Concept of a Fast Breeder Reactor To Transmute MA and LLFP

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Concept of a fast breeder reactor to transmute MA and LLFP

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Abstract

The long-term issues of nuclear power systems are the effective use of uranium resources and the reduction of radioactive waste. Important radioactive wastes are minor actinides (MA: ^{237}Np, ^{241}Am, ^{243}Am, etc.) and long-lived fission products (LLFP: ^{129}I, ^{99}Tc, ^{79}Se, etc.). The purpose of this study was to show a system that can simultaneously achieve the breeding of fissile materials and the transmutation of MA and LLFP in one fast reactor. Transmutation was carried out by loading innovative Duplex type MA fuel in the core region and LLFP containing moderator in the first layer of the radial blanket. Breeding was achieved in the core and axial blanket. As a result, it was clarified that in this fast breeder reactor, a breeding ratio of about 1.1 was obtained, and MA and LLFP achieved a support ratio of 1 or more. The transmutation rate was 10.3%/y for ^{237}Np, 14.1%/y for ^{241}Am, 9.9%/y for ^{243}Am, 1.6%/y for ^{129}I, 0.75%/y for ^{99}Tc, and 4%/y for ^{79}Se. By simultaneously breeding fissile materials and transmuting MA and LLFP in one fast reactor, it will be possible to solve the long-term issues of the nuclear power reactor system, such as securing nuclear fuel resources and reducing radioactive waste.

Introduction

Important issues for nuclear power generation are the effective use of uranium resources and the reduction of radioactive waste while ensuring safety. For the effective use of uranium resources, the fast breeder reactor (FBR) can convert ^{238}U to fissile material, such as ^{239}Pu, with a breeding ratio of 1 or more.

In order to reduce radioactive waste, it is important to reduce the amounts of minor actinides (MA: ^{237}Np, ^{241}Am, ^{243}Am, etc.) and long half-lived fission products (LLFP: ^{99}Tc, ^{129}I, ^{79}Se, etc.). In back-end research, MA is the major element of the potential toxicity of the radioactive waste. By recovering and transmuting minor actinide nuclides, it is thought that the potential toxicity after 1000 years can be reduced to 1/100. In addition, the removal of MA is said to be effective in reducing the area of the disposal site. On the other hand, LLFP is regarded as important from the viewpoint of the radiation safety performance (future exposure dose to the public) of the disposal site.

Many studies have been conducted on the transmutations of MA and LLFP in fast reactors[1-31]. Regarding MA, it is known that if an average of about 5% of MA was added to the fuel region, a transmutation rate of 10%/y or more was achieved without significantly affecting the core characteristics[7,13].

Regarding LLFP, a transmutation study has recently been conducted on 6 important nuclides (^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{129}I, and ^{135}Cs) from the viewpoint of reducing environmental influence. As a result, it was shown that the fast reactor can be used to transmute each of these 6 nuclides with a support ratio exceeding 1 using the YD_2 moderator[28]. The significant reduction in the effective half-life was obtained by analysis. In this case, the six nuclides were used as elements without isotope separation. In addition, a method that can transmute six nuclides at the same time with a support ratio of 1 or more in one fast reactor was clarified[29]. In this case, ^{135}Cs and ^{93}Zr, both with small neutron absorption cross sections were placed in the radial blanket region, and ^{129}I and ^{99}Tc, both with large neutron absorption cross sections were placed in the shield region and axial blanket away from the fuel region Therefore, the transmutation rate of all nuclides was less than 0.5%/y. On the other hand, a study on a method designed to achieve a high transmutation rate (about 8%/y) for four nuclides (^{79}Se, ^{99}Tc, ^{107}Pd, ^{129}I) was conducted[30]. Based on these studies, a system that further improves the transmutation efficiency of the six nuclides was explored. A fast reactor LLFP transmutation system that achieves a support ratio of 1 or more for the entire system was constructed by combining three fast reactors, in addition to using one reactor[31]. From these studies, a significant amount of information about the LLFP transmutation system was obtained.

