Minor Actinides Incineration in a Small Molten Salt Fast Reactor

Chenggang Yu, Chunyan Zou, Yuwen Ma and Jingen Chen

Abstract Minor actinides (MA) accumulated in spent fuel are the primary contributors to the long-term radiological hazards of high-level nuclear waste. Due to its outstanding features such as the function of reprocessing and refueling, large negative temperature feedback coefficient and no fuel assembly fabrication, Molten Salt Fast Reactor (MSFR) is regarded as one of the candidate reactors for MA incineration. In the present work, we evaluate the MA incineration capability for a 500 MWth MSFR by considering FLiBe fuel carrier salts with different initial MA loadings. The simulated results show that the MA transmutation capability has a positive correlation to the MA loading. When MA = 18.17 mol%, the transmutation fraction during 50-year operation can achieve about 95%. The MA feeding into the fuel salt is also analyzed to obtain its influences on the MA transmutation performance. The simulated result shows that both the amount of MA transmutation and the depletion ratio of MA to heavy nuclei also have a positive correlation to the MA feeding.

Keywords Molten salt fast reactor · Minor actinides transmutation

1 Introduction

The open fuel cycle mode is used in the commercial pressurized water reactor (PWR). In this once-through fuel cycle mode, a lot of spent nuclear wastes (SNF) are produced during the PWR operating. Most of the SNF needs geological

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H. Jiang (ed.), *Proceedings of The 20th Pacific Basin Nuclear Conference*,
DOI 10.1007/978-981-10-2314-9_44

disposal or temporary storage up to now. However, the above processing modes are unfavorable to reduce the radioactive dose or to improve the nuclear fuel utilization. Transuranic (TRU) is the primary contributor to the long-lived high-level radioactive hazards in SNF. Transmutation of TRU is one of the most effective ways to solve the SNF safety issues [1–3]. Pu can be made as mixed oxide fuel (MOX) and reused in PWR. However, the transmutation of TRU composing only minor actinides (MA) should be paid more attention, since MA cannot be well burnt in a thermal reactor.

Accelerator-Driven Subcritical System (ADS) [4] and fast reactors including Gas-cooled Fast Reactors (GFR) [3], Sodium-cooled Fast reactor (SFR) [5] and liquid fueled fast reactors [6–8] are recognized as alternative systems to transmute MA for their larger fission-to-capture ratio, deeper burnup compared with a thermal reactor. As one of the six candidate reactors in the Generation IV International Forum (GIF), liquid fueled fast reactor has outstanding features for MA transmutation such as the functions of reprocessing and refueling, large negative temperature feedback coefficient and no fuel assembly fabrication. The MOlten Salt Actinide Recycler & Transmuter (MOSART) proposed by the Kurchatov Institute of Russia within the International Science and Technology Center project 1606 (ISTC#1606) is one of the liquid fueled fast reactors [6]. The original design objective of the MOSART is to effectively transmute TRU. The Molten Salt Fast Reactor (MSFR) proposed by the Centre National de la Recherche Scientifique (CNRS) is another liquid fueled fast reactor concept [7, 8]. The fuel breeding of Th-^{233}U is the main objective for the MSFR.

The MA transmutation capability is evaluated for a small power 500 MWth MSFR (named SMSFR hereinafter) in this work. In what follows, the SMSFR geometry description, molten salt parameters and the burnup calculation tool introduction are presented in Sect. 2; the results are discussed in Sect. 3; and the conclusions are given in Sect. 4.

2 SMSFR and Neutron Physic Parameters

2.1 SMSFR Core Description

Figure 1 shows the quarter vertical section of the SMSFR core [9]. The fuel salt (3.0 m^3) contains the internal fuel salt (1.5 m^3) and the external fuel salt (1.5 m^3). A radial Th fertile salt around the fuel salt is adopted for improving Th-U conversion. Both the compositions of the fuel salt and the fertile salt are listed in Table 1. The heavy nuclides (HN) comprise ^{232}Th, ^{233}U and MA in the fuel salt, while only ^{232}Th is loaded in the fertile salt. It is assumed that the MA has been partitioned from the SNF of a PWR with burnup of 33 GWd/t after 3 years of cooling. The actinide weight ratio is 56.2% for ^{237}Np, 26.4% for ^{241}Am, 12.0% for ^{243}Am, 0.03% for ^{243}Cm, 5.11% for ^{244}Cm and 0.26% for ^{245}Cm [10].

