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NEUTRONIC ANALYSIS ON MOLTEN SALT REACTOR FUJI-12 USING ^{235}U AS FISSILE MATERIAL IN LiF-BeF$_2$-UF$_4$ FUEL

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Neutronic analysis on the Molten Salt Reactor FUJI-12 using the fissile material ^{235}U in LiF-BeF$_2$-UF$_4$ has been carried out. The problem faced in the use of thorium-based fuel is that the amount of ^{233}U is small and not available in nature. ^{233}U was produced through the ^{232}Th breeding at a cost of \$46 million/kg. That is a very high price when compared to ^{235}U enrichment, which is only \$100/kg. The MSR FUJI-12 used in this study is a generation IV reactor with a mixture of liquid salt fuel LiF-BeF$_2$-ThF$_4$-UF$_4$ and thorium-based fuel (^{232}Th+^{233}U). In this study, neutronic analysis was carried out by replacing thorium-based fuel with uranium-based fuel (^{235}U+^{238}U). Neutronic analysis was performed using the OpenMC 0.13.0 code, which is a Monte Carlo simulation-based neutron analysis code. The nuclear data library used for neutronic calculations is ENDF B-VII/1. The fuel is used in a LiF-BeF$_2$-UF$_4$ molten salt mixture with three eutectic compositions: fuel 1, fuel 2, and fuel 3. Each fuel composition is optimized by enriching ^{235}U in UF$_4$ by 3 % to 8 %. The optimization results show the stability of the reactor criticality value, which is the main parameter so that the reactor can operate for the specified time. The optimization results show that fuel 1 cannot reach its optimal state in each variation of ^{235}U enrichment. Fuel 2 and fuel 3 can reach optimal conditions at a minimum enrichment of 8 % and 7 % ^{235}U. The results of the analysis of the distribution of the neutron flux in the reactor core show the distribution of nuclear reactions that occur in the core. The distribution of flux values in fuel 1 shows that the fission chain reaction is not running perfectly. Fuel 2 and fuel 3 are more stable by maintaining maximum flux at the center of the reactor core

Keywords: molten salt reactor, OpenMC, uranium fluoride, thorium fluoride, neutron flux

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1. Introduction

The demand for electrical energy in 2021 increased by 6 % (over 1,500 TWh) and this was the largest absolute increase ever since the recovery from the financial crisis in 2010. The increase is related to the rapid recovery from the Covid-19 pandemic in 2021. The growth in electricity demand is expected to continue at 3 % to 4 % in 2022 and 2 % in 2024 [1]. The growth in electricity demand in 2021 is followed by a growth in the number of coal power plants by 9 % and fulfills almost half of the growth in electricity demand in 2021 [2]. The use of coal drives global electricity sector emissions to grow by close to 7 %, contributing more than 800 Mt of CO_2 emission growth. By 2024, it is estimated that emissions from coal and gas power plants combined can reach more than 13 Gt CO_2 [1].

Nuclear power plant is one of the renewable energy alternatives that can replace the role of coal in balancing the current world's growing demand for electrical energy. Based on the latest data, there are 440 nuclear reactors in operation and spread in 32 countries with a generated capacity of 39.1119 MWe [3]. MSR FUJI-12 is one type of MSR reactor (Molten Salt Reactor), which is included in one of the generation IV reactor designs. This reactor was developed by Japan with the smallest reactor core size when compared to other similar nuclear reactors [4].

MSR reactors such as the MSR FUJI-12 generally use ^{232}Th+^{233}U as the LiF-BeF$_2$-ThF$_4$-UF$_4$ liquid fuel mixture.

The availability of ^{233}U is very low because it is not a natural material and must be produced through ^{232}Th breeding, so it is very difficult to obtain. Another problem faced in the use of $^{232}Th+^{233}U$ fuel is that it is more expensive to produce than natural uranium-based fuels. The production cost for ^{233}U is \$46 million/kg, which is very high when compared to ^{235}U enrichment, which is only \$100/kg [5, 6]. The manufacturing process for thorium-based fuels is also more difficult, which can also increase the cost of manufacturing fuel in MSR.

Uranium materials that occur in nature are ^{234}U, ^{235}U and ^{238}U. The combination of ^{235}U and ^{238}U is a mixture that is commonly used as a fissile and fertile material in nuclear reactor fuels. An investigation to replace the $^{232}Th+^{233}U$-based MSR FUJI-12 fuel with $^{238}U+^{235}U$ needs to be carried out. This is based on the potential for reducing the production cost of the MSR FUJI-12 due to the ^{233}U production process. Manufacturing thorium-based fuels is also more difficult than uranium, so replacing thorium-based fuels also reduces production costs and simplifies the manufacturing process. In addition, the moderator temperature reactivity coefficient value for the $^{238}U+^{235}U$ fuel is considered safer for nuclear reactors. Therefore, it is necessary to conduct a neutronic analysis on the use of ^{235}U and ^{238}U in the LiF-BeF_2-UF_4 fuel mixture as a cheaper and safer MSR FUJI-12 fuel solution.