Among these LLFP nuclides, ^{129}I presents a long-term radioactivity problem in geological disposal as a long-lived nuclide that is soluble and less absorbed by underground materials[32]. ^{99}Tc is the main radioisotope of vitrified radioactive waste, and its potential toxicity is a problem. ^{79}Se has been a determinant of radiation exposure over the period of 10^4-10^5 years. Reducing these three nuclides could reduce the uncertainty of geological disposal. Therefore, these three nuclides were selected as LLFP for transmutation in this study.

If breeding of fissile materials and transmutation of MA (^{237}Np, ^{241}Am, ^{243}Am) and LLFP (^{129}I, ^{99}Tc, ^{79}Se) can be performed simultaneously in one fast reactor, the long-term issues of securing nuclear fuel resources for nuclear power systems and reducing radioactive waste would be solved. In addition, solution of these issues would lead to a better understanding of nuclear power generation among the general public. The purpose of this study was to show a system that simultaneously achieves the breeding of fissile materials and the transmutation of MA and LLFP in one fast breeder reactor.

Here, the support ratio (SR) was defined as the ratio of the amount of each nuclide transmuted by the fast breeder reactor to the amount of each nuclide (MA and LLFP) produced by the fast breeder reactor[28,30,31].

Results and discussions

Studies on MA transmutation in fast reactors have investigated homogeneous loading, in which MA is homogeneously added to the core fuels, and heterogenous loading, in which assemblies of only MA are loaded in the core in a dispersed manner. In the case of homogeneous loading, the effect on the power distribution in the core is small, but due to the strong radiation of MA, large-scale shielded cells were required to prevent exposure during manufacturing. In the case of heterogenous loading, the MA assemblies with large neutron absorption cross sections and the normal MOX fuel assemblies were mixed, so the difference in power among these was large and it was difficult to control the power distribution in the core.

Therefore, Duplex type MA fuel was proposed as an innovative alternative. The concept of the MA fuel is to insert the MA pellet into the center of the hollow MOX pellet, as shown in Fig.1 (a). This is called a Duplex pellet. Normal hollow MOX pellets can be manufactured in glove boxes that do not require significant shielding to prevent high radiation exposure. The central MA pellets are manufactured and inserted into the Duplex pellets in small shielding cells. Since hollow MOX pellets can be manufactured in glove boxes, it is expected that the manufacturing equipment will be simplified compared to MA homogeneous fuels, leading to a reduction in manufacturing costs. Figure. 1 (a) shows the structure of the MA-containing MOX fuel assembly. The number of MA fuel pins in the MA fuel assembly was 271. The MA content of the central MA pellet was set to 20 wt% so that the average of MOX and MA pellets was 5 wt%. The MA containing MOX fuel assemblies were loaded in the inner and outer cores. Figure 2 shows the arrangement of MA-containing fuel assemblies in the fast breeder reactor core. Regarding the fabrication of Duplex pellets, studies were conducted on the effects of Gd_2O_3 placed in the center of oxide fuel pins in LWR[33,34]. In addition, by using Duplex pellets containing neutron absorbers in fast reactors, a new core concept has been proposed that did not achieve re-criticality in case of fuel melting in an accident[35]. From these studies, it is considered that the innovative Duplex type MA fuel used in MA transmutation is sufficiently feasible.

Regarding the transmutation of LLFP, LLFP assemblies in which moderators[36-40] (YH_2, YD_2, etc.) were combined with LLFP nuclides have been studied in order to improve the transmutation performance of LLFP. When an LLFP assembly that combines LLFP and YH_2 moderator is loaded in the blanket region, YH_2 has a high moderating ability, so the transmutation rate increases. However, an increase in the amount of thermal neutrons would create a thermal spike by causing the power of the adjacent fuel assembly to increase. In order to prevent a thermal spike, in the case of ^{129}I transmutation, ^{99}Tc metal pins were installed in the outermost two layers and ^{129}I pins containing a mixture of BaI_2 and moderator YH_2 were installed in the inner part of the assembly. As a result, the thermal neutrons emitted from the fuel regions are absorbed by ^{99}Tc, which has a large neutron absorption cross section, so that a thermal spike of the adjacent fuel can be reduced. Figure 1 (b) shows the arrangement of the ^{99}Tc and ^{129}I pins in the assembly. As shown in Fig. 2, 86 assemblies of ^{99}Tc and ^{129}I were loaded in the first layer of the blanket region.

