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To cite this article: O Ashraf and G V Tikhomirov 2020 *J. Phys.: Conf. Ser.* **1689** 012027

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Test task for transmutation of ^{241}Am in molten salt reactors

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Abstract. Molten Salt Reactor (MSR) is one of the six innovative systems, which has been selected by the international Generation IV forum as a candidate for the next-generation reactors. In this paper, the transmutation of ^{241}Am , the main contributor to the waste toxicity in the nuclear fuel cycle, in both thermal- and fast-spectrum molten salt reactors is compared. The *Am-cylinder* is irradiated in the center of the thermal (SD-TMSR) and fast (SMSFR) spectrum MSRs, respectively, and during a long period of time. The total neutron fluxes in SD-TMSR and SMSFR are 4.1×10^{14} and 1.8×10^{15} n.cm⁻².s⁻¹, respectively. The overall change in actinides and fission products (FPs) mass during the irradiation has been investigated using direct Serpent2 calculations. Under fast spectrum irradiation, transmutation leads to produce short-lived FPs (by fission) rather than the accumulation of heavier actinides. The ratio between the maximum and initial released energy from all isotopes in the *Am-cylinder* has been calculated in this paper.

1. Introduction

Molten Salt Reactor (MSR) is one of the six innovative systems, which has been selected by the international Generation IV forum [1] as a candidate for the next generation reactors. MSR is a very promising concept especially for the Th/U3 fuel cycle which allows nuclear energy production with a relatively low production of radiotoxic minor actinides.

Transmutation of long-lived minor actinides (MAs): Np, Am, and Cm eliminates a very long-term radioactive hazard and convert it to a much shorter-term one [2]. ^{241}Am is the major contributor to the radioactive hazard of the spent fuel. Its transmutation into less radiotoxic isotopes is considered in thermal and fast nuclear reactors [3], [4]. Additionally, accelerator-driven system is proposed to transmute MAs [5]. The thermal neutron spectrum is appropriate to effectively transmute MA due to high capture cross sections in thermal energy regions [2]. However, using thermal neutron spectrum leads to the accumulation of heavier actinides, e.g., Cm and Cf. In contrast, in the fast neutron spectrum, the transmutation of MAs mainly by fission and the transmutation to heavier actinides is relatively small [6].

In this study both thermal- and fast-spectrum molten salt reactors have been compared for the transmutation of the main contributor to the radioactive hazard of the spent fuel (i.e., ^{241}Am). The overall change in actinides and fission products (FPs) mass during the irradiation has been investigated using direct Serpent2 calculations.



2. Model description

The SD-TMSR is a graphite-moderated thermal-spectrum MSR operating in Th/U3 fuel cycle. The core structure of the Single-fluid Double-zone Thorium-based Molten Salt Reactor (SD-TMSR-2,250 MW_{th}) is inspired by the Molten Salt Breeder Reactor (MSBR) [7] and the TMSR [8]. Figure 1 illustrates the whole-core configuration of the SD-TMSR. More details about the description of the SD-TMSR can be found in our recently published papers [9], [10].

The Small Molten Salt Fast Reactor (SMSFR-500 MW_{th}) is a free-moderator fast-spectrum MSR that has a similar core configuration to MSFR [11] but with a smaller core volume. The SMSFR is also designed to operate based on the Th/U3 fuel cycle. Figure 2 demonstrates the whole-core configuration of the SMSFR. More details about the description of the SMSFR can be found in [12].

In the current work, we simulated a cylinder with a radius of 5 cm and a volume of 83.33 cm³. This cylinder is filled with pure ²⁴¹Am with a density of 12 g/cm³. The total mass of ²⁴¹Am in the cylinder is 1000 g. The objective of the current study is to compare the transmutation of ²⁴¹Am in both thermal- and fast-spectrum molten salt reactors. Therefore, the *Am-cylinder* was located in the center of the SD-TMSR and SMSFR, respectively. The power density for SD-TMSR and SMSFR is 0.023 KW/gHM. The total neutron fluxes in SD-TMSR and SMSFR are 4.1×10¹⁴ and 1.8×10¹⁵ n.cm⁻².s⁻¹, respectively. The flux per unit lethargy at Beginning-of-life (BOL) for both SD-TMSR and SMSFR are illustrated in figures 3&4, respectively.

