

Breakeven Feasibility Study for Chlorine-based Molten Salt Fast Reactor

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1. Introduction

As nuclear power is considered one of the solutions to the issue of climate change, nuclear reactors have been under development and improvement for decades to enhance efficiency, safety, and economy. The Molten Salt Reactor (MSR) is one such advanced reactor currently being developed. It utilizes molten salt as both fuel and coolant and is garnering attention due to several safety advantages, including the prevention of severe accidents, stable operation at atmospheric pressure, strong negative feedback from thermal expansion, and a convenient residual heat removal system [1].

In the pursuit of achieving high levels of efficiency, numerous efforts have been dedicated to developing long-lived and high-burnup nuclear systems. Additionally, advanced reactor designs, such as the Molten Salt Reactor (MSR), have emerged with the aim of addressing challenges, including the management of spent nuclear fuel, which significantly impacts the social acceptance of nuclear energy. To address this issue, the concept of a breakeven reactor, which utilizes spent nuclear fuel to sustain criticality, can be envisioned as a potential solution. This study aims to assess the feasibility of achieving such breakeven conditions in an MSFR (Molten Salt Fast Reactor) system.

2. Description of Fuel and Model

The meaning of breakeven in this paper refers to achieving self-supplementation during fuel depletion by establishing an equilibrium between the gain and loss of fissile material. This equilibrium can be influenced by the fuel conversion ratio. To attain a sufficient fuel conversion ratio, a high value of the reproduction factor (η) must be ensured. Figure 1 compares the reproduction factors of three fissile materials: U-233, U-235, and Pu-239. It is evident that Pu-239 has the highest reproduction factor in the fast neutron spectrum with energy levels exceeding 100 keV [2]. This is precisely why the fast neutron spectrum and the U-Pu cycle are employed in this study.

Prior research has indicated that fuels composed of fluoride-based salts are inadequate for achieving breakeven due to their inability to produce a sufficiently hard neutron spectrum [3]. As a result, this study employs chloride-based molten salt as the fuel.

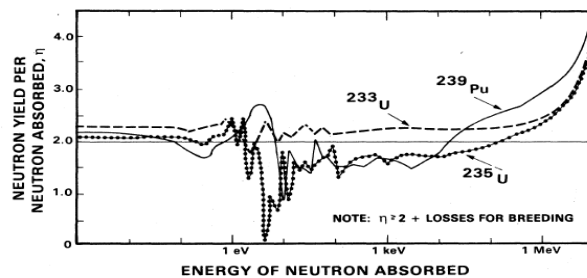


Fig 1. Reproduction factor vs energy of neutron [2]

To ensure a sufficient conversion ratio for achieving breakeven conditions, a combination of Transuranic (TRU) elements and natural uranium has been employed as the fuel at the beginning of the operation. In this mixture, TRU elements play the crucial role of initiating the fission chain reaction, while uranium (U) functions as the primary fertile material responsible for generating plutonium-239. The compositions of transuranic elements extracted from the spent fuel of pressurized water reactors (PWRs) are presented in Table II. The table assumes an ideal composition of TRU with the exclusion of rare earth elements. Notably, the composition contains approximately 85% plutonium.

Table I. Ideal TRU composition extracted from spent fuel of PWR (unit: wt.%) [4]

Ac	1.02E-09	Np	6.131	Cm	0.716
Th	7.40E-05	Pu	85.383	Bk	4.28E-11
Pa	5.92E-06	Am	7.770	Cf	1.92E-07

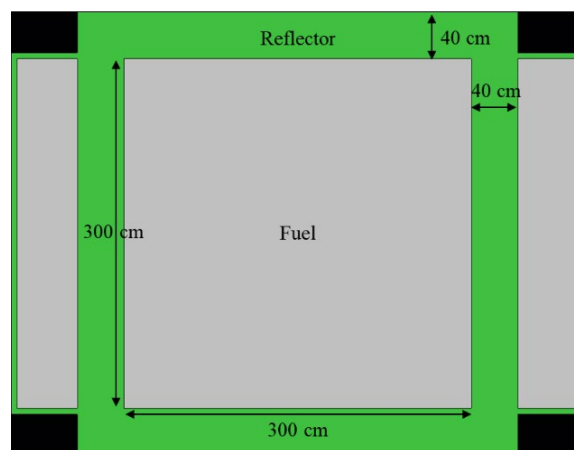


Fig 2. Cylindrical reactor model

The target reactor is composed of distinct elements: an active core, an inactive core, reflectors, and various other components. The active core is cylindrical in shape, with a diameter and height of the same size 300 cm. The inactive core is ring-shaped, enveloping the active core, and separated by a reflector placed in between. Both the active and inactive cores are fuel-filled. It is assumed that the active and inactive cores have identical volumes. The reflector, composed of stainless steel, possesses a thickness of 40 cm. To safeguard against the high-temperature molten salt, the inner surface of the reflector is coated with a 0.1 cm layer of Hastelloy-N. A comprehensive depiction of the reactor's entire design can be observed in Fig. 2.

