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Transition toward thorium fuel cycle in a molten salt reactor by using plutonium

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Abstract The molten salt reactor (MSR), as one of the Generation IV advanced nuclear systems, has attracted a worldwide interest due to its excellent performances in safety, economics, sustainability, and proliferation resistance. The aim of this work is to provide and evaluate possible solutions to fissile ^{233}U production and further the fuel transition to thorium fuel cycle in a thermal MSR by using plutonium partitioned from light water reactors spent fuel. By using an in-house developed tool, a breeding and burning (B&B) scenario is first introduced and analyzed from the aspects of the evolution of main nuclides, net ^{233}U production, spectrum shift, and temperature feedback coefficient. It can be concluded that such a Th/Pu to Th/^{233}U transition can be accomplished by employing a relatively fast fuel reprocessing with a cycle time less than 60 days. At the equilibrium state, the reactor can achieve a conversion ratio of about 0.996 for the 60-day reprocessing period (RP) case and about 1.047 for the 10-day RP case. The results also show that it is difficult to accomplish such a fuel transition with limited reprocessing (RP is 180 days), and the reactor operates as a converter and burns the plutonium with the help of thorium. Meanwhile, a pre-breeding and burning (PB&B) scenario is also analyzed briefly with respect to the net ^{233}U production and evolution of main nuclides. One can find that it is more efficient to produce ^{233}U under this scenario, resulting in a double time varying from about 1.96 years for the 10-day RP case to about 6.15 years for the 180-day RP case.

Keywords Molten salt reactor · Thorium fuel cycle · Plutonium · Reprocessing

1 Introduction

A molten salt reactor (MSR) uses the fluid salt containing fissile and fertile materials as its fuel and the primary coolant with unique features of on-line salt processing and refueling. Under development since 1947 in the Oak Ridge National Laboratory (ORNL), the MSR has been studied in several countries and shown to have remarkable advantages over current light water reactors (LWRs), such as the inherent safety, excellent fuel utilization efficiency, and flexible fuel cycle options [1–3]. Operating on a thorium fuel cycle, MSRs with a thermal or a fast neutron spectrum can achieve desirable breeding capacity, long-term waste reduction, and proliferation resistance [4–8]. Launched in 2011, the Thorium Molten Salt Reactor (TMSR) nuclear energy system in China aims

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to develop both solid and liquid fueled MSRs within 20–30 years in order to achieve effective and sustainable thorium energy utilization [9–12].

With a view to develop and deploy an advantaged MSR operating on a thorium fuel cycle, prior generation of fissile ^{233}U is required, considering that there is no available ^{233}U in nature. It is expected that MSR can be started with the fissile materials (plutonium or enriched uranium) arising from the well-established uranium/plutonium fuel cycle and then produce ^{233}U through neutron irradiation of ^{232}Th [13]. Using low enriched uranium (LEU) in MSR is attractive and it has been studied in both the designs of the Denatured Molten Salt Reactor (DMSR) [14] and the Transatomic Power (TAP) MSR [15]. However, it has generally been ruled out due to the presence of too much of ^{238}U, which makes the ^{233}U production and further the transition toward to thorium fuel cycle difficult. Meanwhile, driven by the proliferation issue, the use of enriched uranium beyond 20% in civilian reactors was abandoned all over the world [13]. As an alternative driver fuel to enriched uranium, plutonium partitioned from LWR spent fuel, is appealing for a MSR startup and ^{233}U production when it couples with thorium. Furthermore, burning plutonium can also close the current once-through fuel cycle used in LWRs and thus make a much more efficient use of plutonium [16].

Much work has been done on the plutonium-started MSR. In the studies of the Molten Salt Breeder Reactor (MSBR) designed by ORNL [17], startup on plutonium was analyzed, and the results indicated that the MSBR startup and the fuel transition from Th/Pu to Th/^{233}U can be accomplished based on the isolation of plutonium isotopes (mainly ^{242}Pu) from the circulating fuel salt when their reactivity contribution is negative after about 3.6-year operation, and the breeding ratio, averaged over a 30-year reactor life, is about 1.052 [18]. A similar work, undertaken by Alexis Nuttin et al. in 2002, also evaluated the feasibility of the transition to a thorium fuel cycle in a MSBR with a single-fluid, one-zone core [8]. Plutonium from a pressurized water reactor (PWR) spent fuel was used as the initial driver fuel and the bred ^{233}U was fed back into the core to maintain criticality after startup. It was found that the thorium fuel cycle can also be achieved and the corresponding double time is 35 years, about 10 years longer than the ^{233}U startup case. However, as presented in Alexis Nuttin's work, the produced ^{233}U is not enough to offset its depletion during the first about 13 years, and the maximum deficit of ^{233}U is more than 300 kg. Regarding the work of FUJI-Pu in Japan, a 250 MWth MSR with a three-zone core was designed, and its startup fuel is the plutonium from PWR 33 GWd/t spent fuel. The detailed calculation showed that the FUJI-Pu can transmute 980 kg of plutonium isotopes and at the same time, produce 489 kg ^{233}U annually for a 1 GWe reactor [19]. Moreover, in the studies of the Molten Salt Fast Reactors (MSFRs), Pu (or TRU) was commonly used for efficient burning plutonium and producing ^{233}U [20, 21]. The results indicated that the ^{233}U production is much better for the Pu-started than the ^{233}U directly started in the MSFR.

