Neutronic Feasibility of a Breakeven Molten Salt Fast Reactor Using a Hybrid Chloride-Fluoride Salt

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1. Introduction

With rising environmental concerns and the increasing need for energy security, nuclear power is being reconsidered as a viable alternative to fossil fuels [1]. However, the sustainable expansion of nuclear energy faces three major challenges: enhancing reactor safety, managing spent fuel, and securing sufficient uranium resources. The inefficient utilization of uranium in LWRs, combined with the diminishing capacity for spent fuel storage, threatens the long-term feasibility of these reactors. A promising approach to addressing these issues lies in utilizing spent nuclear fuel as a new fuel source through the development of advanced reactor technologies.

Molten salt reactors (MSRs), particularly molten salt fast reactors (MSFRs), provide several advantages, including improved passive safety [2], prevention of spent fuel build-up, and enhanced proliferation resistance. Recent studies have explored chloride-fluoride hybrid salts, which are beneficial for reducing reactor size thanks to their exceptionally high fuel density. The BeMFR achieves initial criticality by using transuranic (TRU) elements recovered from PWR spent fuel through pyro-processing [3], a method that extracts TRU, uranium, and rare earth (RE) elements simultaneously, offering a proliferation-resistant solution. This strategy facilitates long-term reactor operation without relying on natural uranium. To address the challenges outlined, this work proposes a closed fuel cycle based on a breakeven molten salt fast reactor (BeMFR) utilizing chloride-fluoride hybrid salts.

2. Fuel Cycle and Reactor Model

2.1 Fuel Cycle

Figure 1 depicts the BeMFR-based closed fuel cycle. In the system, breakeven is achieved by continuously removing fission products (FPs) and supplying make-up fuel. Noble gases are extracted via helium bubbling, valuable fission products like Pd and Nd can be reclaimed after cooling, while highly radioactive species such as Cs and Sr require dedicated storage. For initial startup, LWR spent fuel undergoes pyro-processing to lower uranium content and increase the TRU fraction, ensuring initial criticality. Despite the reduction in uranium, a substantial amount remains, preserving the proliferation resistance of the resulting fuel. After achieving criticality, only simplified pyro-processing for metallization and chlorination is needed, enabling direct feeding of LWR spent fuel into the BeMFR core, enhancing both economic and proliferation resistance aspects.

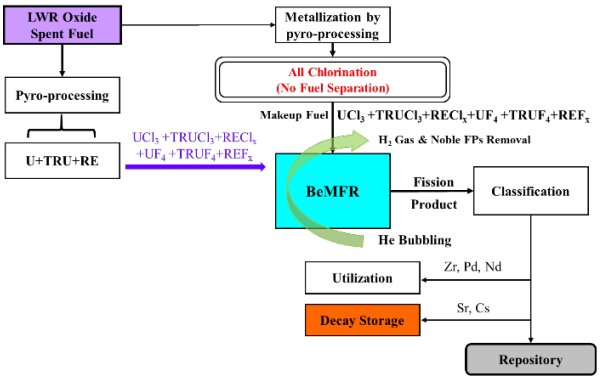

Fig. 1. Closed fuel cycle based on the BeMFR

2.2 Fuel and Reactor Model

The BeMFR is designed to maximize the energy potential of spent nuclear fuel. This study explores operational strategies and assesses the neutronic feasibility of achieving breakeven conditions in the BeMFR through computational analysis. The evaluation utilizes spent fuel from a single fuel assembly of the Korean APR1400 [4], irradiated to a burnup of 50,000 MWD/MTU. Depletion calculations, conducted with the Monte Carlo-based Serpent 2 code, provide the fuel composition after a 10-year cooling period. Spent nuclear fuel serves as make-up fuel after removing non-RE fission products. The composition of the resulting fuel, in terms of mass ratio, consists of 97.2% U, 1.3% TRU, and 1.5% RE, and its detailed composition can be found in Table I. As the make-up fuel is predominantly uranium and undergoes only minimal pyro-processing, proliferation resistance remains uncompromised.