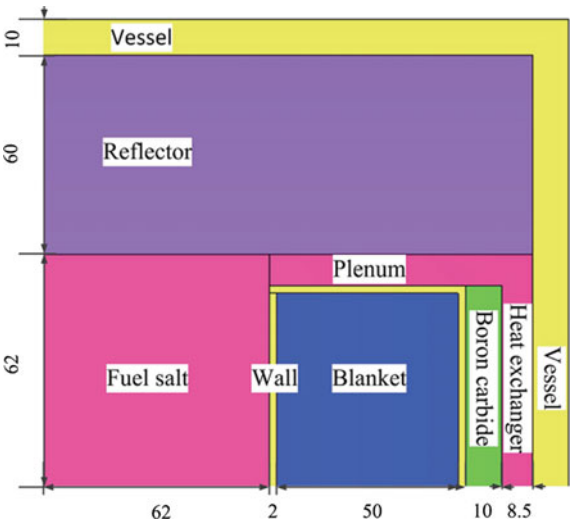

Fig. 1 Quarter vertical section of the SMSFR geometry (units: cm)

Table 1 Molten salt parameters

Parameter	Value
Compositions (mol%)	77.5 LiF-22.5 HNF_4
^{7}Li enrichment (mol%)	99.995
Melting point (°C)	565
Dilatation coefficient (g/cc/°C)	8.82E-4
Fuel salt density at 750 °C (g/cc)	4.1
Fuel salt mean temperature (°C)	750
Fuel salt volume (m^3)	3.0
Fertile salt mean temperature (°C)	700
Fertile salt volume (m^3)	2.9

2.2 Neutron Simulation Tool

All the results in this work are obtained from the calculations with SCALE6 which was developed at ORNL for reactor criticality and safety analyses [11]. To perform the burnup calculation for a two-flow MSR with online fuel reprocessing, a special MSR reprocessing sequence (MSR-RS) was developed by coupling with the CSAS6, TRITON and ORIGEN-S modules in the SCALE6 program [9, 12]. The CSAS6 module is responsible for criticality analysis. The TRITON module performs the problem-dependent cross-sectional processing followed by a multi-group neutron transport calculation. The ORIGEN-S module is used for depletion and decay calculations. The nuclear cross-sectional data used in the MSR-RS is from the ENDF/B-VII library. The flowchart of MSR-RS is shown in Fig. 2.

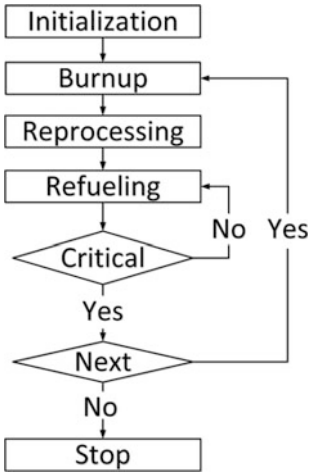

Fig. 2 MSR-RS flowchart

3 Results and Discussion

The main content in this paragraph is divided into two parts. First, the effective multiplicity factor (k_{eff}) varying with MA loadings is studied to analyze the MA transmutation performance. Second, the MA transmutation capability with online Th-U and MA refeeding into the fuel salt is studied.