The previous work presented above has demonstrated the fact that it is feasible to efficiently produce ^{233}U by using plutonium in both thermal and fast MSRs. The aim of this work is to discuss and compare the possible solutions to achieve the fuel transition from plutonium to thorium fuel cycle. Two main solutions are proposed according to whether the produced ^{233}U from the decay of the extracted ^{233}Pa is fed back into the core or accumulated outside the core to restart a new MSR. If the produced ^{233}U is fed and progressively replaces the plutonium, we name it as the breeding and burning (B&B) scenario. Otherwise, if the produced ^{233}U is stored outside the core and used as the startup fuel for a new MSR, it is called as the pre-breeding and burning (PB&B) scenario.

The methodology for the geometry and the calculation tool are introduced in Sect. 2. The discussions on the B&B scenario are presented in Sect. 3, while that on the PB&B scenario are given in Sect. 4. Comparison of the two scenarios is carried out in Sect. 5 and Sect. 6 gives the conclusions.

2 Methodology

2.1 Core geometry and material description

As shown in Fig. 1, a cylinder core (2.1 m radius and 4.6 m height) comprising of a lattice of 10 cm pitch hexagonal elements with hollow cylindrical channels to allow fuel salt circulation is chosen. The radius of these fuel channels is

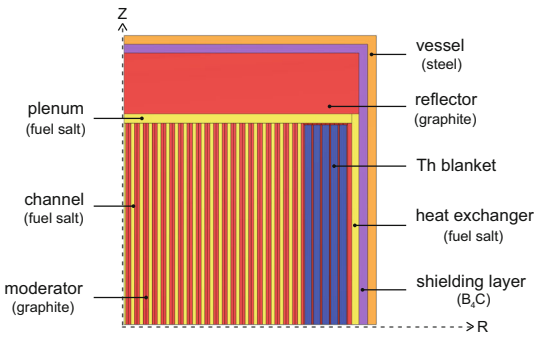

Fig. 1 (Color online) Geometrical description for the quarter of the reactor core

2.03 cm, corresponding to a salt fraction of about 15%. The total volume of the fuel salt in the reactor is about 20 m^3, which contains three parts: the salt in the graphite channels in the core (9 m^3), the plenum salt above and below the core (4.5 m^3), and the salt in the heat exchangers that are external to the core (6.5 m^3). A 50-cm-thick radial thorium blanket is arranged to improve the breeding capacity and it also consists of the same graphite hexagonal elements but with a larger channel radius of about 4 cm. This geometry is based on the optimization by L. Mathieu et al. [22], but the core height to diameter ratio is adjusted to about 1.095 instead of 1.0 in order to improve the temperature reactivity coefficients in such a thermal MSR.

In order to enhance the solubility of PuF_3 to convert ^{232}Th to ^{233}U in such a Pu-started MSR, a fuel salt with 78 mol% LiF-22 mol% ($ThF_4 + PuF_3$), rather than the LiF/BeF_2 salt used in MSBR [17], is chosen. Its density at the mean operation temperature (973 K) is about 4.125 g/cm^3 with a dilatation coefficient of 8.82×10^{-4} g/cm^3/K [23]. ^7Li is enriched to 99.995% to lower the neutron capture in ^6Li. The salt used in the blanket is the same as the fuel salt except for the absence of plutonium. The nuclear grade graphite used as the moderator and reflector has a density of 1.843 g/cm^3.

2.2 Calculation tool descriptions

In this work, an in-house developed tool, named MSR reprocessing sequence (MSR-RS) [24, 25], is employed, which can be used to simulate on-line processing and refueling by coupling with various functional modules in SCALE6.1 [26]. Figure 2 presents the detailed flowchart of MSR-RS. The criticality calculation performed by KENO-VI, a three-dimensional monte carlo criticality computer code, is based on a whole core. The Couple calculation is used to produce the binary nuclear data libraries for Origen-s code by combining problem-dependent, one-group cross sections with state-of-the-art ENDF/B-VII nuclear decay data and energy-dependent fission product yields. The one-group cross sections are generated by using a multigroup cross-sectional library and the problem-dependent multigroup fluxes which are prepared in the KENO-VI sequence with a 238-group ENDF/B-VII nuclear base. The Origen-s calculation is performed with on-line refueling and reprocessing. Afterward, the effective multiplication factor (k_{eff}) is obtained based on the KENO-VI module to determine whether the feed rate is appropriate for critical operation. If k_{eff} is within the range, the next step calculation will be performed similarly until all burn-up steps are accomplished. It should be noted that the core power (2250 MWth) is maintained constant in each step of depletion calculation.