2. Literature review and problem statement

The paper [7] presents a comparison of the utilization of ^{233}U and plutonium in MSR on thorium-based MSR fuel. The results showed that the average number of neutrons produced by the ^{233}U fuel per neutron absorbed in the thermal energy region was higher than that of plutonium. This shows that it takes less concentration of ^{233}U to operate the FUJI-12 MSR reactor at a power of 350 MWt compared to the use of plutonium, either in reactor grade or weapon grade enrichment. The problem is that using ^{233}U in ^{232}Th-based fuel is still very expensive. This is due to the large production cost of ^{233}U and the relatively more difficult manufacturing cost of thorium-based fuel.

An approach to replace ^{233}U with ^{235}U in ^{232}Th-based fuels has been implemented in [8]. The results show that the operation of 50 MWt in a mini-FUJI reactor for 5 years can be carried out with a minimum enrichment of 95 % ^{235}U with a UF_4 composition of 1.96 %. This study shows that it is possible to replace ^{233}U with ^{235}U in ^{232}Th-based fuels. The problem is that the ^{235}U enrichment required is very large and is in the weapon grade uranium range. This should not be applied to nuclear power plants that are only allowed to enrich the reactor grade uranium range.

The research [9] conducted an approach by comparing fuels based on $(^{232}Th+^{233}U)O_2$ and $(^{235}U+^{238}U)O_2$ in the PWR Russian VVER-1200 reactor using the SERPENT code. The results for operating power of 3200 MWt show that fuel based on $^{232}Th+^{233}U$ has a lower reproduction factor and number of neutron production than fuel based on $^{235}U+^{238}U$ for an operating duration of 1440 days. In addition, it is shown that the fuel based on $^{235}U+^{235}U$ has a higher moderator temperature reactivity coefficient value. This research shows that uranium-based fuels have the potential to replace thorium-based fuels. In addition, based on the moderator temperature reactivity coefficient, fuel based on $^{235}U+^{235}U$ is safer than that based on $^{232}Th+^{233}U$.

An approach to MSR has been carried out in [10] by comparing LiF-BeF_2-UF_4 (FLiBe) and NaF-BeF_2-ThF_4-UF_4 (FNaBe) fuels in the NERTHUS Thermal Molten Salt Reactor. The total amount of uranium required for FLiBe is greater than for FNaBe. The ^{235}U enrichment in FLiBe was smaller (2.0935 %) than in FNaBe (19.4512 %). But for continuous power operation, the fuel refueling ratio of FLiBe is much smaller than that of FNaBe. The results of this study [10] are also in line with research [9] with a higher moderator temperature reactivity coefficient value for the beginning of the reactor operation and the end of the reactor operation. Another study [11] compared the corrosiveness of several variations of molten salt fuel in MSR. The level of corrosiveness and LiF-BeF_2-UF_4 in Hastelloy-N alloy for each variation is 0.5–1.0 m/year, while for LiF-BeF_2-ThF_4-UF_4 – 0.4–3.5 m/year. This value indicates that the corrosiveness of LiF-BeF_2-UF_4 is relatively smaller. Based on the research [7–11], identification of the use of fuel based on $^{235}U+^{238}U$ in the LiF-BeF_2-UF_4 fuel mixture in the MSR FUJI-12 reactor needs to be done.

3. The aim and objectives of the study

The aim of the study is to replace FUJI-12 MSR based on $^{232}Th+^{233}U$ with $^{238}U+^{235}U$.

To achieve this aim, the following objectives are accomplished:

– to calculate the eutectic composition of LiF-BeF_2-UF_4, which has the potential to be used as fuel for MSR FUJI-12;

– to calculate the flux distribution in the reactor core of MSR FUJI-12 with a power of 350 MWt and an operating time of 5 years with LiF-BeF_2-UF_2 fuel.

4. Materials and methods

4. 1. Research procedures

The object of this research is the variation of the eutectic composition of the LiF-BeF_2-UF_4 fuel and the variation of ^{235}U enrichment in UF_4. This research focuses on neutronic analysis using the OpenMC 0.13.0 code and nuclear data library ENDF B-VII/1. The OpenMC code is an open-source neutronic analysis code based on a Monte Carlo calculation simulation. The research proceeded with the flow following Fig. 1.

The optimal operation in Fig. 1 is obtained by looking at the graph of the *k-eff* value for the reactor operation for 5 years. The optimal composition is a variation of ^{235}U enrichment that can maintain the *k-eff* value near the critical state (*k-eff*=1) and does not decrease in the subcritical state (*k-eff*<1). This situation must last from the start of the reactor operation to the end of the reactor operation.