3.1 k_{eff}

The mole fraction of HN in both the molten salt is set as 22.5 mol% as shown in Table 1. To keep an enough excess reactivity, 5.2 mol% ^{233}U is adopted for different MA loadings. Therefore, the 17.3 mol% of the rest fraction is composed by MA and Th.

k_{eff} is defined as the ratio of neutron production to neutron disappearance in a reactor [9]:

$$k_{\text{eff}} = \frac{R_{\text{p}}}{R_{\text{d}}} = \frac{\sum_i R_{\text{f}}(i) \cdot \bar{\nu}(i)}{\sum_j R_{\text{a}}(j) + L} \tag{1}$$

where R_{p} denotes the productive rate of neutron; R_{d} denotes the disappear rate of neutron; $R_{\text{f}}(i)$ denotes the fission rate of neutron for nuclide i; $\bar{\nu}(i)$ denotes the fission neutron number for nuclide i; $R_{\text{a}}(j)$ denotes the absorption rate of neutron for nuclide j; and L denotes the leakage rate. So, the single nuclear fission contribution to total k_{eff} can be written as

$$k_{\text{eff}}(i) = \frac{R_{\text{f}}(i) \cdot \bar{\nu}(i)}{\sum_i R_{\text{a}}(i) + L} \tag{2}$$

One can see from Eqs. (1) and (2) that the k_{eff} relates to neutron spectrum and the inventory of HN. The single HN contribution (includes ^{233}U, MA and ^{232}Th) and the total k_{eff} with different MA loading are shown in Fig. 3. And the neutron spectrum with different MA loading is shown in Fig. 4. The spectrum hardens with the increasing MA loading. The hardening spectrum leads to the reduction of the ^{233}U one-group fission cross section. And this causes the k_{eff} (^{233}U) declining with the increasing MA loading as shown in Fig. 3. Due to the increasing of MA loading, its reactivity contribution rises with the increasing MA loading. Since the fission cross section is small, the reactivity contribution of Th is slight with the increasing MA loading.

Considering the relatively large fractions of ^{237}Np and ^{241}Am in the MA, a considerable amount of Pu can be produced from the following approaches:

$$^{237}\text{Np} + \text{n} \rightarrow ^{238}\text{Np} \rightarrow \beta^- + ^{238}\text{Pu}$$
$$^{241}\text{Am} + \text{n} \rightarrow ^{242}\text{Am} \rightarrow \begin{cases} \beta^- + ^{242}\text{Cm} \rightarrow \alpha + ^{238}\text{Pu} \\ \beta^+ + ^{242}\text{Pu} \end{cases}$$

The burnup for different MA loading is shown in Fig. 5. Since there are not enough fissile nuclides in the core, the k_{eff} decreases with operating time in all cases. One can also see that as MA loading increases, the k_{eff} loss decreases with operating time. The main reason of this phenomenon is the prompt reactivity provided by MA itself and the delayed reactivity by the produced Pu isotopes.

