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Assessment of Fission Product Migration in Molten Salt and Metal Reactor (MSMR)

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1. Introduction

For the mitigation of climate change and the replacement of fossil fuels, which are expected to be depleted soon, nuclear power is regarded as one of the best alternatives. Gen IV reactors are being developed and researched worldwide to achieve safety, fuel economy, appropriate spent fuel disposal, and other important objectives. Molten Salt Reactor (MSR) is one of the Gen IV reactors that uses molten salt as fuel and coolant. [1] Among them, the Molten Salt Fast Reactor (MSFR) is considered to have the greatest potential. MSFR is a type of MSR that utilizes a fast neutron spectrum. [2]

A specialized purpose mobile reactor, known as the Molten Salt and Metal Reactor (MSMR), has been introduced for use in various applications such as military bases, mining industries, small villages in remote areas, and space exploration, among others. The MSMR is an application of MSFR and utilizes two liquid layers consisting of liquid metal and molten salt, which work as fuel and coolant, respectively. It has been developed to overcome the technical difficulties of MSFR in achieving criticality at an ultra-micro size that can fit inside a container. Basic properties and feasibility studies of the MSMR have been investigated and performed in previous works [3, 4]. Therefore, this work focuses on assessing the behavior of fission products.

2. Basic Concept of MSMR

2.1 Explanation of MSMR Model

As discussed in the introduction, MSMR utilizes liquid metal and molten salt as layers, which are contained in the core. The liquid metal in the lower layer generates heat, and the molten salt in the upper layer transfers heat to the secondary system and contains some of the fission products. The gas plenum on the top of the upper layer accommodates gaseous fission products and the heat expansion of the lower and upper layers. The reactor vessel is composed of corrosion-resistant and high-melting-temperature materials and it contains and conserves the fuel and the coolant. In the outer region, there are the reflector, drum-shaped reactivity control device, and the secondary system. The reflector helps achieve a high neutron economy, and the reactivity control device controls excess reactivity. The secondary system removes the heat from the primary system and uses it for the generation of electrical energy. Note that the heat removal is performed by cooling down the surface of the reactor vessel and primary heat exchanger in some cases. The overall conceptual design is depicted in Fig. 1.

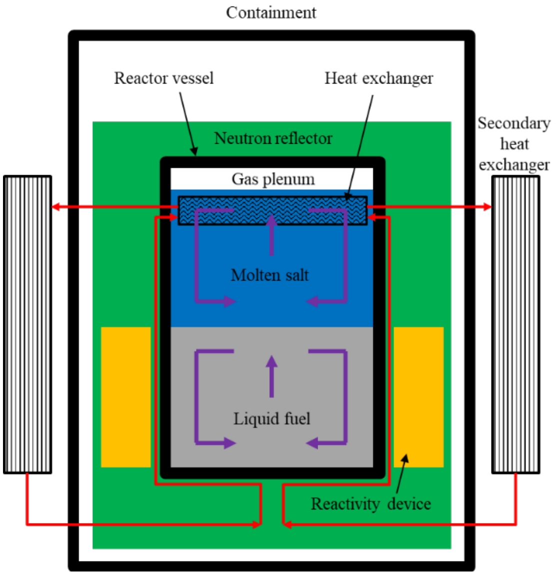

Fig. 1. Conceptual structure of MSMR

2.2 Detailed Structure and Material of MSMR

For computer program analysis, a specification of the model is required, and appropriate materials need to be selected. Figures 2 and 3 provide the details, where the overall structure can be found in the former, and the structure of the drum-shaped reactivity control device can be understood in the latter.

The lower layer that generates nuclear heat is composed of U-Fe, and the upper layer that plays the role of coolant is composed of $NaCl$-KCl-$MgCl_2$. The two layers have the same dimensions, with a diameter of 65 cm and a height of 62.3 cm, respectively. The height of the gas plenum is 5 cm, and it is filled with air. The reactor vessel's thickness is 1 cm, and its material is Tantalum, which has strong corrosion resistance [5] and high melting temperature. A secondary system has been implemented outside of the reactor vessel in the form of a 2 cm gap filled with molten salt $NaCl$-KCl-$MgCl_2$. The reflector, which is 40 cm thick and composed of stainless steel, wraps around the reactor vessel and the secondary system. Note that there is a 0.1 cm protective layer composed of Tantalum between the secondary molten salt region and the reflector. Finally, 8 drum-shaped reactivity control devices are installed in the reflector region, with a diameter of 38 cm and a height of 62.3 cm. Their body is composed of stainless steel, and 4 cm thickness of B_4C pads are attached for neutron absorption.