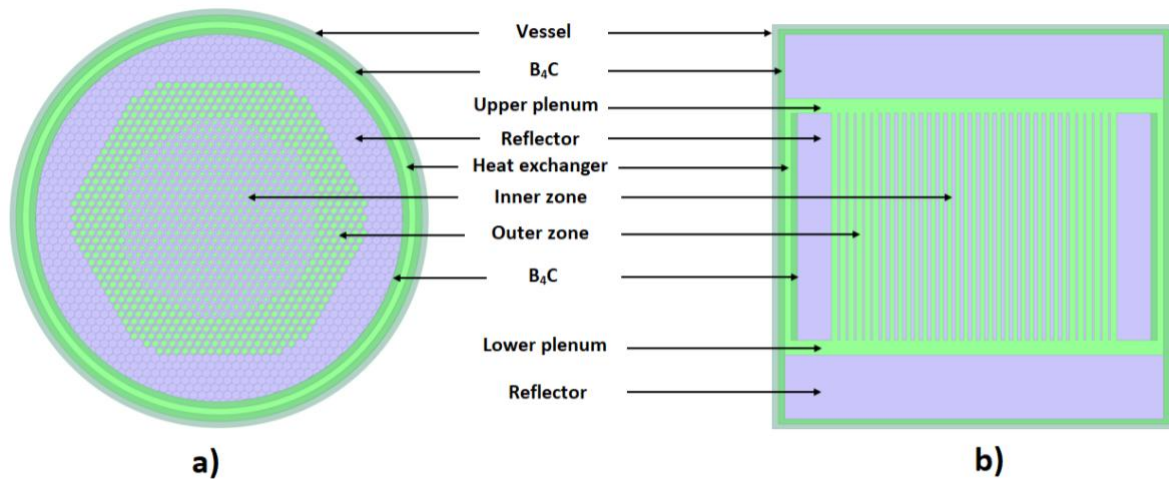


Figure 1: XY (a) and XZ (b) sections of the core model of the SD-TMSR.

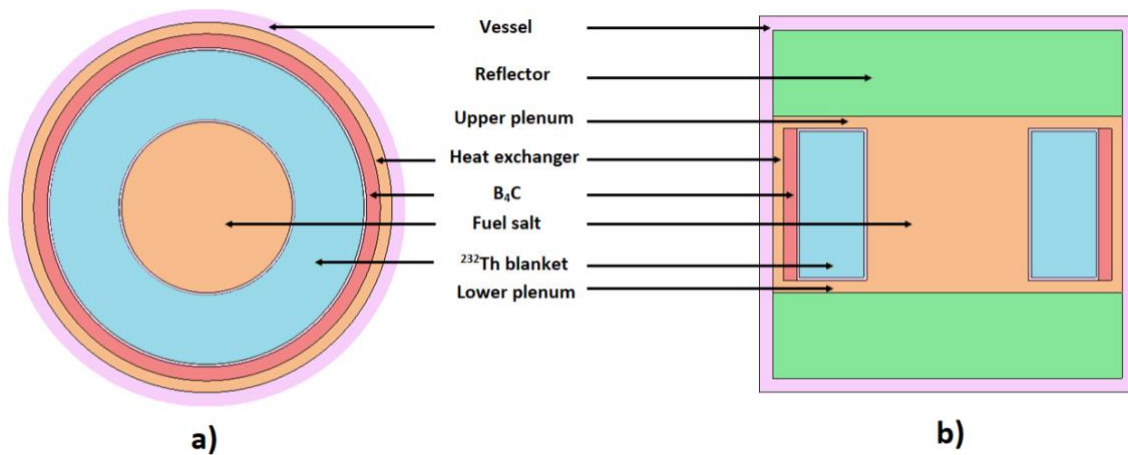


Figure 2: XY (a) and XZ (b) sections of the core model of the SMSFR.