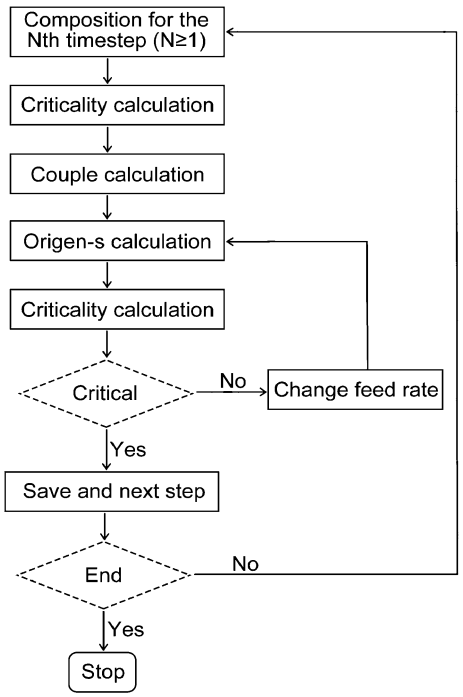

Fig. 2 Flowchart of MSR-RS

2.3 Feeding modes and processing options

Considering the depletion of the initial plutonium and thorium, and the accumulation of the fission products (FPs), the fissile and fertile materials should be fed into the core to ensure the reactor steady-state operation. In both of the B&B scenario and the PB&B scenario, the amount of ^{232}Th is kept constant, while the concentration of fissile fuel is adjusted as required to maintain the criticality of the core. ^{233}Pa is extracted continuously and placed outside the core to decay into ^{233}U. However, the feeding of the fissile materials is different from each other for the two scenarios. In the B&B scenario, if the produced ^{233}U is enough to maintain the reactor criticality, no additional plutonium is needed to be fed and the excess ^{233}U is stored. Otherwise, extra plutonium would have to be added into the core to compensate for the lack of ^{233}U. While in the PB&B scenario, all the ^{233}U produced from the decay of the extracted ^{233}Pa will be accumulated outside the core until it reaches the required startup mass for a new reactor. Thus, the criticality of the core is maintained by refueling plutonium.

The salt processing, mainly associated with the protactinium extraction and the FPs removal, is complicated but essential to obtain a high breeding performance. In this work, the fuel processing scheme used in the MSBR project [27] is chosen and the influence of the reprocessing period (RP) on the fuel transition is analyzed by increasing the reference period (10 days) to 2 and 6 months (the

extraction efficiencies are assumed constant). The processing of the fertile salt in the blanket is similar to that in the core, but the ^{233}U produced in the blanket is also extracted by effective fluorination.

3 Results and discussions for B&B scenario

3.1 Evolution of uranium and plutonium isotopes

The initial plutonium obtained from LWR (burn-up of 60 GWd/t) spent fuel consists of about 4% ^{238}Pu, 51% ^{239}Pu, 25% ^{240}Pu, 12% ^{241}Pu, and 8% ^{242}Pu [28]. At the initial time, about 6.85 t plutonium and 41.5 t ^{232}Th dissolved in the fuel salt are required to reach criticality. Figures 3 and 4 give the evolutions of the inventories of main plutonium isotopes and uranium isotopes in the fuel salt for all the three RP options respectively. It should be noted that the initial load of Pu between the three RP cases is equal, and the slight difference shown in Fig. 3 is mainly caused by the additional Pu within a relatively short time after startup. For both of the 10-day and the 60-day RP cases, a relatively small amount of additional plutonium (RP = 10 days: 268 kg; RP = 60 days: 533 kg) is required before the reactor becomes self-sustaining. Thereafter, the concentrations of ^{239}Pu, ^{240}Pu, and ^{241}Pu decrease rather sharply (see Fig. 3a, b), while those of ^{238}Pu and ^{242}Pu decrease slowly before 10 years due to the key reactions from ^{239}Pu and ^{241}Pu, which are shown as follows:

$$^{239}\mathrm{Pu} \xrightarrow{(n,\gamma)} {}^{240}\mathrm{Pu} \xrightarrow{(n,\gamma)} {}^{241}\mathrm{Pu} \xrightarrow{(n,\gamma)} {}^{242}\mathrm{Pu}$$

$$^{241}\mathrm{Pu} \xrightarrow{\beta} {}^{241}\mathrm{Am} \xrightarrow{(n,\gamma)} {}^{242}\mathrm{Am}$$

$$^{242}\mathrm{Am}(83.1\%) \xrightarrow{\beta} {}^{242}\mathrm{Cm} \xrightarrow{\alpha} {}^{238}\mathrm{Pu}$$

After 20 years, the inventories of all plutonium isotopes approach their equilibrium. Meanwhile, one can find from Fig. 4a, b that the inventory of ^{233}U in both the RP cases first increases quickly from the startup to gradually replace the fissile plutonium isotopes (^{239}Pu and ^{241}Pu) to maintain the reactor criticality and then decreases owing to the depletion of the other three Pu isotopes (^{238}Pu, ^{240}Pu, ^{242}Pu) and the accumulated Minor Actinides (MAs), mainly ^{241}Am, ^{243}Am, and ^{244}Cm, which have a much higher capture cross section than their fission cross section. An equilibrium inventory of ^{233}U is reached at about 30 years. The equilibrium value for ^{233}U is about 790 kg for the 10-day case and about 850 kg for the 60-day case, both of which are a little more than the required critical mass (about 753 kg) to start a new reactor with ^{233}U.

As a result of the above fuel transition, fission rate of the main fissile materials (^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu) in the core has also changed. In order to illustrate this, the 10-day RP case is taken as an example to present the detailed evolution of the fraction of total fission rate, as shown in Fig. 5. The fraction of ^{239}Pu decreases rather quickly from about 72.4% at initial time to about 3.7% at 10 years, while ^{241}Pu's contribution to fission rate rises to 32.5% at about 5 years due to the captures in ^{240}Pu, which extends the fission of Pu beyond 10 years after startup. Meanwhile, ^{233}U plays a more and more important role in maintaining the criticality of the core and its fraction exceeds that of the ^{239}Pu and ^{241}Pu after about 3.8 years. At equilibrium state, ^{233}U contributes about 87.4% of the total fission rate, and ^{235}U is about 10%.