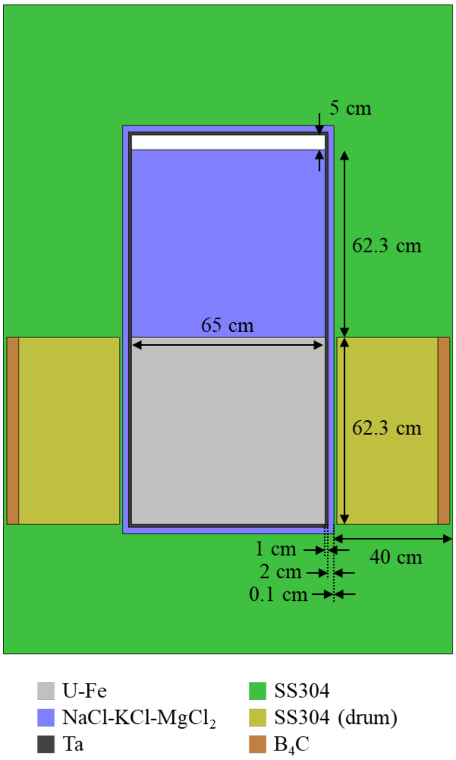

Fig. 2. Detailed structure of MSMR (Side view)

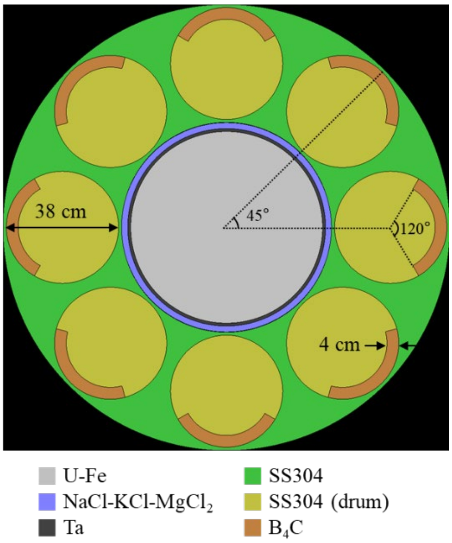

Fig. 3. Detailed structure of MSMR (Top view)

U-Fe and NaCl-KCl-MgCl$_2$, which are used as fuel and coolant, have eutectic compositions. Their detailed information is tabulated in Table I. Figures 4 and 5 show the phase diagrams of U-Fe and NaCl-KCl-MgCl$_2$ that provide the eutectic information. Please note that the heavy metal inventory can be calculated as 2,818 kg.

Table I: Information about of U-Fe and NaCl-KCl-MgCl$_2$

	U-Fe	NaCl-KCl-MgCl$_2$
Eutectic composition	89-11 (mass)	30.2-22.7-47.1 (mol)
Melting temperature	723°C	385°C
Enrichment	12 wt.% U-235	99 at.% Cl-37

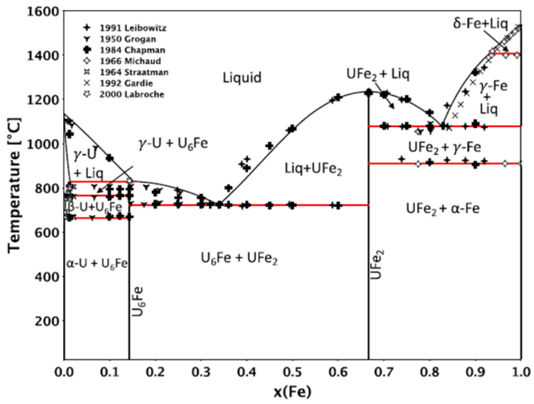

Fig. 4. Phase diagram of U-Fe [6]

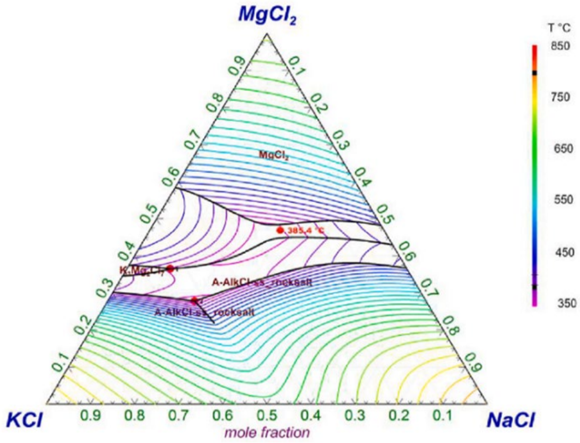

Fig. 5. Phase diagram of NaCl-KCl-MgCl$_2$ [7]

