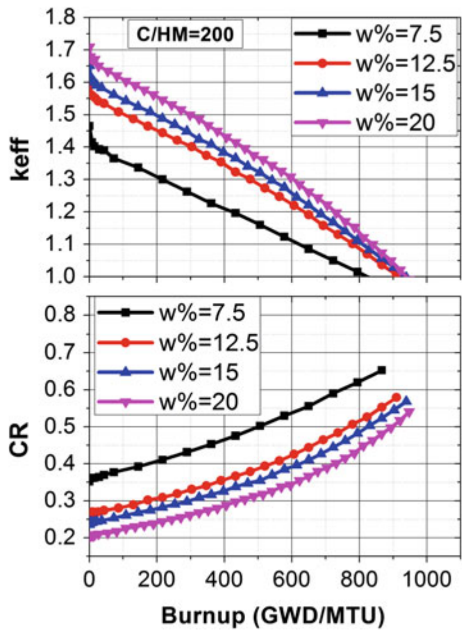

Fig. 7 Evolution of k_{eff} (*top*) and CR (*bottom*) with the burnup of C/HM = 200 for different U-233 weight fractions

U-233 weight fraction for different C/HM. All of such phenomena can be explained from the perspective of neutron balance or the six-factor formula, and it is not discussed here.

4 Conclusions

In order to improve the TCR for thorium-based fuel (Th2/U3), the influences of the U-233 weight fraction and the number of TRISO-coated particles (corresponding to C/HM) upon neutronics for a FHR core are investigated, including the spectrum, the initial k_{eff}, the TCR, and the burnup. The conclusions drawn from the above analysis are shown as below:

- A harder spectrum shows a larger spectrum factor. The lower the C/HM, the higher the U-233 weight fraction, the larger spectrum factor can be achieved. The U-233 weight fraction is of great importance for the spectrum factor with low C/HM while the C/HM is more significant for the spectrum factor when U-233 weight fraction becomes greater.
- An improved TCR can be obtained by reducing C/HM. As the U-233 weight fraction increases, the negative TCR tends to be a larger value for the FHR core with a higher C/HM ($\geq$ 200) but trends toward a smaller value for the one with a lower C/HM (e.g., 100).
- A deeper burnup can be achieved for the FHR core with a lower C/HM and/or a higher U-233 weight fraction, which displays a relatively higher initial k_{eff} and/or a comparatively better conversion ratio benifited from a harder spectrum.

From the aspects of improved TCR and deeper burnup, a FHR core with C/HM of 200 and U-233 weight fraction of 12.5 % is recommended for the optimization of thorium utilization. Multizone with different C/HM is also required to improve the conversion ratio of thorium. Moreover, elaborate fuel pebble arrangement and fuel pebble recycling are expected to obtain a deeper burnup.

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