Compared with the other two RP cases, the system with the 180-day RP is complicated regarding to the evolutions of the inventories of Pu isotopes and U isotopes, as shown in Figs. 3c and 4c. After an almost similar evolution to above two RP cases for about 45 years, the inventories of plutonium isotopes increase sharply. In order to explain this, the fractions of fission rate for the fissile nuclides are also given, as presented in Fig. 6. It can be found that the contribution of ^{233}U to the total fission rate increases continuously and exceeds 80% at about 28 years, thus resulting in an increase of the added ^{233}U and therefore, a

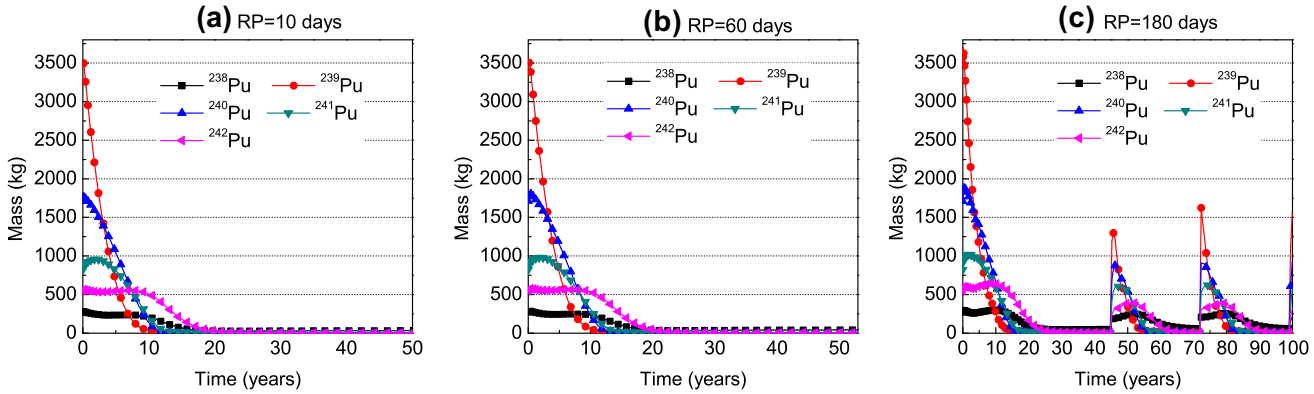

Fig. 3 (Color online) Evolution of plutonium isotopes in the core for different RP options. **a** RP = 10 days. **b** RP = 60 days. **c** RP = 180 days

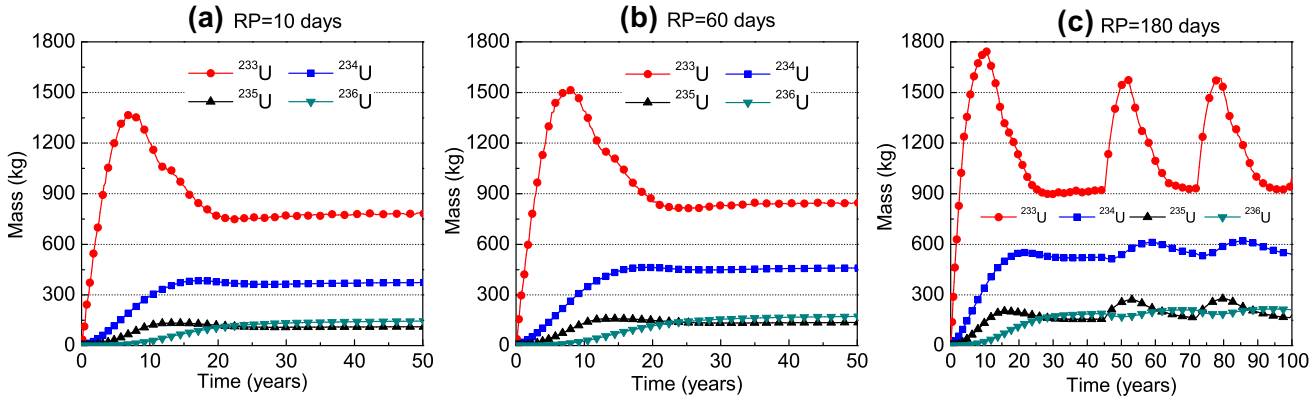

Fig. 4 (Color online) Evolution of uranium isotopes in the core for different RP options. **a** RP = 10 days. **b** RP = 60 days. **c** RP = 180 days

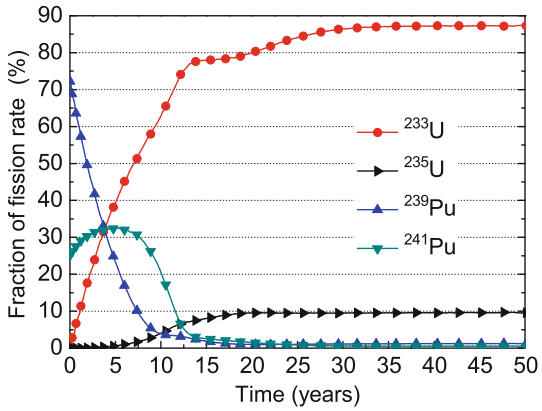

Fig. 5 (Color online) Fission rate fraction of ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu for the 10-day RP case

decrease of the accumulated ^{233}U that is stored outside the core. At about 45 years, the accumulated ^{233}U has been almost burnt off and the external plutonium would have to be refueled into the core to maintain the criticality of the core, which accounts for the sharp increase of the inventories of plutonium isotopes. Influenced by the refueled