4. 2. Monte Carlo method

Neutron analysis is always related to the neutron transport equation. The equation states that the rate of variation of the neutron density ($\partial n/\partial t$) is equal to the rate of neutron absorption minus the leakage rate:

$$\frac{1}{v}\frac{\partial}{\partial t}\psi(r,\Omega,E,t)+\Omega\cdot\nabla\psi(r,\Omega,E,t)+$$
$$+\sum_t(r,E)\psi(r,\Omega,E,t)=q_{ext}(r,\Omega,E,t)+$$
$$+\int_{4\pi}\mathrm{d}\Omega'\int_0^\infty \mathrm{d}E'\sum_s(r,\Omega'\to\Omega,E'\to E)\psi(r,\Omega',E',t)+$$
$$+\frac{\chi(r,E)}{4\pi}\int_{4\pi}\mathrm{d}\Omega'\int_0^\infty \mathrm{d}E'v(r,E')\sum_f(r,E')\psi(r,\Omega',E',t). \quad (1)$$

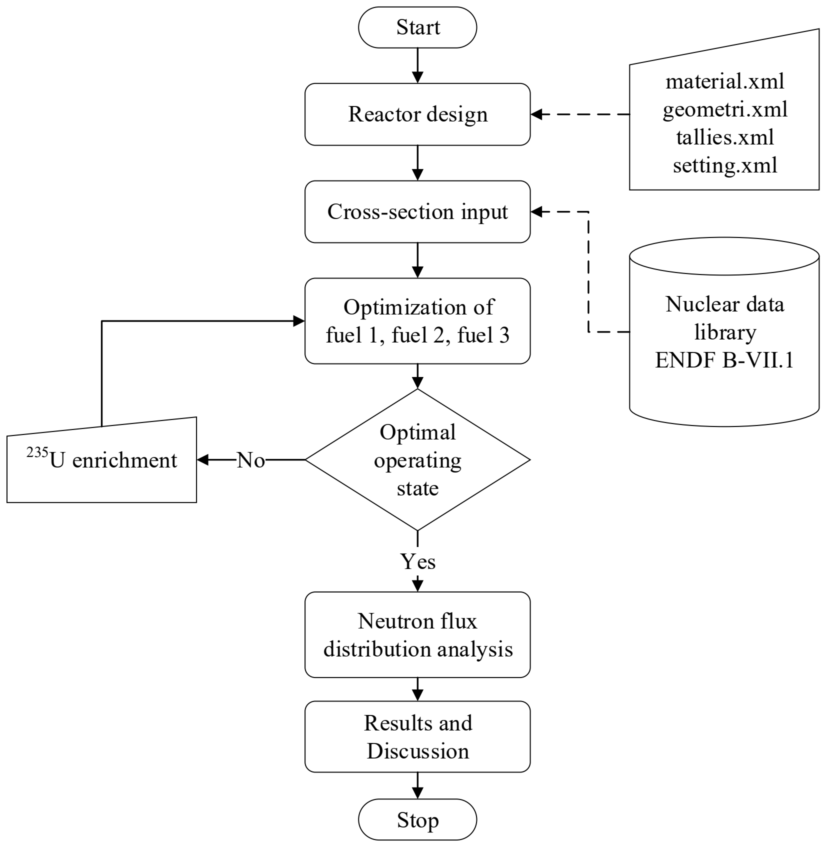

Fig. 1. Research procedure

The Monte Carlo method approach in OpenMC simulates the reactor criticality at a steady state. The *k-eff* value is formulated as the eigenvalue (k) obtained by dividing the number of fission $v(r, E')$ by the k factor [12 13] so that the equation for calculating reactor criticality can be written as:

$$\Omega\,.\nabla\psi(r,\Omega,E)+\Sigma_t(r,E)\psi(r,\Omega,E)-$$
$$\int_{4\pi}\mathrm{d}\Omega'\int_0^{\infty}\mathrm{d}E'\Sigma_s(r,\Omega'\to\Omega,E'\to E)\psi(r,\Omega',E')=$$
$$=\frac{1}{k}\frac{\chi(r,E)}{4\pi}\int_{4\pi}\mathrm{d}\Omega'\int_0^{\infty}\mathrm{d}E'\,v(r,E')\Sigma_f(r,E')\psi(r,\Omega',E'). \quad (2)$$

(2) can be simplified by defining the operators **M** and $S(r)$:

$$M\psi(r,\Omega,E)=\Omega\cdot\nabla\psi(r,\Omega,E)+\Sigma_t(r,E)\psi(r,\Omega,E)$$
$$-\int_{4\pi}d\Omega'\int_0^{\infty}dE'\Sigma_s(r,\Omega'\to\Omega,E'\to E)\psi(r,\Omega',E'), \quad (3)$$

$$S(r)=\frac{1}{k}\frac{\chi(r,E)}{4\pi}\int_{4\pi}\mathrm{d}\Omega'\int_0^{\infty}\mathrm{d}E'\,v(r,E')\times$$
$$\times\Sigma_f(r,E')\psi(r,\Omega',E'), \quad (4)$$

by substituting (3), (4) into (2), we get the equation (5) [13]:

$$\mathbf{M}\psi(r,\Omega,E)=\frac{1}{k}\frac{\chi(r,E)}{4\pi}S(r). \quad (5)$$

The operator **M** denotes the total rate of neutron loss and $S(r)$ is the operator for the fission source. If we define Green's function (G) for (5) as:

$$M\cdot G(r_0,E_0,\Omega_0\to r,E,\Omega)=$$
$$=\delta(r-r_0)\delta(E-E_0)\delta(\Omega-\Omega_0), \quad (6)$$

where the notation «0» [12] denotes the starting point in the space phase, (2), can be rewritten only in fission source form;

$$S(r)=\frac{1}{k}\int\mathrm{d}r_0H(\mathbf{r}_0\to\mathbf{r})S(\mathbf{r}_0), \quad (7)$$