The eutectic composition of KCl- UCl_3 exhibits exceptional characteristics for molten salt fuel, characterized by its low melting temperatures and elevated uranium concentrations. Detailed information regarding these properties is outlined in Table II. For the achievement of a high conversion ratio, the reactor employs a mixture of TRU and U, resulting in the use of KCl- $TRUCl_3$ - UCl_3 as the reactor's fuel. The compositions of the non-fuel salts are derived from the eutectic composition of KCl- UCl_3 . The cumulative proportion of $TRUCl_3$ and UCl_3 in the overall fuel composition is about 54%. The specific composition of $TRUCl_3$ and UCl_3 has been adjusted to achieve the initial criticality required for the reactor. The fuel composition and the inventory of heavy metals are enumerated in Table III. It's important to note that UCl_3 employs natural uranium as its initial source material.

Table II. Eutectic data of KCl- UCl_3 [5]

	KCl- UCl_3
Molar composition	46-54
Melting temperature	558°C
U density at 650°C	2.179 g/cm ³

Table III. Composition of fuel and heavy metal inventory of KCl- $TRUCl_3$ - UCl_3

	KCl- $TRUCl_3$ - UCl_3
Mole fraction	46.0-7.6-46.4
U mass	79,598 kg
Pu + MA mass	13,132 kg

3. Numerical Results

To assess the feasibility of achieving breakeven in Molten Salt Fast Reactors (MSFRs), fuel depletion calculations were conducted using the Monte-Carlo-based reactor analysis code Serpent 2.2.0. The calculations employed the ENDF-B/VII.1 nuclear library as a reference. For the history of a single cycle, a sample size of 50,000 was utilized, with 50 samples for the inactive cycle and 200 samples for the active cycle. The reactor's full-power operational capacity was set at 1,680 MWth.

One of the primary objectives in studying breakeven MSFRs is to achieve semi-permanent operation. However, even if initial breakeven is attained, the reactor cannot maintain criticality without undergoing fuel processing during operation. This is due to the depletion of fertile materials and the accumulation of fission products over the course of operation. As a result, usage of fuel processing is essential for achieving long-term breakeven.

The fuel processing employed in this study encompasses the removal of fission products, refueling, elimination of activation products, and replenishment of non-fuel materials. Fission products are categorized into noble gases, noble metals, and soluble fission products. Noble gases, which are gaseous and insoluble fission products, can escape from the fuel. Noble metals refer to fission products insoluble in the molten salt. Other fission products are classified as soluble fission products.

In the context of this study, it is assumed that 100% of gaseous fission products are extracted from the core, while noble metals and soluble fission products are removed in fractions when fuel processing is applied. When the fission products are removed from the reactor, spent nuclear fuel from a PWR is newly added to the core. In this study, to acquire the composition of spent nuclear fuel, the depletion calculation of a lattice of APR1400 with a burnup of 50 MWth/kgU followed by a 10-year cooling period has been performed. The obtained composition is enumerated in Table IV, where the primary isotope in the supplement is U-238. Note that the number of actinide atoms refueled is half the number of fission products removed, as two fission products originate from one fission event.

Furthermore, accumulation of activation products resulting from interactions between neutron and non-fuel components, such as Ar-38 which is produced through neutron capture by Cl-37, can occur. Such activation products need to be suitably extracted from the core, and an equivalent amount of non-fuel materials (K and Cl) should be replenished for compensation. Depending on the removal fractions for noble metals, soluble fission products, and activation products, four distinct fuel processing strategies have been considered in this study as shown in Table V. Note that the amount of provided spent fuel is determined accordingly to the amount of fission products being removed.