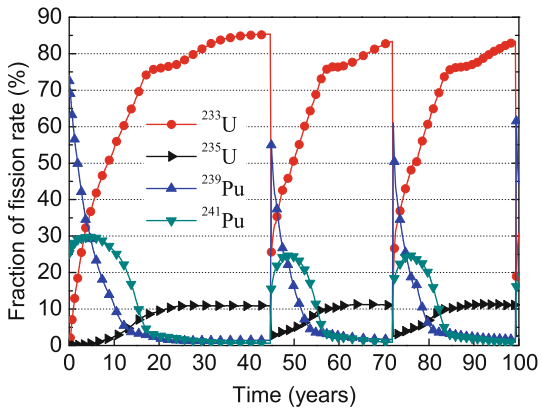

Fig. 6 (Color online) Fission rate fraction of ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu for the 180-day RP case

plutonium, the fission fraction of ^{233}U decreases drastically and then increases with the inventory of ^{233}U in the core. This process is very similar to that of startup and such complicated transition behaviors involved with the increase and decrease of plutonium and ^{233}U will repeat periodically in the following years, as indicated in Figs. 3c and 4c. Therefore, for the 180-day RP case, the fuel transition can only be accomplished by refueling external fissile material successively during the operation, which is significantly different from the other two RP cases. More details on the accumulated ^{233}U stored outside the core will be discussed in the next subsection in order to better understand these transition behaviors.

3.2 Net ^{233}U production and conversion ratio

Two main parameters of interest to evaluate the transition performance, the net ^{233}U production, and the conversion ratio, are discussed in this section. After startup with the plutonium, the reactor can sustain a critical operation on both the additional supplied plutonium and its self-produced ^{233}U. When the reactor operates solely on its self-produced ^{233}U and the reactivity for the core is beyond the allowable level ($k_{\text{eff}} = 1$), some excess ^{233}U as a net production should be stored outside the core to control the reactivity. It is obvious that the stable increase of net ^{233}U production can only be achieved in a breeder reactor. Figures 7 and 8 give the evolutions of net ^{233}U production and associated conversion ratio for various RP options. It is found that the net ^{233}U can be produced even before the conversion ratio actually exceeds unity, which is due to the fact that ^{233}U is more reactive than ^{239}Pu in such a thermal neutron reactor. One can also find that the net ^{233}U production is strongly dependent on the processing rate, and this will be discussed below.

The net ^{233}U production can be analyzed from the aspects of ^{233}U breeding and depletion. The transition case

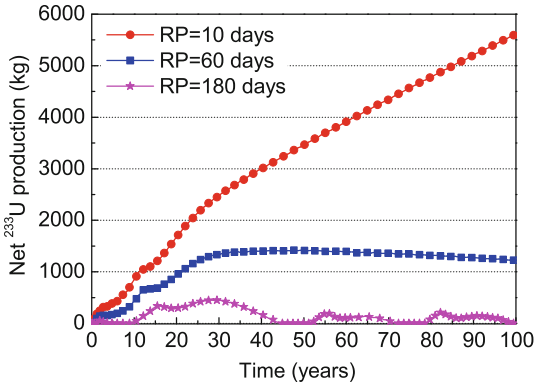

Fig. 7 (Color online) Net ^{233}U production for different RP options in B&B scenario

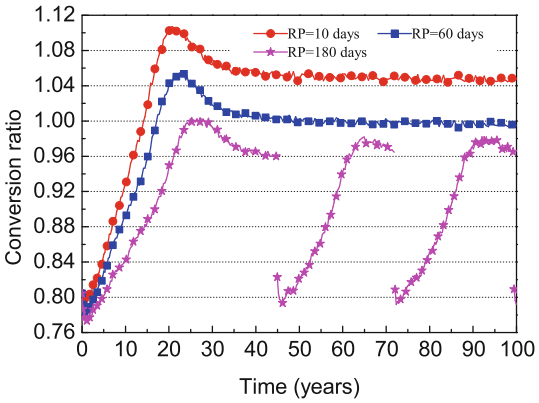

Fig. 8 (Color online) Conversion ratio for different RP options

with a 10-day RP is chosen to discuss the evolution of the net ^{233}U production. During the first 7 years of this case, the ^{233}U inventory increases relatively quickly to its peak value in order to supply the reactivity loss caused by two factors: the produced MAs and the depleted initial fissile plutonium isotopes. As shown in Fig. 5, ^{233}U's contribution to the total fission rate has increased to about 50% at 7 years. Meanwhile, the conversion ratio increases quickly during this stage. A combined result of the ^{233}U production and depletion is a slow increasing of the net ^{233}U production. While in the following 5 years (7–12 years), the conversion ratio increases continuously and more quickly than the ^{233}U depletion rate (^{233}U's fraction of the total fission rate at 12 years is about 73%), thus leading to a higher net ^{233}U production rate than that in the first 7 years. The third stage (12–28 years) is similar to the second stage but with an even higher conversion ratio. The last stage is the equilibrium stage, which corresponds a constant annual yield of about 42 kg. For the 60-day RP transition case, the behavior of the net ^{233}U production before 28 years is almost similar to that of the 10-day RP case but with a much lower net ^{233}U production. The conversion ratio at the equilibrium state is very close to 1.0 (about 0.996), resulting in a very slow decrease of net ^{233}U production.