$H(\mathbf{r}_0\to\mathbf{r})$ is the expected number of fissionable neutrons at **r**, since the parent fission neutrons are born at $\mathbf{r_0}$.

$$H(\mathbf{r}_0\to\mathbf{r})=$$
$$=\iint\iint\mathrm{d}\Omega\mathrm{d}E\mathrm{d}\Omega_0\mathrm{d}E_0v\Sigma_f(\mathbf{r},E)\times$$
$$\times\frac{\chi(\mathbf{r},E_0)}{4\pi}G(\mathbf{r}_0,E_0,\Omega_0\to\mathbf{r},E,\Omega). \quad (8)$$

By assuming the right hand side of (7), with the fission operator F, the eigen equation can be formulated as:

$$S=\frac{1}{k}FS. \quad (9)$$

The value of k_0=*k-eff*, so that the eigenvalue iteration equation for the Monte Carlo method can be written as:

$$S^{(i+1)}=\frac{1}{k^{(i)}}FS^{(i)}, \quad (10)$$

$S^{(i)}$ shows the sample of neutrons for the initial state in the Monte Carlo simulation. Neutrons are then simulated for their transport so that the location of the next generation $S^{(i+1)}$ is determined. Iteration starts from the initial source distribution at $S^{(0)}$ and eigenvalue $k^{(0)}$ [12].

4. 3. Design and parameters

The reactor design used is the MSR FUJI-12 with a cylindrical core, single core arrangement and composed of a core, reflector, and absorber. The configuration of each FUJI-12 MSR core structure is shown in Fig. 2.

The moderator is a component that moderates the neutrons so that the fission chain reaction can be maintained. The moderator is in the form of 271 hexagonal fuel line pins and arranged in a circle. MSR fuel is in the liquid phase, so there is a cylindrical tube through which the fuel flows called the fuel duct in the center of the hexagonal pin. The arrangement of the reactor core components and the arrangement of the fuel pins follow Fig. 3.

There are three compositions of MSR FUJI-12 fuel used in this study. The eutectic material shows the lowest melting point of the composition of a mixture of the same homogeneous compound. The combination of the wrong mole fraction of the fuel mixture can cause clumping and clog the flow rate of fuel in the fuel duct. The eutectic composition variations for the LiF-BeF$_2$-UF$_4$ fuel mixture are shown in Table 1.

The MSR FUJI-12 reactor was operated with an operating power of 350 MWt for 5 years. The reactor operates with the operating parameters in Table 2.

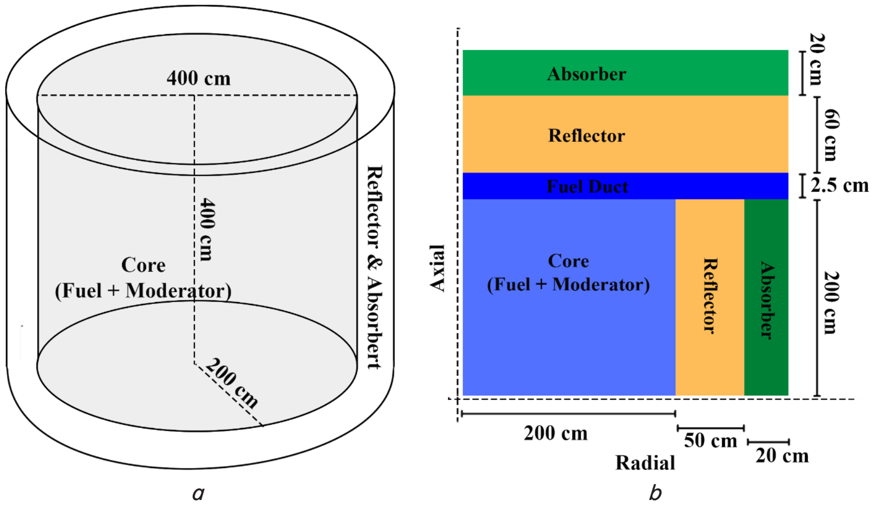

Fig. 2. Reactor design: *a* — the shape of the reactor core; *b* — the configuration of the core constituent material