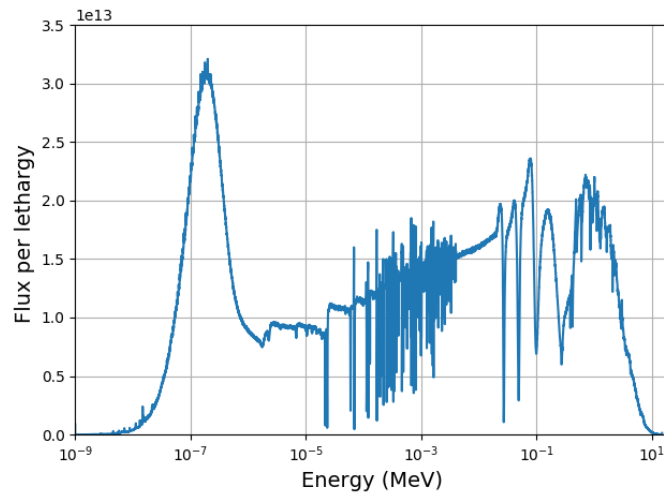


Figure 3: The neutron flux energy spectrum at BOL for SD-TMSR.

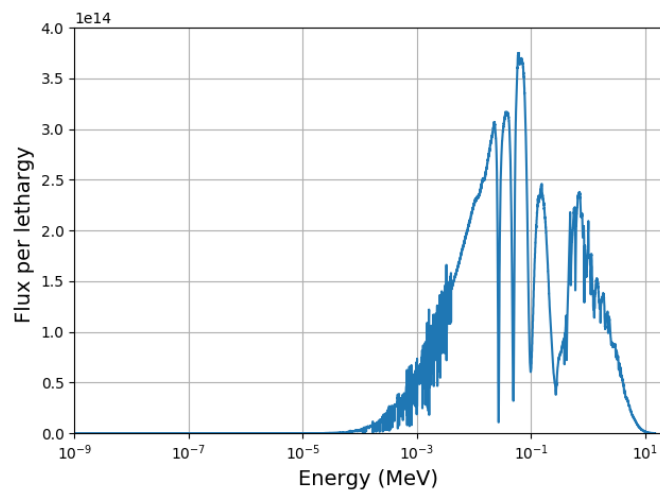


Figure 4: The neutron flux energy spectrum at BOL for SMSFR.

3. Methodology and tools

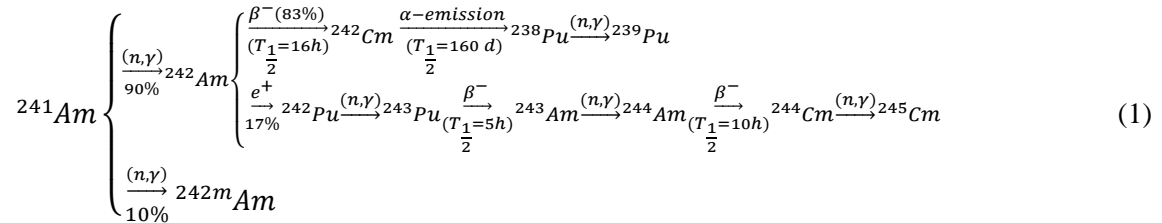
The Serpent2 [13] is a three-dimensional continuous energy Monte Carlo neutron transport and burn-up code. The Serpent2 has been extended to model the online reprocessing and refueling in MSRs [14]. In the present work, Serpent2 has been utilized for the simulation of the full-core model of SD-TMSR and SMSFR with online reprocessing and refueling. The overall change in actinides and fission products (FPs) mass during the irradiation has been investigated using direct Serpent2 calculations. The results were obtained after whole-core runs of 12.5 million neutron history and 50 inactive cycles per burn-up step for the convergence of the fission source distribution. The statistical error in multiplication factor ≤ 36 pcm. During the burnup step, the core was maintained critical ($K_{\text{eff}} \approx 1.003$) and the total fuel mass in the core and blanket was almost constant ($dm \leq 0.1\%$).

4. Results and discussion

4.1. Irradiation of ^{241}Am

The main reaction of the ^{241}Am in both thermal- and fast-spectrum is radiative capture (n,γ). However, the capture to fission ratio is only 3 in the fast spectrum for ^{241}Am . Thus, there is more competition

between capture and fission in the fast spectrum [15]. Equation 1 shows the possible reaction paths for ^{241}Am under thermal neutron irradiation.