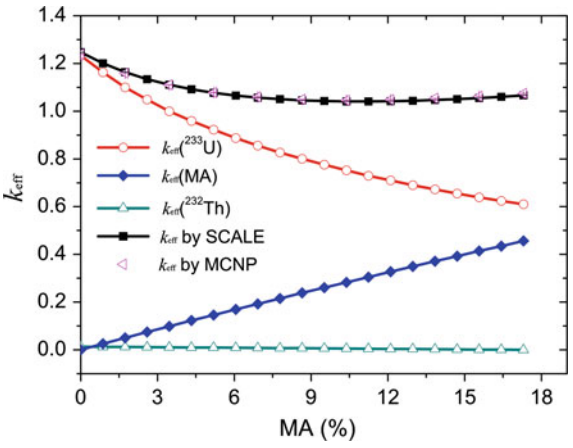

Fig. 3 k_{eff} and primary HN separate contributions

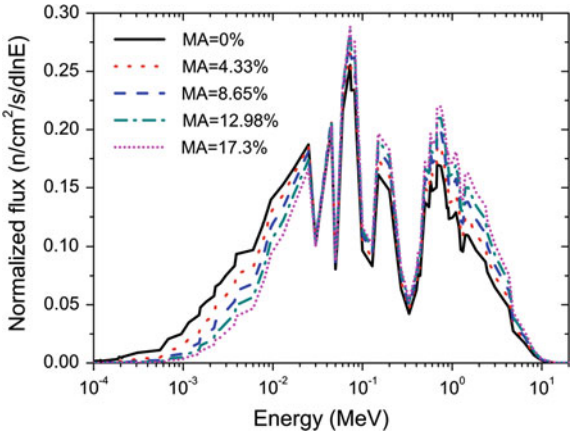

Fig. 4 Neutron spectra for different MA loadings

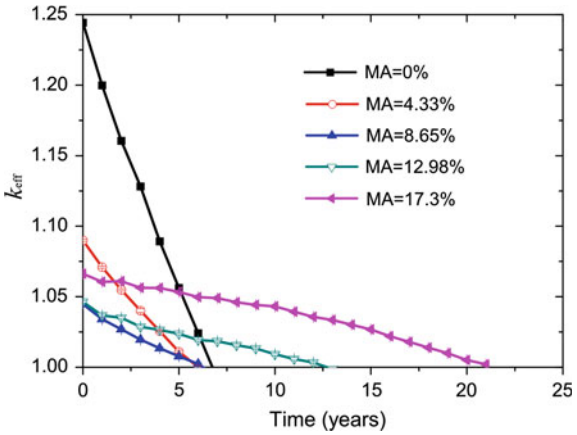

Fig. 5 Burnup performance without HN refueling for different MA loadings

When MA <10 mol%, the initial k_{eff} declines with the increasing MA loading as shown in Figs. 3 and 5. However, the effective full power year (EFPY) keeps almost constant during the operation since the k_{eff} compensation from MA and Pu. The burnup deepens significantly for MA >10 mol% since the delayed reactivity compensation from the Pu isotopes is larger than the depletion of k_{eff}.

A small reactivity at the beginning of burnup (BOB) is helpful to the SMSFR safety controlling. And a slow loss of k_{eff} during the time operating is favorable to improve the neutron economy and the MA transmutation performance. Both the above cases are desirable to transmute MA for the SMSFR. Therefore, a larger MA loading can meet the above two requirements as shown in Fig. 5.

3.2 MA Transmutation Capability

As discussed above, the k_{eff} decreases during the operating time due to the fissile fuel depletion in the fuel salt. Therefore, a HN feeding into the fuel salt case will be investigated to further deepen the burnup and improve the MA transmutation capability. As displayed in our previous work, a stable condition with $k_{\text{eff}} \approx 1$ is set by feeding HN into fuel salt. Under the small excess reactivity situation, the maximum loading of MA = 18.17 mol% is determined (slightly larger than 17.3 mol% shown in Figs. 3 and 5), corresponding to the 4.33 mol% ^{233}U loading.

A 50-year operating time of the SMSFR is analyzed considering the lifetime of the Hastelloy N alloy. In this simulation, the gaseous and non-soluble metal fission products (Xe, Kr, etc.) are online removed through the helium bubbling system with an extraction time constant of 30 s [13]. The other soluble fission products are reprocessed by the chemical reprocessing system with an extraction time constant of 180 days. To improve the utilization of ^{233}U, Pa in the fuel salt and the fertile salt is also extracted.

To evaluate the MA transmutation capability, a parameter of fractional transmutation (FT) is defined as the MA depletion to the initial MA loading [14]:

$$\text{FT}(t) = 1 - \frac{M(t)}{M_{\text{BOB}}} \tag{3}$$

where M denotes the MA inventory.

The FT with different MA loading is calculated and shown in Fig. 6. One can see from Fig. 6 that the FT increases at first and then decreases when MA <0.19 mol%. The main reason of this phenomenon is that the MA transmutation inventory is larger at first and smaller than its production inventory afterward. The MA transmutation capability increases during the first 30 years and then becomes stable for MA = 0.19 mol%. When MA loading is greater than 0.94 mol %, about 95% of the MA transmutation efficiency can be achieved during 50-year operating.

Although the FT is about 95% at the condition of MA >0.94 mol%, their respective net transmutation mass is different significantly. The net transmutation mass is 292 kg for MA = 0.94 mol%, while it is 5620 kg for MA = 18.17 mol%. The MA transmutation mass of the latter is 19 times greater than that of the former. Therefore, FT is insufficient to draw a definite conclusion for MA transmutation capability evaluation.