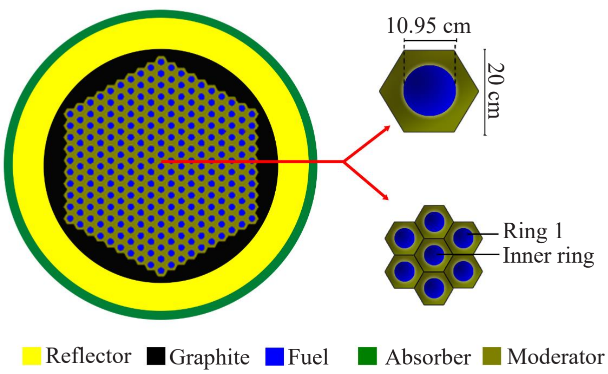

Fig. 3. Configuration and size of the fuel duct

Table 1

Eutectic composition of LiF-BeF_2-UF_4 [14, 15]

Mole Fraction (%)			Eutectic Temperature (K)	Fuel Name
LiF	BeF_2	UF_4		
48	51.5	0.5	623.00	Fuel 1
69	23	8	699.00	Fuel 2
70	12	18	700.15	Fuel 3

Table 2

Operating parameters of MSR FUJI-12

Parameter	Value
Power	350 MWt
Average power density	7 kW/liter
Burn-up	5 Years
Fuel	LiF-BeF_2-UF_4
Density	2.9 gr/cm^3
Thermal output	840 K
Moderator	
Composition	Graphite (C)
Density	1.84 gr/cm^3
Reflector	
Composition	Graphite (C)
Density	1.76 gr/cm^3
Absorber	
Composition	Boron Carbide (CB_4)
Density	2.52 gr/cm^3

Natural lithium (Li) consists of 7.6 % ^{6}Li, which has a very high absorption cross section. It is not recommended for nuclear reactor fuel. ^{7}Li enrichment needs to be carried out in the fuel mixture by 99.95 % to avoid feedback reactivity due to tritium generation by neutron absorption of the Li_6 isotope [16].

5. Results of the neutronic analysis of molten salt reactor Fuji-12 using fissile 235U in LiF-BeF2-UF4 fuel

5. 1. Optimization of LiF-BeF_2-UF_4 fuel

Optimization of LiF-BeF2-UF4 fuel for all variations is shown in Fig. 4. The optimization was carried out by simulating 15,000 neutron particles in 50 batches, consisting of 40 active batches and 10 inactive batches. This limitation results in a standard deviation error value in the range of 110 pcm to 115 pcm. Fig. 4 shows the value of *k-eff*, which is a reactor criticality parameter for the three compositions of fuel 1, fuel 2 and fuel 3 with 3–8 % ^{235}U enrichment. Based on Fig. 4, the optimal *k-eff* value for power operation of 350 MWt for 5 years can only be achieved by fuel 2 and fuel 3. The optimal state can be achieved when the *k-eff* value approaches the critical state (*k-eff*=1) from the beginning of the operating period to the end of the operating period. The minimum enrichment of ^{235}U required for fuel 2 and fuel 3 to reach a critical state is 8 % and 7 %, respectively. Fuel 1 cannot reach the optimal operating state for each variation of ^{235}U enrichment. The highest ^{235}U enrichment of 8 % can only help fuel 1 reach a critical point at the beginning of life (BOL) and then drop to a sub-critical state (*k-eff*<1).

Fig. 5 shows a neutron spectrum analysis of the three variations of the fuel mixture. The peak flux values of all variations of the fuel mixture are in the thermal energy range. Fuel 1 has the highest flux value and indicates that the burn-up of fissile fuel (^{235}U) in fuel 1 occurs more quickly. This could lead to a shortage of fissile fuel (^{235}U) to sustain reactor operation for 5 years. The peak flux of fuel 1 has a large difference from fuel 2 and fuel 3. The characteristics of fuel 2 and fuel 3 are almost similar. The peak flux difference between the two is not too far, so it has almost similar *k-eff* characteristics for 350 MWt power operation for 5 years.

The amount of ^{3}H (tritium) greatly affects the corrosiveness of the fuel to the fuel pipe in MSR. Fig. 6 shows that the amount of ^{3}H in fuel 1 is the largest and continues to increase until the end of operation. A drastic increase in the amount of ^{3}H in fuel 1 occurred at the beginning of operation until the 2nd year of operation. The amount of ^{3}H in fuel 2 and fuel 3 is relatively stable and slightly decreased from the beginning of operation to the end of operation.