For ^{79}Se, it is known that the transmutation rate did not change significantly regardless of whether the moderator is YD_2 or YH_2; therefore, YD_2 was used to address the issue of thermal spike. As shown in Fig. 1 (c), 169 pins in the form of mixed ZnSe[41,42] and YD_2 were arranged in the assembly[30,31]. As shown in Fig. 2, ten ^{79}Se assemblies were placed in the first layer of the blanket region. This is because the amount of ^{79}Se produced in the fast breeder reactor is as small as 0.22 kg/y.

It is considered possible to achieve the breeding of fissile materials by the axis blanket, as significant breeding by the radial blanket cannot be expected because the LLFP assemblies are loaded in the first layer of the radial blanket. The thickness of the upper and lower axial blankets were 30 cm and 40 cm, respectively. Figure 3 shows a cross-sectional view of the core.

Table 1 shows the analysis results of MA transmutation. The composition of the loaded MA nuclides was based on the composition of fuel discharged from the fast breeder reactor, as shown in Table 2, and the composition of fuel discharged from a light water reactor was also studied as a reference. ^{244}Cm was excluded from the loaded MA nuclides because it has a short half-life of 18.1 years and Cm element can be separated from MA[43]. Since the MA composition of the fuel discharged from the fast breeder reactor changes due to transition from Pu, Am nuclides increase. On the other hand, since the MA composition of fuel discharged from the LWR changes due to transition from U, ^{237}Np increases. In the case of the MA composition of fuel discharged from the fast breeder reactor, it was found that a support ratio of 1 or more can be achieved for each of the three MA nuclides. The transmutation rate was 10.3%/y for ^{237}Np, 14.1%/y for ^{241}Am, and 9.9%/y for ^{243}Am. Since the transmutation characteristics are excellent, it is considered that the fast breeder reactor system can transmute MA flexibly according to the MA inventory. In the case of the MA composition of the fuel discharged from LWR, the support ratio was as large as 25.6 for ^{237}Np, but it was just 1.0 for ^{243}Am. Regarding the transmutation rate, ^{237}Np and ^{241}Am were greater than 10%/y, but ^{243}Am was as small as 5.8%/y. When using MA with the composition of the fuel discharged from LWR, it is considered that efficient MA transmutation can be achieved by mixing it with the MA composition of fuel discharged from the fast breeder reactor.

Table 3 shows the analysis results of LLFP transmutation. The SR is 1.13 for ^{99}Tc, 1.24 for ^{129}I, and 3.3 for ^{79}Se, all exceeding 1. Regarding the transmutation rate, ^{99}Tc was 0.79%/y, ^{129}I was 1.24%/y, and ^{79}Se was 3.3%/y. The transmutation rates of ^{99}Tc and ^{129}I were lower than the values obtained for 300 MWe class fast reactors (^{99}Tc: 2.47%/y, ^{129}I: 3.41%/y). This is because the leakage of neutron flux in the radial direction is reduced due to the increase in the core diameter. Since the number of loaded assemblies of ^{79}Se is as small as 10, the transmutation rate of ^{79}Se was not significantly affected by the size of the core.

Table 4 shows a comparison of the breeding ratios between the fast breeder reactor loaded with MA and LLFP and a normal large-scale fast breeder reactor (with radial blanket). Compared with a normal large-scale fast breeder reactor, the breeding ratio of the fast breeder reactor loaded with MA and LLFP was slightly lower, but it was found that about 1.1 of the breeding ratio could be obtained. This is because the contribution of the breeding ratio of the axial blanket in a large-scale fast breeder reactor is larger than that of the radial blanket; and the breeding ratio of the entire core does not decrease significantly even if the first layer of the radial blanket is replaced with the LLFP assemblies.