The disappearance of ^{241}Am can be done by three basic reactions; capture, fission, and decay. According to Equation 1, ^{241}Am undergoes radiative capture to produce ^{242}Am with a 90% branching ratio and ^{242m}Am with a 10% branching ratio under thermal neutron capture. However, under fast neutron capture, the branching ratios are 85% for ^{242}Am and 15% for ^{242m}Am . ^{242m}Am transmutes mainly by nuclear fission. In contrast, ^{242}Am ($T_{1/2} = 16\text{ h}$) has two decay modes: β -decay to produce ^{242}Cm (83% branching ratio) and e^{+} (electron capture) to produce ^{242}Pu (17% branching ratio), as shown in Equation 1. ^{242}Cm decays, with a half-life of 160 days to ^{238}Pu by α -emission. By radiative capture, ^{238}Pu turns into ^{239}Pu then this fissile isotope undergoes fission. ^{242}Pu , by radiative capture, turns into ^{243}Pu which then decays, with a half-life of 5 h, to ^{243}Am . By decay and capture, ^{243}Am gives rise to ^{244}Am , ^{244}Cm and ^{245}Cm . Finally, ^{245}Cm is removed predominantly by fission.

4.2. Evolution of total actinide mass

To compare the transmutation efficiency of ^{241}Am in both thermal- and fast-spectrum molten salt reactors, Serpent2 calculations have been made. The *Am-cylinder* was irradiated in the center of the thermal (SD-TMSR) and fast (SMSFR) spectrum MSRs, respectively, and during a long period of time. The total neutron fluxes in SD-TMSR and SMSFR were 4.1×10^{14} and 1.8×10^{15} n.cm⁻².s⁻¹, respectively.

Figure 5 shows the time evolution of the actinides masses for SD-TMSR and SMSFR. In figure 5, the total amount of actinides is decreasing sharply in the thermal spectrum compared to the fast spectrum. Under fast spectrum irradiation, about 50% of the actinides are fissioned after about 2350 days (i.e., 6.5 years); however, the same percent of the actinides disappears after about 2000 days (5.5 years) under irradiation in SD-TMSR. The disappearance of the actinides reaches 99.3% after around 9000 days (less than 25 years). Under fast spectrum irradiation, the disappearance of the actinides reaches 88% after the same period (9000 days). The transmutation rate of ^{241}Am is higher in SD-TMSR than in SMSFR.

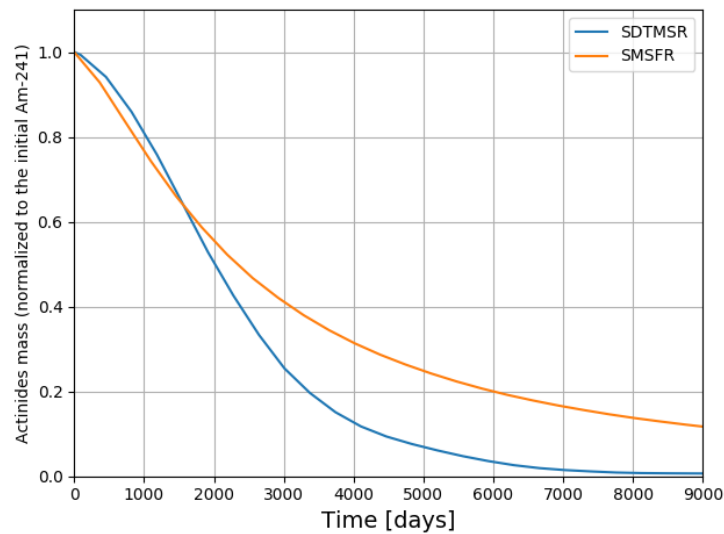


Figure 5: Time evolution of the actinides masses for SD-TMSR and SMSFR.

4.3. Evolution of major isotopes in the Am-cylinder

Figure 6 illustrates the time evolution of the ^{241}Am mass in the *Am-cylinder* for SD-TMSR and SMSFR. Under thermal spectrum irradiation, about 98.5% of ^{241}Am mass in the *Am-cylinder* is transmuted to other minor actinides and fission products, however, under fast spectrum irradiation, the transmutation rate is much lower (89.8%). This is because of the higher microscopic cross-section in the thermal region compared to the fast neutron region.