Table IV. Composition of spent fuel for supplement

Isotope	Mole fraction	Isotope	Mole fraction
U234	0.00292%	Pu244	0.00003%
U235	0.05035%	Am241	0.06407%
U236	0.23419%	Am242m	0.00004%
U238	98.40118%	Am243	0.04643%
Np237	0.04623%	Cm243	0.00009%
Pu238	0.03229%	Cm244	0.02296%
Pu239	0.48859%	Cm245	0.00229%
Pu240	0.33611%	Cm246	0.00063%
Pu241	0.09810%	Cm247	0.00001%
Pu242	0.17350%		

Table V. Schemes of processing of the fuel

Removal fraction of	Case A	Case B	Case C	Case D
Noble gas	100%	100%	100%	100%
Noble metal	100%	75%	50%	25%
Soluble FPs	50%	35%	20%	10%
Activation products	50%	35%	20%	10%

For the preliminary test, batch fuel treatment based on fuel processing strategies A, B, and C was applied to the reactor every 10 years. The calculated evolutions of reactivity and conversion ratio for each case with an operation period of 50 years are illustrated in Figs. 3 and 4. After the onset of fuel processing, the reactivity is increased in a step-wise manner and gradually dwindles throughout the operation. Particularly, the extent of reactivity decrement during the operation between two consecutive fuel processing events becomes more significant as burnup increases. Regarding the conversion ratio, it exhibits an upward surge after each process and subsequently diminishes within a single cycle. As the cycles progress, these values gradually approach an equilibrium state, although they do not yet ensure the normal operation of the reactor.

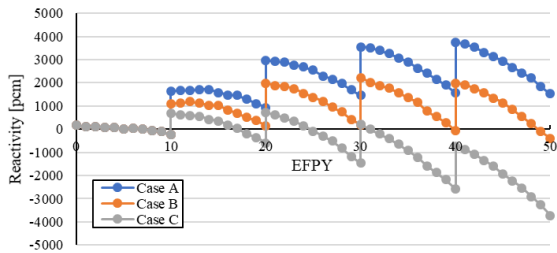


Fig. 3. Reactivities vs full-power operation time for Cases A, B, and C

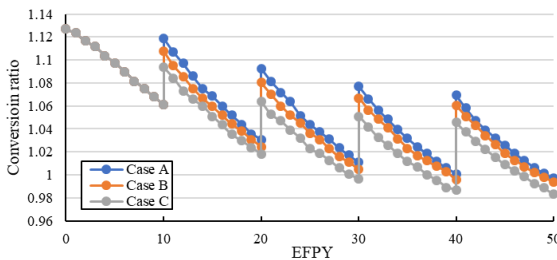


Fig. 4. Conversion ratios vs full-power operation time for Cases A, B, and C

Since the results of the preliminary tests cannot guarantee normal operation in terms of reactivity, the treatment scenario has been adjusted to maintain reactivity within the range of 0 to 1,000 pcm with an operation period of 40 years. In total of 10 fuel processing have been considered throughout the operation, where each processing scheme is summarized in Table VI.

Table VI. Adjusted scheme of processing of the fuel

Number	Scheme	Interval year	Elapsed year
1	Case C	6	6
2	Case D	8	14
3	Case D	4	18
4	Case D	3	21
5	Case D	3	24
6	Case D	3	27
7	Case D	3	30
8	Case D	3	33
9	Case D	3	36
10	Case D	2	38

The calculated results can be found in Figs. 5 and 6, where the former indicates the reactivity, and the latter depicts the conversion ratio throughout the operational period. The overall behavior of these figures is similar to that observed in the preliminary test results, where fuel processing strategy D is constantly required with a time interval of 2-3 years. The calculated reactivity ranges between 0 and 800 pcm asymptotically, while the conversion ratio ranges between 1.02 and 1.04.

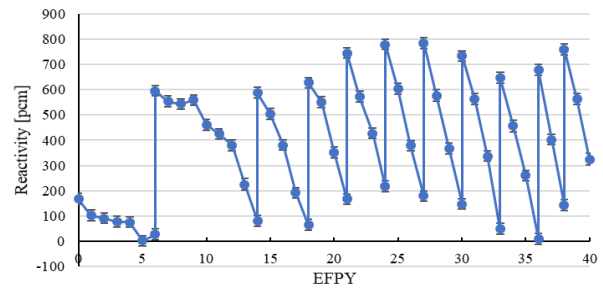


Fig. 5. Reactivity vs full-power operation time with adjusted fuel processing

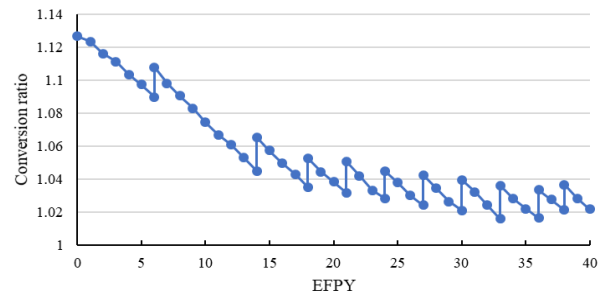


Fig. 6. Conversion ratio vs full-power operation time with adjusted fuel processing