The sodium void reactivity and the Doppler coefficients related to the safety of the fast breeder reactor loaded with MA and LLFP were analyzed. Table 5 shows a comparison of the sodium void reactivity and the Doppler coefficient between the fast breeder reactor loaded with MA and LLFP and a normal large-scale fast breeder reactor. The sodium void reactivity of the fast breeder reactor loaded with MA and LLFP was about 30% higher than that of the normal large-scale fast breeder reactor. The absolute value of the Doppler coefficient was about 40% smaller. This is because the neutron spectrum of the core became harder due to the addition of MA. However, it is considered that there is no major safety problem with such changes in the Doppler coefficient and sodium void reactivity.

Regarding MA, the difference in transmutation rate between MA-containing Duplex pellets and homogeneous MA pellets was analyzed. As shown in Table 6, it was found that there was no significant difference in transmutation rate between MA-containing Duplex pellets and homogeneous MA pellets, and it was found that MA-containing Duplex pellets were effective. Since the energy spectrum of neutrons is hard in the core, the self-shielding effect by inserting MA in the central region is considered to be small.

Conclusions

From this study, the new concept of fast breeder reactor system that can transmute MA (^{237}Np, ^{241}Am, ^{243}Am) and LLFP (^{129}I, ^{99}Tc, ^{79}Se) with a support ratio of 1 or more was constructed while breeding fissile materials in one fast breeder reactor. It was clarified that this fast breeder reactor achieved a breeding ratio of about 1.1, and MA and LLFP support ratios of 1 or more. The transmutation rate was 10.3%/y for ^{237}Np, 14.1%/y for ^{241}Am, 9.9%/y for ^{243}Am, 1.6%/y for ^{129}I, 0.75%/y for ^{99}Tc, and 4%/y for ^{79}Se.

Based on these studies, the following can be considered impacts on nuclear power development, effective utilization of uranium resources, and reduction of radioactive waste.

- By simultaneously breeding fissile materials and transmuting MA and LLFP in one fast reactor, it will be possible to solve the long-term issues of the nuclear power systems, such as securing nuclear fuel resources and reducing radioactive waste. In addition, solving these issues would promote a better understanding of nuclear power systems among the general public.
- The ability to breed fissile materials and transmute MA and LLFP in one fast breeder reactor shows

the high potential of the fast breeder reactor and will promote research and development of the fast breeder reactor.

- This new system can contribute to the effective utilization of uranium resources and the reduction of radioactive waste without significantly changing the conventional nuclear fuel cycle system.

Method

Core conditions

The fast breeder reactor used in this study is a large sodium-cooled fast breeder reactor designed for the commercial stage. Table 7 shows the main specifications. The thermal power of the reactor is 3570 MWt and the electric power is 1500 MWe. The core was a homogeneous two-region core, with 316 MA-containing fuel assemblies in the inner core, 278 MA-containing fuel assemblies in the outer core, and 55 control rods. The outside of the core was composed of 96 LLFP assemblies, 102 radial blanket assemblies, and 222 radial shielding assemblies. The height of the core is 80 cm. The core equivalent diameter is 490 cm. The Pu enrichment of the inner and outer cores are 20.7 W% and 23.3 wt%, respectively. The Pu enrichment of the outer core is higher than that of the inner core to achieve power flattening. Table 8 shows the specifications of the MA-containing fuel assembly and the LLFP assembly. There are 271 MA-containing fuel pins in the fuel assembly and 169 LLFP pins in the LLFP assembly. The isotopic compositions of MA and LLFP loaded in the fast breeder reactor shown in Tables 2 and 9 were based on the results of 80 GWd/t burnup simulation in the fast breeder reactor by MVP-BURN code. The MA composition of the discharged LWR fuel was based on the results of a 40 GWd/t burnup simulation of LWR UO_2 fuel.