As discussed in the previous subsection, the transition behaviors are complicated for the 180-day RP case, which is determined by the degraded breeding capacity stemming from the slow fuel reprocessing. It can be found from Fig. 7 that the net ^{233}U production decreases when ^{233}U contributes about 80% of the total fission rate (see Fig. 6) and slightly increases when external Pu is refueled into the core, which can greatly reduce the consumption of ^{233}U. Meanwhile, the corresponding conversion ratio, shown in Fig. 8, first increases, then decreases, and then fluctuates periodically. Consequently, the fuel transition from the initial plutonium to thorium fuel cycle cannot be accomplished, and the reactor operates as a converter with such a limited processing option.

3.3 Neutron spectrum shift

During the fuel transition, the neutron spectra in both the fuel zone and the fertile blanket are shifted downward (toward softer) from the initial time to the equilibrium state ($T = 100$ years), as illustrated in Fig. 9, which gives the spectrum shift for the 10-day RP case. The sufficiently strong thermal neutron captures in plutonium isotopes, including the resonance captures in ^{240}Pu at about 1.06 eV (The capture cross section is about 1.15×10^5 barns) and ^{242}Pu at about 2.67 eV (The cross section is about 3.21×10^4 barns), are the primary contributors to the harder spectrum at the initial time (see Fig. 9a). While at the equilibrium state for the Th/^{233}U fuel cycle, the plutonium isotopes are almost burnt off and simultaneously, the moderator-to-fuel ratio becomes higher due to decrease of the heavy mental inventories, both of which result in softening the neutron spectrum. Such a distinct spectrum shift can certainly affect the core behaviors, one of which is the temperature reactivity coefficients that will be discussed in the next section.

The neutron spectra for the 60-day RP case are almost similar to those for the 10-day case, but a little harder at the equilibrium state due to more FPs accumulation. As to the neutron spectra shift for the 180-day RP case, it is more complicated owning to the re-supplied plutonium at about 45, 72 and 99 years. In order to illustrate this, a new parameter, energy of the average lethargy causing fission (EALF) is introduced to evaluate how fast or thermal the reactor spectrum is Ref. [29], which is shown in Fig. 10. It can be found from the comparison that the EALF for the 180-day RP case fluctuates periodically, which is caused by the refueling of Pu fuel as discussed in Sect. 3.1.

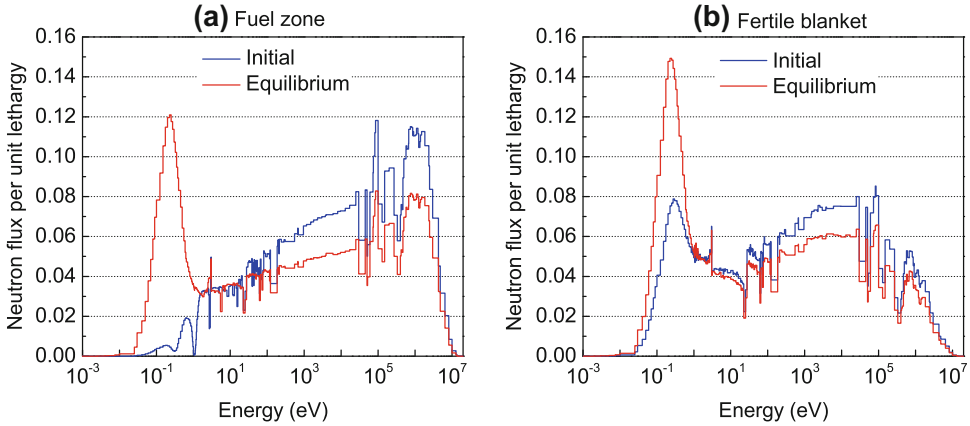

Fig. 9 (Color online) Initial and equilibrium normalized neutron spectra of the fuel zone and fertile blanket for the 10-day RP case. **a** Fuel zone. **b** Fertile blanket

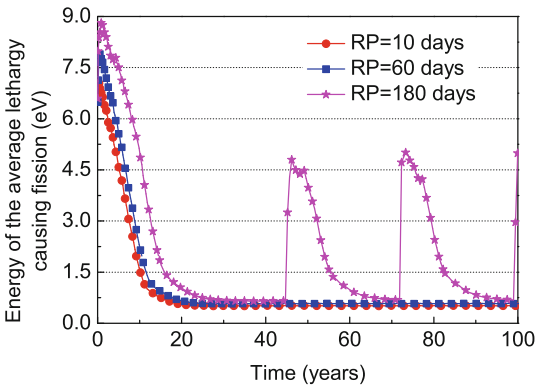

Fig. 10 (Color online) Energy of the average lethargy causing fission (EALF) for different RP options

3.4 Temperature reactivity coefficients

The temperature reactivity coefficients (TRC) is an important parameter to evaluate the safety performance during the transition. It is responsible to the reactivity transient and can be calculated based on reasonable approximation that the core behaves isothermally. Actually, due to the fact that the most nuclear fissions take place in the core active region, the fuel salt temperature changes much faster and stronger than the moderator temperature and the fertile salt temperature. Accordingly, the TRC of the three components at the mean operation temperature (973 K) are calculated separately and their combined effects are also given.