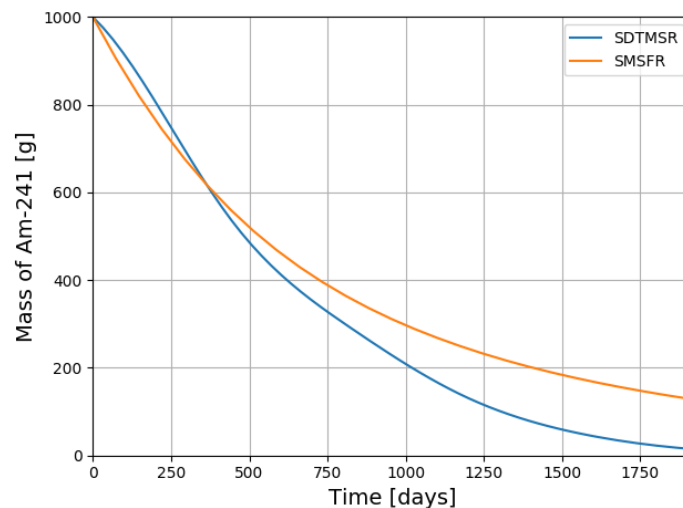


Figure 6: Time evolution of the ^{241}Am mass in the *Am-cylinder* for SD-TMSR and SMSFR.

As shown in figure 7, the fission products (FPs) accumulated in the *Am-cylinder* as a result of capture, fission, and decay of ^{241}Am in both thermal and fast spectrum MSRs. Results show that the accumulation of FPs in SMSFR is higher than in SD-TMSR; in the fast neutron spectrum, the transmutation of MAs mainly by fission and the transmutation to heavier actinides is relatively small [6]. After about 1500 days, the mass of FPs in SD-TMSR overrides the FPs mass in SMSFR, this because the reaction rates in the SMSFR decrease during operation.

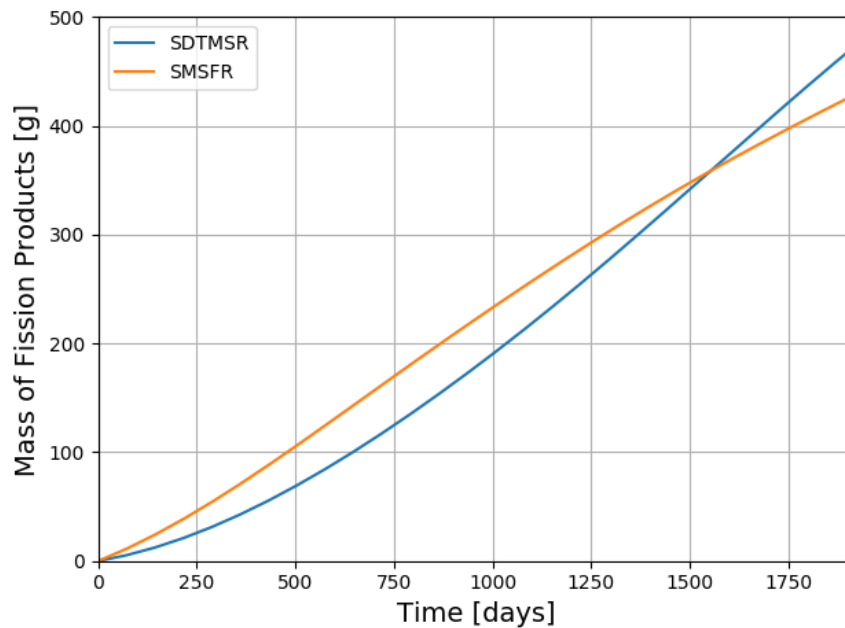


Figure 7: Build-up of FPs in the *Am-cylinder* for SD-TMSR and SMSFR.

Figure 8 shows the time evolution of the ^{241}Am and its major reaction products. As shown in figure 8, the mass of ^{241}Am decreases sharply. The decay of ^{242}Am resulting in a build-up of ^{242}Cm with a maximum mass of 16% of initial ^{241}Am at 455 days. ^{238}Pu , a decay product of ^{242}Cm , accumulates during irradiation. The maximum mass of ^{238}Pu reaches 31% of initial ^{241}Am at 1185 days. ^{242}Pu , a decay product of ^{242}Am , is also accumulate quickly during operation. As shown in figure 8, ^{242}Pu and ^{244}Cm take relatively long time to disappear. Its burnup time is quite long (5200 & 7000 days). ^{239}Pu , produced by capture of ^{238}Pu , accumulates with a relatively small amount then this fissile isotope undergoes fission. ^{245}Cm and ^{246}Cm accumulate quite slowly and will be the last isotopes to disappear.