Table I: Spent fuel composition after removing non-RE fission products

Element	Mass fraction [%]	Element	Mass fraction [%]	Element	Mass fraction [%]

Y	5.122E-04	Eu	2.129E-04	Th	1.307E-09
La	1.797E-03	Gd	4.361E-04	Pa	2.430E-11
Ce	3.486E-03	Tb	7.060E-06	U	9.718E-01
Pr	1.618E-03	Dy	3.658E-06	Np	4.530E-04
Nd	5.925E-03	Ho	4.905E-07	Pu	1.120E-02
Pm	1.280E-05	Er	2.090E-07	Am	1.106E-03
Sm	1.209E-03	Tm	1.246E-09	Cm	2.625E-04

Figure 2 shows the phase diagram of the KCl-UCl_3-UF_4 system, referred to as the chloride-fluoride hybrid salt. This hybrid salt has a melting point of about 475°C at the 28%-36%-36% eutectic composition. In this study, fuel salt compositions are formulated based on these systems, keeping the KCl fraction constant at 28%. The total content of the remaining chlorides (UCl_3, $TRUCl_3$, $RECl_3$) and fluorides (UF_4, $TRUF_4$, REF_4) are each set at 36%. The initial core's criticality is managed by modifying the combined fraction of U, TRU, and RE, with TRU and RE compositions assumed to be identical to those of spent nuclear fuel (SNF) in Table I.

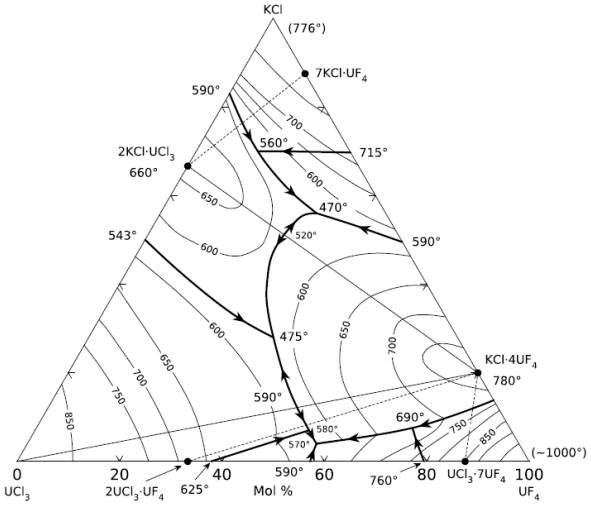

Fig. 2. Phase diagram of KCl-UCl_3-UF_4 [5]

In this study, a cylindrical core was modeled as an example of the simplified structure of the BeMFR, as shown in Figure 3. The active core, filled with liquid fuel, is surrounded by a stainless steel-based reflector to enhance neutron economy. Outside the reflector, a heat exchanger connected to the active core is positioned, facilitating the circulation of molten salt fuel between the core and the heat exchanger. Additionally, reactivity control devices are installed within the reflector. The active core is designed with a diameter and height of 300 cm, maintaining equal dimensions.

In the MSR system, freshly generated nuclides in the active core need to be diluted by inactive salt found in the heat exchangers and pipelines of the primary loop. As a result, the volume of this inactive salt significantly influences the reactor's behavior. In this study, the inactive salt volume is fixed at 15 m^3 for both models. Each reactor operates at a thermal power of 3,000 MWth, with further design parameters detailed in Table II.

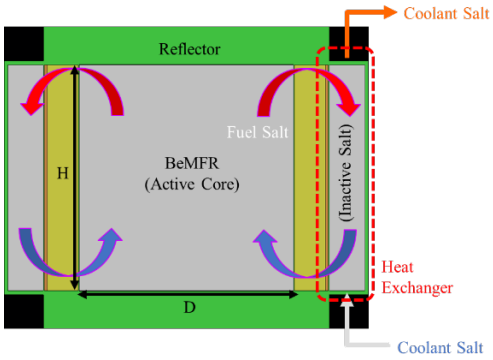

Fig. 3. Side view of BeMFR

Table II: Design parameter of initial BeMFR

Power	3,000 MWth
Initial molar composition of KCl-$TRUCl_3$-UCl_3-$RECl_3$-$TRUF_4$-UF_4-REF_4	28.00-6.60-19.40-10.00-6.60-19.40-10.00
Density (650°C)	4.538 g/cm^3
Diameter	300 cm
Height	300 cm
Active core volume	2.121E+7 cm^3
Inactive salt volume	1.500E+7 cm^3
U mass	71,524 kg
TRU mass	21,069 kg
RE mass	18,696 kg
Average fuel temperature	923.15 K
Pressure of the system	1 atm

3. Numerical Results

This section outlines the operational strategies and corresponding numerical results. The simulations were conducted with the Monte Carlo-based Serpent 2.2.1 code, utilizing the ENDF/B-VII.1 nuclear data library.