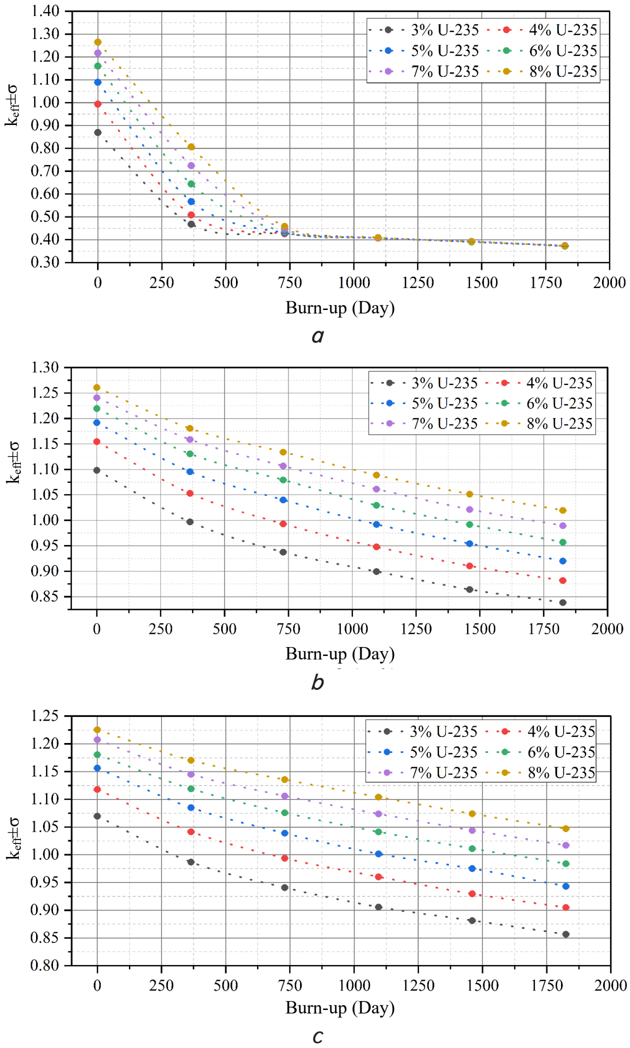

Fig. 4. Optimization of each fuel composition:
a – fuel 1; *b* – fuel 2; *c* – fuel 3

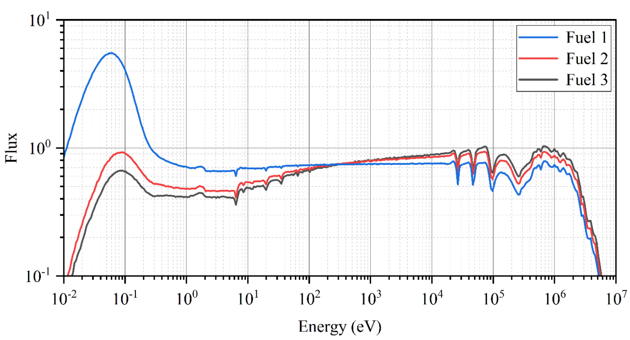

Fig. 5. Neutron spectra for each fuel

Fig. 7 shows the value of macroscopic cross section fission and absorption as a function of energy (eV). The difference can be seen by reviewing the thermal, epithermal, and fast energy ranges. Fuel 3 has the highest macroscopic cross section fission and absorption values in the thermal energy range. In the epithermal energy range, the macroscopic cross section absorption of fuel 3 is lower than that of the other two fuels. Meanwhile, in the fast energy range, the macroscopic cross section fission of fuel 3 is higher than that of the other two fuels.

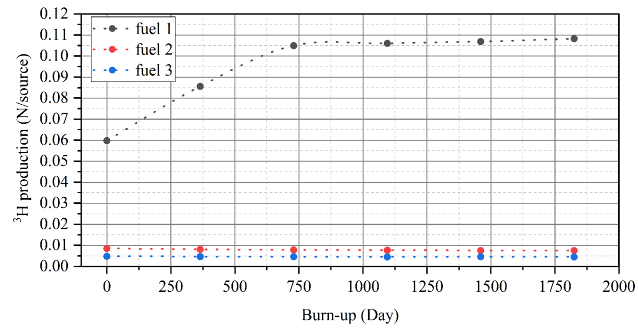

Fig. 6. Production of ^{3}H (tritium) for each fuel composition

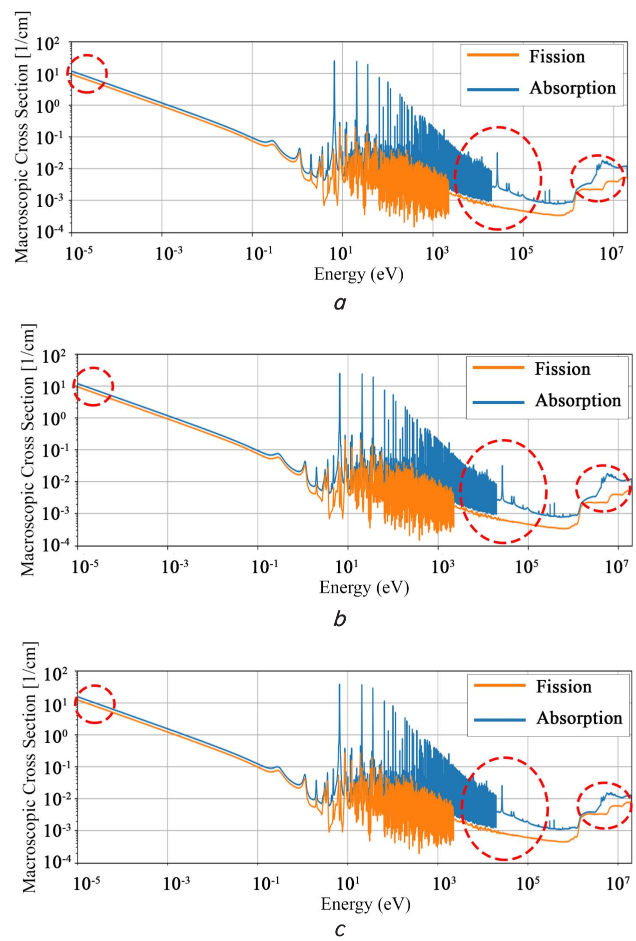

Fig. 7. Macroscopic cross section fission and absorption:
a – fuel 1; *b* – fuel 2; *c* – fuel 3