The incinerated MA relative to the total depleted HN is also an important factor for evaluating MA transmutation capability, which is defined as:

$$\text{DR} = \frac{\text{MA(depletion)}}{\text{HN(depletion)}} \tag{4}$$

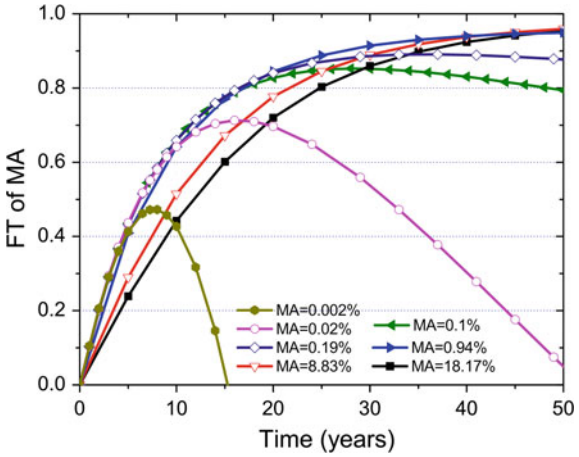

Fig. 6 MA transmutation fraction during 50 years operating

where MA(depletion) and HN(depletion) represent the incinerated MA and the depleted HN, respectively.

The time evolution of DR for MA = 18.17 mol% is shown in Fig. 7. The DR increases gradually during the first 20 years due to the considerable production of the Pu isotopes from MA and then tends to decline. It indicates that after 20 years of operation, the MA left in the fuel salt is not sufficient to be burned, which results in the decreases of both the inventory of Pu and the DR. One can also find that the DR is smaller than 0.5 after about 45 years, which implies more ^{233}U and ^{232}Th are consumed than MA. Therefore, MA is continually injected into the fuel salt to maintain the constant amount of total TRU inventory during the whole operation. In this case, the transmutation amount of MA at 50 years is about 8030 kg, improving by 42.9% than the no MA feeding condition. Moreover, the DR for the MA feeding condition increases from 0.435 at BOB to 0.775 at 50-year without any decline phenomenon.

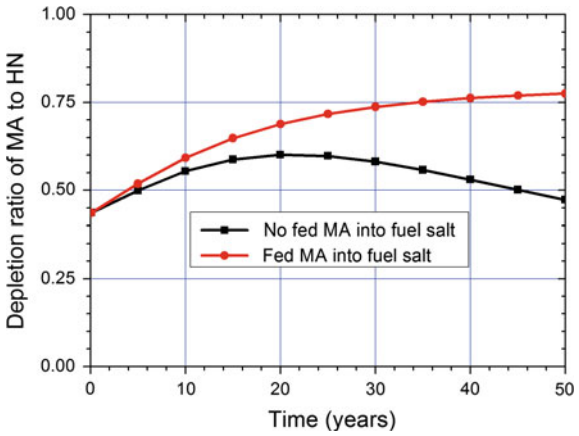

Fig. 7 Time evolutions of depletion ratio of MA to HN for different MA transmutation scenarios at MA = 18.17%

4 Conclusions

A 500 MWth MSFR is used for evaluating the MA transmutation performance. When maintaining the amount of ^{233}U at 5.2 mol%, the decreasing of k_{eff} (^{233}U) slows down, while the k_{eff} (MA) increases linearly with the increasing MA loading. The combination of the above two contributions makes the k_{eff} firstly reduce and then increase with the increasing MA loading. As the MA loading increases, the k_{eff} loss decreases with operating time due to the delayed reactivity compensation of Pu isotopes. And the low initial k_{eff} and deepen burnup are favorable to the reactor safety controlling and MA transmutation.

The online refueling of Th, ^{233}U and MA into the fuel salt are also investigated to evaluate the MA transmutation performance. A larger MA loading is helpful to improve the MA transmutation capability. When MA = 18.17 mol%, the fractional transmutation is 95% corresponding to the 5620 kg MA transmutation mass for no MA online feeding. The MA transmutation amount and the DR are 8030 kg and 77.5%, respectively, when MA is fed into fuel salt to maintain the total TRU inventory constant during the whole operation for MA = 18.17 mol%.

The initial fuel temperature coefficient of SMSFR is −3.8 pcm/K for MA = 18.17%, and it does not change notably during the whole operation.

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