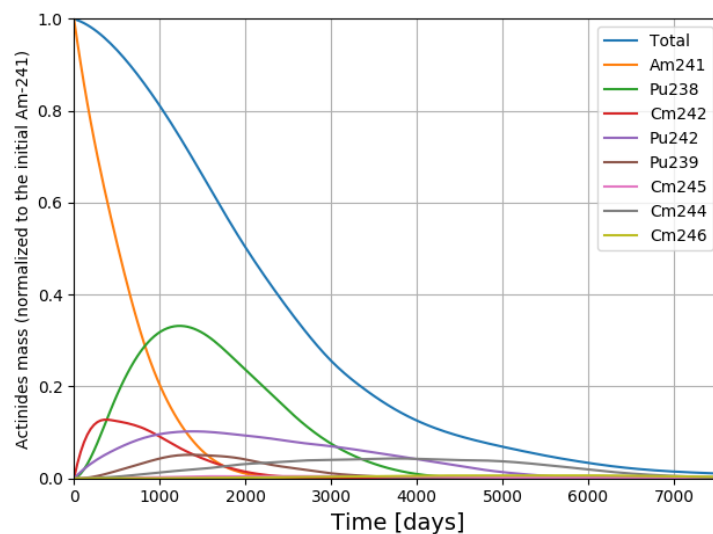


Figure 8: Time evolution of the ^{241}Am and its main reaction products for SD-TMSR.

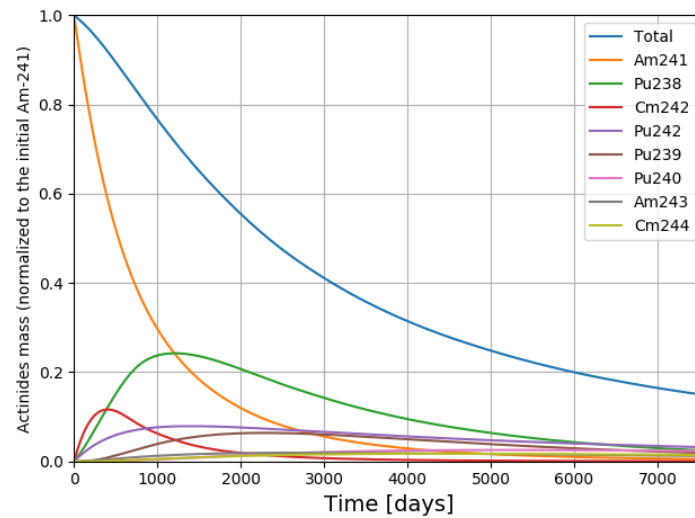


Figure 9: Time evolution of the ^{241}Am and its main reaction products for SMSFR.

Figure 9 illustrates the time evolution of ^{241}Am and its main reaction products in the *Am-cylinder* for SMSFR. As shown in figure 9, the mass of ^{241}Am decreases slowly compared to the case in figure 8. The main result here is the actinide mass is relatively small compared to this in Fig 8. Thus, under fast spectrum irradiation, transmutation leads to produce short-lived FPs (by fission) rather than the accumulation of heavier actinides. Notably, all Pu isotopes take relatively long burnup time to disappear, since the spectrum is fast.

The total decay heat (Watt) released from all isotopes in the *Am-cylinder* can be calculated from the following Equation:

$$q(t) = \sum_{i=1}^n E_i \lambda_i N_i(t) \quad (2)$$

where n is the number of isotopes in the *Am-cylinder*. E_i is the decay energy (MeV), λ_i is the decay constant (s^{-1}), and $N_i(t)$ is the number of atoms of isotope i in time t .