2.3 Basic Properties of MSMR

For the analysis of the basic properties of MSMR, the Monte Carlo-based reactor analysis program Serpent 2.2.0 was used, with ENDF/B-VII.1 selected for the nuclear data. 100,000 were utilized for history, and 100 and 300 cycles were chosen for inactive and active cycles, respectively. The burnup-dependent reactivities are displayed in Fig. 6, while burnups and conversion ratios are presented in Fig. 7. The reactor lifetime can be estimated to be more than 140 years, with a maximum reactivity estimated of around 250 pcm. The discharge burnup is estimated to be approximately 45 MWd/kgU, and the conversion ratios range from 0.68 to 0.73 throughout the lifetime. In addition, the reactivity-drum worth is calculated to be 2,629 ± 20 pcm. Additionally, the fuel temperature coefficients (FTCs) of MSMR were calculated for four temperature intervals, using 10 times the number of samples for history. These are shown in Table II.

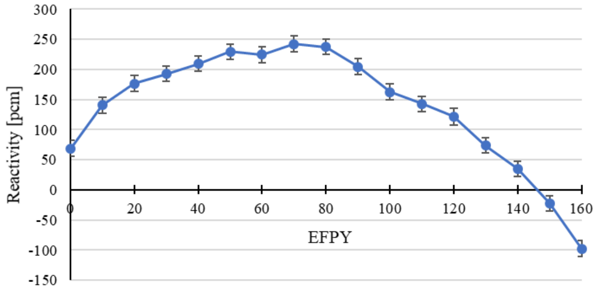

Fig. 6. Burnup-dependent reactivities for 2.5 MWth power

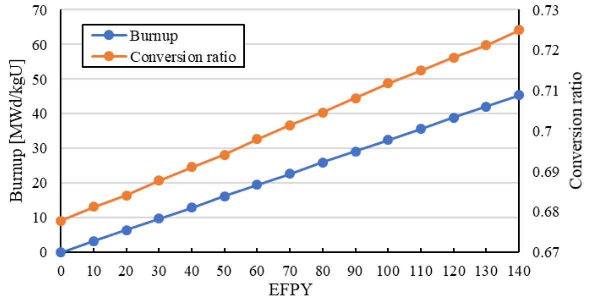

Fig. 7. Burnups and conversion ratios for 2.5 MWth power

Table II: Fuel temperature coefficient of MSMR

Temperature range	Temperature coefficient
800 - 900	-2.48 ± 0.06
900 - 1000	-2.48 ± 0.06
1000 - 1100	-2.51 ± 0.06
1100 - 1200	-2.67 ± 0.06

3. Numerical Results of Fission Product Study

In the MSR, fission products can be classified into three groups: noble gases, noble metals, and ‘others.’ Noble gases include gaseous fission products that do not dissolve in liquid fuel and molten salt. Noble metals include non-gaseous fission products that do not dissolve in molten salt. ‘Others’ refer to fission products that are not included as noble gases or noble metals and can be dissolved in molten salt. Table III enumerates the fission products that have been produced in the MSMR at a 45.4 MWd/kgU burnup, with a total mass of 131.2 kg.

Table III: Fission products from MSMR at 45.4 MWd/kgU

Element	Type	Mass fraction [%]
Xe	Noble gas	12.58606
Kr	Noble gas	1.163383
Rn	Noble gas	8.42E-14
Zr	Noble metal	12.44069
Nb	Noble metal	0.002414
Mo	Noble metal	9.971559
Tc	Noble metal	2.471687
Ru	Noble metal	5.93603
Rh	Noble metal	1.767941
Pd	Noble metal	2.280483
Ag	Noble metal	0.124034
Cd	Noble metal	0.214767
In	Noble metal	0.034521
Sn	Noble metal	0.325362
Sb	Noble metal	0.081617
Hf	Noble metal	4.52E-17
Hg	Noble metal	1.41E-24
Tl	Noble metal	4.64E-14
Pb	Noble metal	7.71E-07
Bi	Noble metal	8.63E-11
Te	Noble metal	1.446093
Se	Noble metal	0.192263
Po	Noble metal	1.8E-12
I	Noble metal	0.693799
At	Noble metal	3.78E-21
La	Others	3.629297
Ce	Others	6.794389
Pr	Others	3.39548
Nd	Others	12.10858
Pm	Others	0.038028
Sm	Others	2.675983
Gd	Others	0.123585
Dy	Others	0.002517
Ho	Others	8.84E-05
Er	Others	0.000111
Tm	Others	7.55E-06
Yb	Others	4.25E-06
Y	Others	1.481246
Rb	Others	1.226256
Cs	Others	8.660187
Sr	Others	1.629292
Ba	Others	6.435432
Br	Others	0.066805

During operation of the MSMR, fission products may move and affect the reactor's properties. Noble gases are expected to be released from the core and gather in the gas plenum. Noble metals are expected to remain in the fuel region as liquid or precipitation, and some of the ‘others’ may move to the upper coolant layer.