Based on Fig. 5, the FUJI-12 MSR reactor is a type of reactor with a spectrum of thermal energy. Referring to this, in Fig. 7, fuel 1 and fuel 2 have similar fission and absorption probabilities for the thermal energy range.

5. 2. Neutron flux distribution analysis for the optimal composition of each fuel

Fig. 8, 9 show the value of the flux distribution in the core reactor on the XY and XZ sections. Fig. 8 shows the flux distribution for the initial state of the reactor mass (BOL). Fig. 9 shows the flux distribution for the end state of the reactor life.

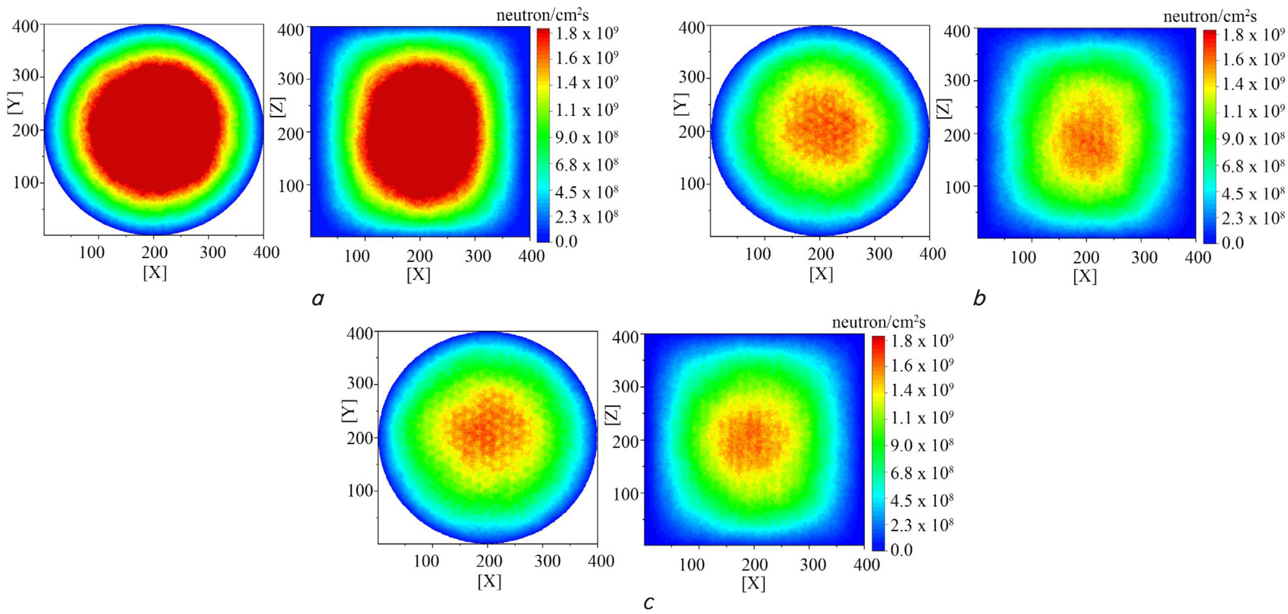

Fig. 8. Distribution of neutron flux at the beginning of life: *a* – fuel 1; *b* – fuel 2; *c* – fuel 3