The fuel salt TRC is a combined effect of the Doppler effect and the fuel dilatation effect, and therefore it can be calculated by changing the fuel salt temperature (corresponds to the Doppler effect) and density (corresponds to the dilatation effect caused by temperature change) simultaneously (the parameters of the moderator and the fertile salt are kept constant). In this calculation, the effective multiplication factor (k_{eff}) at three points (893, 973, and 1053 K) is calculated and the fuel salt TRC can be obtained from a linear fit to the data plotted as reactivity versus $1/T$ ($\delta\rho/\delta T$, T is the fuel salt temperature). At 973 K, the reactivity is about zero in order to simulate the transient response of a critical system. Similar calculations for the fertile salt TRC are conducted. The TRC of the moderator in the core active region and fertile zone (the reflector graphite is not included) corresponds to the effect of increasing of the moderator temperature (Similarly, the parameters of the fuel salt and fertile salt are kept constant), and it can also be calculated by using linear fitting method. The graphite density coefficient is usually small and can be essentially negligible [30]. The total TRC combines above three effects together assuming that all the three components undergo equal temperature changes. It should be noted that the selected temperatures (893 and 1053 K) are based on the consideration of the mean operation temperature (973 K) and the range of validity of the formula for salt density given by Ignatiev et al. [23].

Figure 11 presents the fuel salt TRC, fertile salt TRC, moderator TRC, and the total TRC for the 10-day RP case

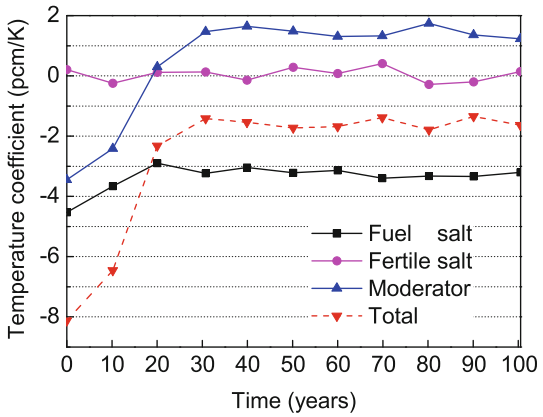

Fig. 11 (Color online) Temperature reactivity coefficients for the 10-day RP case

respectively. One can firstly find that the total TRC is always negative during the operation, but varies from −8.17 pcm/K at initial time to about −1.75 pcm/K at about 100 years. This remarkable change is mainly due to the graphite TRC which appears to be quite sensitive to the fuel composition and the neutron spectrum in the core. When the temperature of the graphite increases, the spectrum shifts upwards, and further results in relative cross-section changes between fissions in fissile nuclides and captures in fertile nuclides, which is the primary source of the moderator reactivity effect. The calculation shows that the captures in fertile nuclides increase, especially for ^{240}Pu, while the fissions in ^{239}Pu and ^{241}Pu decrease if the graphite temperature increases at the initial time. With respect to the positive graphite TRC after the transition, it is also caused by the spectrum shift, which has been discussed in detail [8, 30]. As to the fuel, the TRC increases from −4.37 pcm/K at the initial time to about −3.0 pcm/K at the equilibrium state owning to both of the increase of the fuel doppler effect and density effect.

The TRC for the 60-day RP case is similar to that for the 10-day RP case and the total TRC at 100 years is about −1.68 pcm/K. As to the 180-day RP case, the total TRC fluctuates between a sufficiently negative value (about −7 pcm/K) and a relatively small value (about −1.51 pcm/K), which is also caused by the re-supplied plutonium.

4 Results and discussions for PB&B scenario

It can be concluded from the discussion in Sect. 3 that the MSR can achieve the fuel transition from a plutonium to a thorium fuel cycle under the B&B scenario when employing a RP of no more than 2 months. In this section, the PB&B scenario, defined above, is analyzed in order to provide an alternative option for ^{233}U production and fuel transition, particularly under limited processing conditions. The PB&B scenario stores the bred ^{233}U outside the core rather than refueling it back into the core in the B&B scenario, which is advantageous to produce enough ^{233}U as quickly as possible for starting a new reactor operating on the Th/^{233}U fuel cycle. Similarly, three groups of RP options are employed. Figure 12 gives the net ^{233}U production for different RP options. For the startup of the reactor described in Sect. 2, about 753 kg of ^{233}U is required at the initial time. The maximum net ^{233}U production shown in Fig. 12 is about 1000 kg which is enough for startup and feeding before the reactor becomes self-sustaining.

It can be found from Fig. 12 that it is efficient to produce ^{233}U by employing this scenario, in which the criticality of the reactor is maintained mainly by the external plutonium,

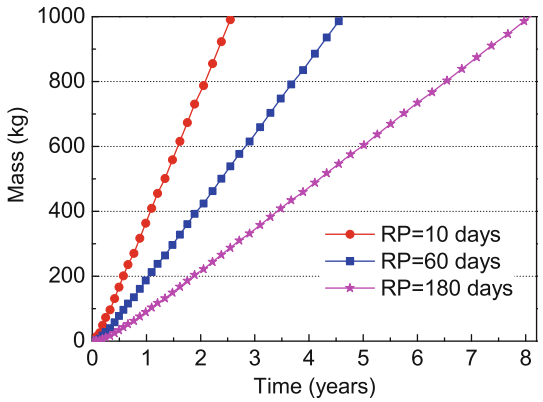

Fig. 12 (Color online) Net ^{233}U production for different RP options in PB&B scenario

particularly for the case with the 10-day RP. The double time (corresponding to the critical mass of 753 kg) varies from about 1.96 years to about 6.15 years for the chosen RP options. Actually, in this scenario, it is also feasible for ^{233}U production if the uranium isotopes (mainly ^{233}U) are periodically isolated from the core by applying fluorination, and this is a major difference compared with a similar scenario for ^{233}U production that uses the enriched uranium as driver fuel rather than plutonium.