Figure 10 shows the total decay heat versus time. Results showed that the initial decay heat released from *Am-cylinder* (due to the 1000 g ^{241}Am) is about 115 Watt. Then the decay heat increases with time due to the production of heavier actinides and FPs, which have high decay heat. For example, ^{242}Cm , ^{242}Am , ^{104}Tc , ^{134}I , ^{138}Cs , ^{99}Mo , ^{103}Mo , ^{144}La , ^{241}Am , and ^{103}Ru . The ratio between the maximum and initial released energy form *Am-cylinder* is about 230 and 240 for SD-TMSR and SMSFR, respectively. This may reflect the possibility to use such an *Am-cylinder* as a heat source for a radioisotope thermoelectric generator (RTG, RITEG). Transmutation and burnup of MAs and FPs decrease their masses; consequently, decrease the released decay heat as shown in figure 10 after about 600 days.

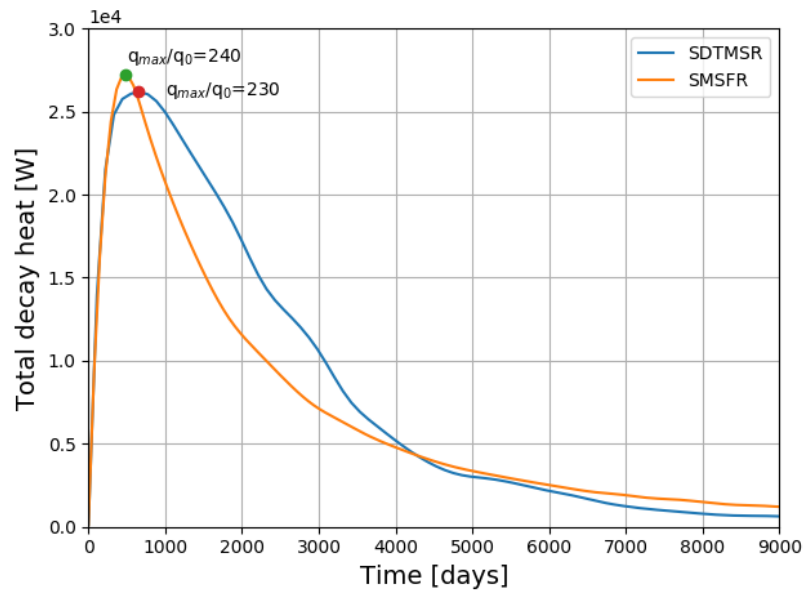


Figure 10: Total decay heat from all isotopes in the *Am-cylinder* for SD-TMSR and SMSFR.

5. Conclusion

In the current work, we compared the transmutation of ^{241}Am in both thermal- and fast-spectrum molten salt reactors. An *Am-cylinder* was located in the center of the SD-TMSR and SMSFR, respectively. The total neutron fluxes in SD-TMSR and SMSFR are 4.1×10^{14} and 1.8×10^{15} $\text{n.cm}^{-2}.\text{s}^{-1}$, respectively. The flux per unit lethargy at Beginning-of-life (BOL) for both SD-TMSR and SMSFR has been calculated. The overall change in actinides and fission products (FPs) mass during the irradiation has been investigated using direct Serpent2 calculations. Results showed that the overall disappearance rates of ^{241}Am are higher in SD-TMSR than in SMSFR due to the high capture cross sections in thermal energy regions. However, the accumulation of FPs in SMSFR is higher than in SD-TMSR; in the fast neutron spectrum, the transmutation of MAs mainly by fission and the transmutation to heavier actinides is relatively small. Thus, under fast spectrum irradiation, transmutation leads to produce short-lived FPs (by fission) rather than the accumulation of heavier actinides. Notably, all Pu isotopes in this case take relatively long burnup time to disappear, since the spectrum is fast. The ratio between the maximum and initial released energy from *Am-cylinder* is about 230 and 240 for SD-TMSR and SMSFR, respectively. This may reflect the possibility to use such an *Am-cylinder* as a heat source for a radioisotope thermoelectric generator (RTG, RITEG).

Conflict of interest

The authors declare no conflict of interest.

Acknowledgments

Osama Ashraf would like to thank the Egyptian Ministry of Higher Education (MoHE), as well as MEPhI's Competitiveness Program for providing financial support for this research, as well as Andrei Rykhlevskii for his valuable suggestions and comments. The facility and tools needed to conduct this work were supported by MEPhI.

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