For the assessment, several assumptions have been made. Firstly, all noble gases are fully removed from the core and any gathering in the gas plenum is neglected. Next, some of the fission products that are classified as ‘others’ - so-called salt-seeker elements - are assumed to move to the upper molten salt zone. Their ratios are determined as constant: 0%, 50%, or 100%. These are classified into Case A, B, and C, respectively, as arranged in Table IV. Lastly, the change in volume of the fuel zone is neglected, but the corresponding density change is considered. The assessments have been performed under the same conditions as in Section 2.3.

Table IV: Fission products removal Case summary

	Noble gas	Salt-seeker elements
Reference	Not removed	Not removed
Case A	Completely removed	Not removed
Case B	Completely removed	50% removed

Case C	Completely removed	100% removed

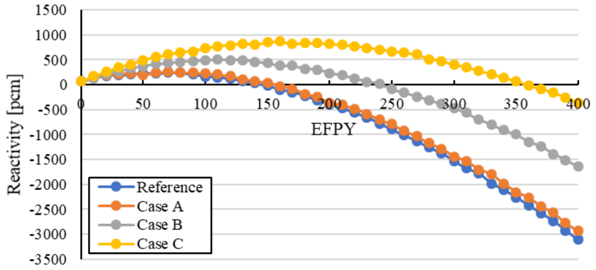

Fig. 8. Burnup-dependent reactivities of MSMR with 2.5 MWth power under various Cases

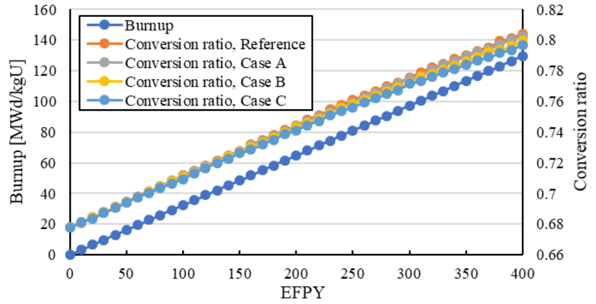

Fig. 9. Burnups and conversion ratios of MSMR with 2.5 MWth power under various Cases

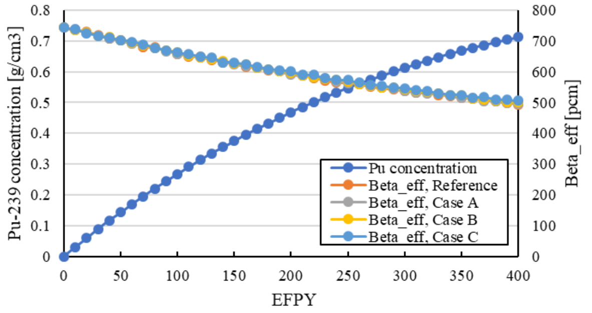

Fig. 10. Burnup-dependent Plutonium-239 concentrations and effective delayed neutron fraction of MSMR with 2.5 MWth power under various Cases

When looking at Fig. 8, the estimated lifetimes for Case A, B, and C are around 150, 240, and 350 years, respectively, and the reactivity swings are estimated to be 260, 510, and 830 pcm, respectively. Removing a greater fraction of fission products leads to higher reactivity and longer estimated lifetime due to the decrease in neutron absorptions caused by fission products. Figure 9 shows that removing more fission products leads to a lower conversion ratio, although the differences are not significant. The discharge burnup for reactors of Case A, B, and C can be calculated at 49, 78, and 113 MWd/kgU, respectively, indicating higher fuel utilization performance for the reactor removing a greater fraction of fission products. In Fig. 10, the concentration of plutonium-239 increases as burnup increases, whereas the effective delayed neutron fraction decreases due to the increasing contribution of plutonium-239. The effective delayed neutron fraction can be reduced to around 500 pcm if the removal of fission products is maximized as in Case C.

4. Conclusions

Without considering the behavior of fission products, the Molten Salt and Metal Reactor (MSMR) has a lifetime of around 140 years at 2.5 MWth, with a discharge burnup of about 45 MWd/kgU. However, when taking into account the movement of fission products, the lifetime and fuel utilization can increase to more than double if all ‘other’ fission products are removed. This is advantageous, but it may also pose a challenge in terms of reactivity control, as the reactivity swing increases accordingly. In future work, a more precise understanding of the behavior of fission products can be achieved by assessing their solubility in the molten salt. Additionally, it may be necessary to distinguish between noble metals that remain in a liquid state and those that are deposited on the inner surface of the reactor vessel.

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