During the ^{233}U production stage, the total inventory of the heavy nuclides in the fuel salt increases in all the RP cases, but it is not serious and the physicochemical properties of the fuel salt can still be kept stable. In order to illustrate this, the evolution of the main heavy nuclides inventories for the 180-day RP is presented in Fig. 13. One can find that the ^{233}U inventory increases quite slowly compared with the same RP case in the B&B scenario. The fissile ^{241}Pu also increases due to a strong neutron capture of ^{240}Pu. At about 8 years, the relative fission fractions for ^{233}U, ^{239}Pu, and ^{241}Pu are approximately identical (29.44, 33.13, and 30.95%, respectively). As a result of such a

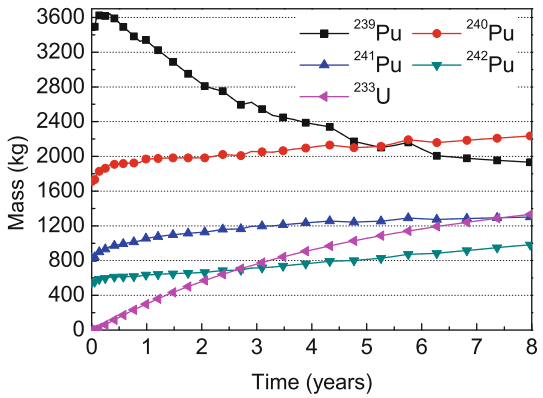

Fig. 13 (Color online) Evolution of the inventories of the main nuclides for the 180-day RP

variation of the inventories, the mole fraction of the total heavy nuclides increases by about 0.9%.

If the produced ^{233}U is enough for startup, it can be used to start directly the reactor with the processed carrier salt. In such a way, the fuel transition to thorium fuel cycle, and then deploying a thorium-based MSR, can be realized.

5 Comparison and discussions of the two scenarios

In Sects. 3 and 4, two scenarios for the transition toward the thorium fuel cycle in MSR by using plutonium have been discussed. Table 1 presents an overview of initial Pu and added Pu to produce critical ^{233}U mass (753 kg) for a new MSR. The corresponding double time varies from about 1.96 years for the PB&B scenario with a 10-day RP to about 17 years for the B&B scenario with a 60-day RP, which is mainly due to the different feeding modes described in Sect. 2.3. A parameter named M/C ratio is introduced to evaluate the ratio of the critical ^{233}U mass to the consumed Pu (initial Pu and added Pu). One can find that the M/C ratio in the B&B scenario is a little larger than that in the PB&B scenario when employing the same reprocessing period.

Fast reprocessing employed in both scenarios is attractive with respect to double time and net ^{233}U production. However, it is challenging considering the complexity of the salt reprocessing, particularly for the Th–U–Pu mixed system discussed in this work in view of the fact that the reprocessing of thorium-based fuel cannot be achieved with processes that are currently used for uranium fuel [13]. Therefore, the 10-day reprocessing period used in this work may be a long-term consideration for commercial MSRs. At present, salt reprocessing is still in the research and development phase and an off-line, batch processing solution based on current chemical partitioning technology is preferred. Fortunately, much work has been done to enhance the reprocessing technology and an on-line, fast reprocessing scheme is expected to be well dealt with in the following years.

6 Conclusion

In this work, two scenarios for achieving the MSR fuel transition from the Th/Pu to the Th/^{233}U fuel cycle have been put forward and studied, respectively, by employing an in-housed developed calculation tool. The B&B scenario, which uses the produced ^{233}U to gradually replace the initial starter fuel of the plutonium, is firstly analyzed in detail. It is found that the fuel transition can be accomplished if the reprocessing period is within 60 days. Under this condition, a relatively small amount of additional plutonium is required to supply the reactor after startup, and the breeding ratio at the equilibrium state varies from about 0.996 for the 60-day reprocessing period to about 1.047 for the 10-day reprocessing period. Meanwhile, the neutron spectrum shifts downward due mainly to the decrease of the thermal captures in plutonium isotopes. Moreover, the analysis also indicates that the total temperature reactivity coefficient is always negative at the operating temperature during the operation although it becomes less negative due to the depletion of the plutonium isotopes.

With limited processing (reprocessing period is 180 days), the fuel transition cannot be accomplished and the transition behavior is complicated due to a periodical plutonium supplement. The conversion ratio fluctuates between about 0.8–0.96 and the reactor operates as a converter. In such a system, the reactor can be used to burn the plutonium isotopes from LWR spent fuel with the help of thorium.

The PB&B scenario, as an alternative solution to ^{233}U production and further utilization in a MSR, has also been analyzed briefly in this work. The results indicate that it is more efficient to produce ^{233}U and the double time corresponding to the 180-day reprocessing period is just about 6.15 years. In such a system, the total proportion of the heavy nuclides increases by about 0.9%, ensuring the relatively stability of the fuel salt.

Two scenarios for the fuel transition presented in this work are both attractive; however, much work is still needed to be done to improve the feasibility of such a Th/

Table 1 Overview of initial Pu, added Pu and double time for the B&B and PB&B scenarios

Scenarios	RP (days)	Initial Pu (kg)	Added Pu (kg)	Double time (years)	M/C ratio
B&B	10	6841	268	9.31	0.106
	60		533	17	0.102
	180		15580	–	–
PB&B	10	6841	3405	1.96	0.073
	60		3590	3.51	0.072
	180		4868	6.15	0.064

Pu to Th/^{233}U transition considering the complexity of the reprocessing associated with a Th/Pu/U mixed system.

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