In MSRs, fission products can be classified into noble gases, noble metals, and soluble FPs based on their behavior. Noble gases are FPs that escape in gaseous form, noble metals are those that do not dissolve in molten salt and precipitate, and soluble FPs are those that dissolve in molten salt. The elements in each group are summarized in Table III.

Table III: Fission product classification [6]

Noble gas	Kr, Xe, Rn
Noble metal	Co, Ni, Cu, Ge, As, Se, Mo, Tc, Ru, Rh, Pd, Ag, Sn, Sb, Te, W, Re, Os, Ir, Au, Hg, Bi, Po
Soluble FP	Cr, Mn, Fe, Zn, Ga, Br, Rb, Sr, Y, Zr, Nb, Cd, In, I, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Ta, Pt, Tl, Pb, At, Fr, Ra

In this BeMFR, to accurately replicate the behavior of noble gases prone to easy removal and ensure proper hydrogen extraction, the noble gas removal rate is set at 1% per second, while hydrogen is removed at a rate of 90% annually. Make-up fuel is added at a rate that

closely compensates for actinide consumption during operation, with actinides supplied at 3.074 kg/day and RE at 0.0474 kg/day. The noble metal removal rate is maintained at 60% per year. These parameters remain consistent throughout all operational periods, as summarized in Table IV.

Table V shows the soluble FP removal rates obtained from various simulations. To maintain reactivity at an appropriate level, the operational period was divided into multiple intervals, with the length of each interval and the soluble FP removal rate during each period precisely adjusted. The reactor operation simulation conditions include 200,000 histories per cycle, 100 inactive cycles, 300 active cycles, and an operational period of 100 years. For the removal of soluble fission products, the removal rate initially starts high, drops sharply, and then gradually rises. At equilibrium, the soluble fission product removal rate stabilizes at 2.6% annually. To maintain the KCl ratio and prevent Cl deficiency from the cation increase due to fission product buildup, appropriate amounts of K and Cl are supplied.

Table IV: Reactor operation strategy (common)

Noble gas removal rate	1 %/s
Noble metal removal rate	60 %/y
Hydrogen removal rate	90 %/y
Makeup fuel feeding rate	3.074 kg/d
RE feeding rate	0.0474 kg/d

Table V: Soluble FP removal rate

	Period [y]	Soluble FP removal rate
Period I	0-1	7.0 %/y
Period II	1-2	7.0 %/y
Period III	2-3	5.0 %/y
Period IV	3-4	4.0 %/y
Period V	4-5	2.5 %/y
Period VI	5-20	1.5 %/y
Period VII	20-50	2.5 %/y
Period VIII	50-	2.6 %/y

Figure 4 illustrates the calculated variations in reactivity, effective delayed neutron fraction (beta), and conversion ratio. During the transitional stages, reactivity fluctuations are observed due to the accumulation of fission products and the conversion of actinides. Towards the end of the process, these initial fluctuations gradually stabilize, leading to convergence in both reactivity and conversion ratio. As mentioned earlier, this scenario is suboptimal, suggesting that dividing the periods into smaller intervals and fine-tuning the fission product removal rates can significantly reduce or even eliminate these fluctuations. In this case, reactivity stabilizes between 200 and 500 pcm. The effective delayed neutron fraction remains around 400 pcm throughout the entire operational period. Considering a reactivity and beta reduction of around 200 pcm due to molten salt behavior [7], it can be observed that the surplus reactivity remains at approximately 1.5$ throughout the entire operational period. For the conversion ratio, it begins at 1.01 and rises to 1.03 after an increase and subsequent drop in the ratio.