Figure 7 illustrates the number densities of U-238 and Pu-239, where the former represents primary fertile material and the latter represents main fissile material. Throughout each cycle, U-238 is consumed and replenished, and Pu-239 accumulates over the course of operation due to the higher supplement from the fuel conversion compared to the consumption occurring through nuclear fission events. Asymptotically, the accumulation process reaches equilibrium as production and consumption balance out. Figure 8 visually depicts

the ratio of U-238 to Pu-239, which effectively represents the fertile-to-fissile ratio, as U-238 serves as the main fertile material and Pu-239 serves as the main fissile material. Such ratio decreases during operation, except for having U-238 supplementation, which accounts for the diminishing conversion ratios as the cycles progress.

Figure 9 indicates the number densities of Cs-135 and Ru-102. The former represents the behavior of the soluble fission products, and the latter represents the behavior of the noble metal. The behavior of fission products can elucidate the fluctuations in reactivity observed. In the initial two cycles, a modest swing in reactivity could be observed. This is due to the combined accumulation of Pu-239 and fission products, with their opposing effects on reactivity largely offsetting each other. However, during later cycles, the reactivity within the cycle rapidly diminishes. This phenomenon arises from the fact that while Pu-239 accumulation ceases, fission product accumulation persists. The general behaviors of fissile, fertile, and fission product reaches stationary and they affect the asymptotic behaviors of the reactivity and conversion ratio.

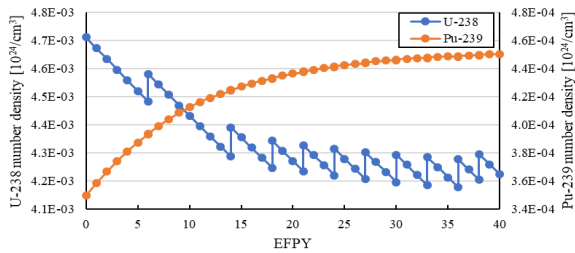


Fig. 7. Number densities of Pu-239 and U-238 vs full-power operation time with adjusted fuel processing

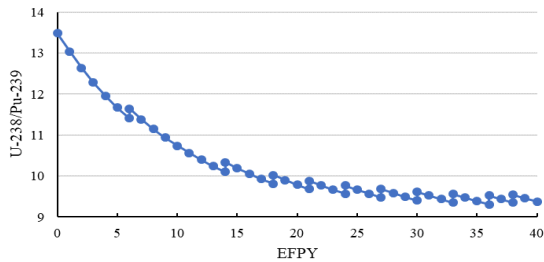


Fig. 8. Ratio of number densities of U-238 and Pu-239 vs full-power operation time with adjusted fuel processing

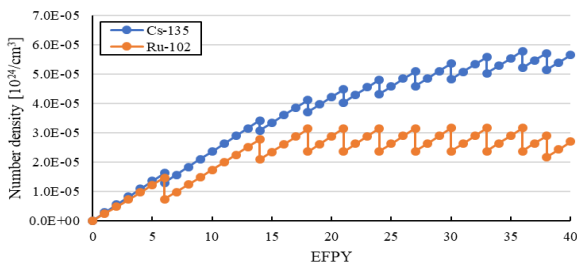


Fig. 9. Number densities of Cs-135 and Ru-102 vs full-power operation time with adjusted fuel processing

4. Conclusions

This study has observed the impact of fission product removal and refueling processes, highlighting the establishment of equilibrium between the production and consumption of Pu-239. The results indicate that achieving breakeven is possible by removing fission products and resupplying fuel every 2-3 years using the treatment method described as Case D, which involves removing the complete fraction of noble gases, a 20% fraction of noble metals, and 10% of soluble fission products and activation products. However, further research into optimization for enhanced economic feasibility and the development of reactor core operation strategies are needed.

The optimization process may include adjustments to the core dimensions or modifications to the strategy for fission product removal and refueling. Subsequent research efforts focused on breakeven should be performed to aim at the ultimate objective of significantly enhancing both the economy and efficiency of MSFRs. Achieving such goals involves enabling semi-permanent reactor operation and substantially reducing the amount of nuclear spent fuel through the consumption of existing spent nuclear fuel. This research holds the potential to fully realize the benefits of the breakeven concept and effectively address key challenges in the nuclear energy landscape.

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