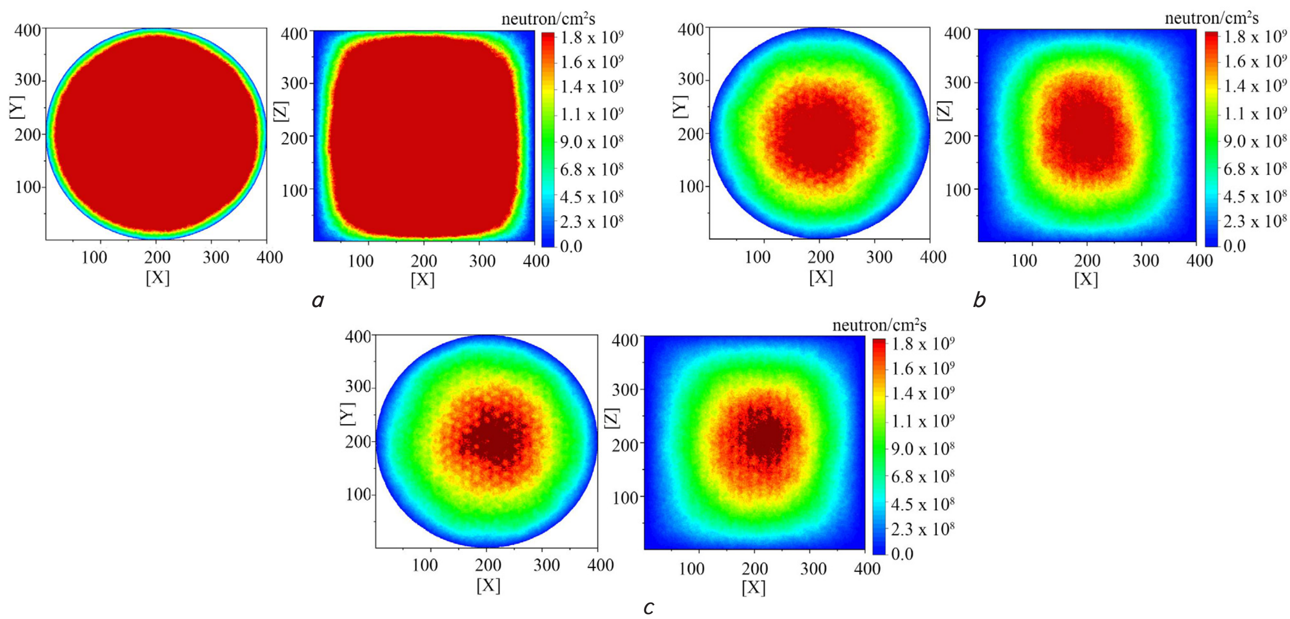

Fig. 9. Distribution of neutron flux at the end of life: *a* – fuel 1; *b* – fuel 2; *c* – fuel 3

Fig. 8, 9 show the distribution of neutron flux at the beginning of operation (BOL) and the end of operation (EOL). Based on Fig. 8, 9, fuel 1 has the widest maximum flux distribution at EOL and BOL. The distribution of neutron flux in fuel 2 and fuel 3 is almost the same and has a much different total area from fuel 1 in the BOL and EOL conditions.

6. Discussion on the composition of uranium-based fuels that have the potential to replace thorium-based fuels

Based on the research results, fuel 2 and fuel 3 have almost the same characteristics and have the greatest potential to become MSR FUJI-12 fuel for 350 MWt power operation for 5 years. Fuel 1 has a flux value that is too high, so that fissile fuel (^{235}U) is not enough for reactor operation for 5 years (Fig. 5). The composition of the eutectic mixture of LiF-BeF$_2$-UF$_4$ for fuel 1 also produces a larger amount of ^{3}H (Fig. 6). LiF turns into hydrogen fluoride (^{3}HF) during the reactor operation. The material is very corrosive and dangerous for the reactor fuel pipe [17]. The optimization results also show that only fuel 2 and fuel 3 were able to reach the optimal state for 350 MWt power operation for 5 years (Fig. 4). Fuel 2 requires a minimum of 8 % ^{235}U enrichment and fuel 3 – a minimum of 7 % ^{235}U enrichment. Fuel 1 cannot reach the optimal state for each variation of ^{235}U enrichment. This is because the total amount of uranium in fuel 1 is very small. Uranium is the main material in fission reactions. Fuel 1 requires more ^{235}U enrichment to last 5 years. However, ^{235}U enrichment that is too large can increase excess reactivity and potentially cause the reactor to explode due to an uncontrolled fission chain reaction.

Based on Fig. 8, 9, the distribution of the maximum neutron flux is spread over the center of the reactor either on the *XY* or *XZ* axes. The distribution characteristics for fuel 2 and fuel 3 are almost the same, in contrast to fuel 1, which has a maximum neutron flux distribution with a larger area. The difference is very visible in the state of BOL and EOL. Larger maximum neutron flux distribution in fuel 1 indicates greater fission production or burning of almost all fissile materials. In contrast to fuel 2 and fuel 3, which are better at maintaining the fissile material so that the maximum neutron flux can be maintained at the center of the reactor core.

Neutronic analysis in molten salt reactor FUJI-12 using OpenMC 0.13.0 shows that fuel 2 and fuel 3 have potential neutronic parameters to replace thorium-based fuel. These results need to be developed further because the simulation method with OpenMC 0.13.0 uses a static system. MSR is a reactor with fuel that is constantly moving, so it needs further analysis with a dynamic system. Combination with thermal-hydraulic analysis needs to be done as a comprehensive step in the nuclear reactor safety analysis.

7. Conclusions

1. The eutectic composition of LiF-BeF_2-UF_4 in fuel 2 and fuel 3 has the potential to be used as a liquid salt fuel mixture in MSR FUJI-12 with an operating power of 350 MWt for 5 years.

2. The distribution of maximum neutron flux in fuel 2 and fuel 3 is more stable than in fuel 1 for both BOL and EOL conditions.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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