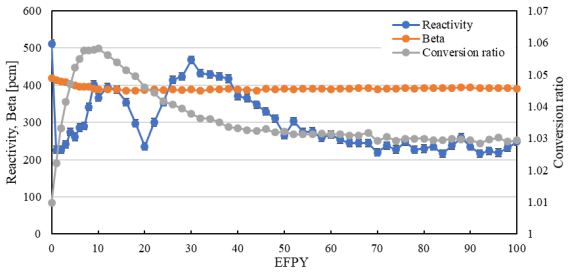

Fig. 4. Reactivity, beta and conversion ratio evolutions

Figure 5 shows the variations in volume during operation, ranging from -2% to +4%. Although a system that can accommodate these volume changes may be feasible, adjustments to the design can be made to manage these fluctuations if needed. It is important to note that the main factor affecting volume change is the RE content in the initial core.

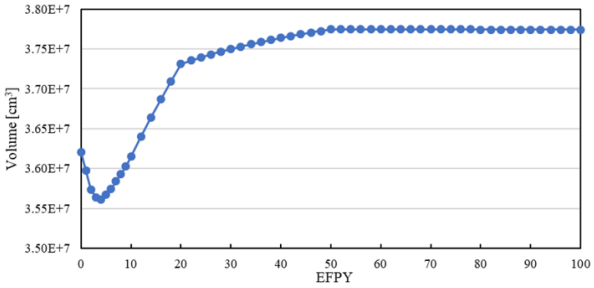

Fig. 5. Fuel volume evolution

Figure 6 illustrates the mass changes of U-238 and Pu-239 over time. Both U-238 and Pu-239 gradually increase in mass, nearing equilibrium. As the rates of feeding and consumption reach balance, both reactivity and the masses of U-238 and Pu-239 stabilize at their equilibrium levels. At equilibrium, the mass of U-238 exceeds 64 tons, while the mass of Pu-239 is around 9.2 tons.

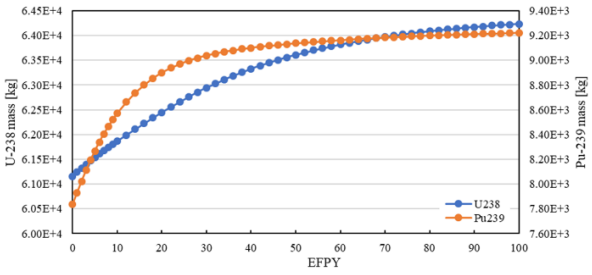

Fig. 6. U-238 and Pu-239 mass evolutions

Figure 7 shows the masses of noble metals and soluble fission products. Noble metals reach equilibrium faster than soluble fission products, owing to their higher

removal rate. Consequently, the mass of noble metals remains much lower than that of soluble FPs throughout the operation. The mass of noble metals stabilizes at approximately 600 kg. In contrast, soluble FPs exhibit a piecewise linear pattern in the initial and transition cores, gradually converging to a stable value over time. The sharp linear behavior observed at the boundaries of each period in the initial and transition cores is due to the sudden change in the soluble FP removal rate at those boundaries. At equilibrium, the mass of soluble FPs is around 20 tons. This study demonstrates that equilibrium can be achieved through proper feeding of make-up fuel and removal of fission products.

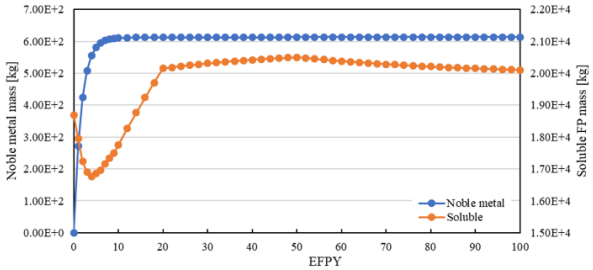

Fig. 7. Noble metal and soluble FP mass evolutions

4. Conclusions

The core concept of the hybrid salt-based BeMFR proposed in this study is to minimize the reactor size by achieving a high fuel density. Using the operational strategy outlined, excess reactivity could be maintained within 500 pcm throughout the entire operational period by adjusting the removal rates of noble metals and soluble fission products. After approximately 50 to 60 years, most operational parameters stabilize and approach a quasi-equilibrium state. The reactor is capable of operating solely on spent nuclear fuel, eliminating the need for natural uranium. This highlights the technology's potential to decrease dependence on fossil fuels and contribute to carbon neutrality. By utilizing spent fuel, the BeMFR can play a crucial role in combating the climate crisis and facilitate a transition to a more sustainable and secure energy.

Acknowledgments

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