Calculational method

Core characteristics were analyzed with a continuous neutron energy Monte Carlo code MVP[44] with JENDL-4.0[45] neutron cross section library. The number of neutron histories was 10,000, the number of batches skipped for accurate source distribution was 100, and the number of effective batches was 1,000. From this Monte Carlo simulation, the neutron energy spectra and the reaction rates of MA and LLFP in various regions of the fast breeder reactor were obtained. The typical statistical error for k-effective was about 0.015% with a 1σ error. The statistical errors of the MA reaction rate in the core fuel and the reaction rate of the LLFP in the LLFP assembly were also sufficiently low, ranging from 0.1 and 0.5%. Burnup calculations were performed with the MVP-BURN code[46].

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Fuel composition	MA abundance of discharged FBR fuel			MA abundance of discharged LWR fuel		
Nuclide	^{237}Np	^{241}Am	^{243}Am	^{237}Np	^{241}Am	^{243}Am
Transmutation rate (%/y)	10.3	14.1	9.9	12.4	14.0	5.8
SR	4.6	6.3	4.4	25.6	4.6	1.0

Table 1 Transmutation rate and support ratio of MA

	Fuel composition		Half-life (year)
	MA abundance of discharged FBR fuel (%)	MA abundance of discharged LWR fuel (%)	
^{237}Np	11.3	50.5	2,144,000
^{241}Am	51.3	37.0	433
^{243}Am	37.4	12.5	7370

Table 2 Isotope abundance and half-life of loaded MA nuclides

	^{99}Tc	^{129}I	^{79}Se
Transmutation rate(%/y)	0.75	1.4	4.0
SR	1.13	1.24	3.3

Table 3 Transmutation rate and support ratio of LLFP in fast breeder reactor with MA and LLFP

Core arrangement	FBR with MA and LLFP	FBR without MA and LLFP
Core	0.76	0.76
Axial blanket	0.32	0.32
Radial blanket	0.01	0.11
Total	1.09	1.19

Table 4 Comparison of breeding ratios between with and without MA and LLFP

Core arrangement	FBR with MA and LLFP	FBR without MA and LLFP
Sodium void reactivity ($)	6.8	5.2
Doppler coefficient (Tdk/dT)	-4.0E-3	-6.6E-3

Table 5 Characteristics of sodium void reactivity and Doppler coefficient in the core of fast breeder reactor

	Transmutation rate (%)	
Pellet arrangement	Duplex pellet with MA in the center region (MA:20%)	Homogeneous pellet with MA (MA:5%)
^{237}Np	14.1	14.3
^{241}Am	10.3	10.5

Table 6 Comparison of transmutation rate between Duplex pellet and homogeneous pellet

Thermal power (MWt)	3570
Electric power (MWe)	1500
Core type	Homogeneous two-region core
Operation cycle length (months)	18
Number of refueling batches (Core/Blanket)	4/4
Core height (cm)	80
Thickness of axial blanket (cm) (Upper／Lower)	30/40
Number of core fuel assemblies (Inner/Outer/Total）	316 / 278 / 594
Number of LLFP assemblies	96 (^{129}I and ^{99}Tc assembly:86, ^{79}Se assembly:10)
Number of radial blanket assemblies	102
Pu enrichment (wt%) (Inner/Outer core)	20.7/23.3
Number of control rods （Main/backup）	40 / 15
Number of radial shielding assemblies	108 /114
Volume ratio of core (Fuel/Structure/Coolant)	44.1/24.2/31.7

Table 7　Specification of large-scale fast breeder reactor for MA and LLFP transmutation

	MA-containing fuel assembly	LLFP assembly
Pin diameter (mm)	8.8	11.5
Thickness of pin (mm)	0.52	0.5
Pellet diameter (mm)	7.6	10.3
Number of pins in the assembly	271	169

Table 8 Specifications of MA-containing fuel assembly and LLFP assembly

Isotopes of loaded LLFP elements	Abundance (%)	Half-life of LLFP (year)
^{76}Se	0.027	
^{77}Se	2.786	
^{78}Se	5.587	
^{79}Se	13.32	295,000
^{80}Se	22.75	
^{82}Se	55.52	
^{99}Tc	100.00	211,000
^{127}I	23.91	
^{129}I	76.09	15,700,000

Table 9 Isotope abundance and half-life of loaded LLFP nuclides

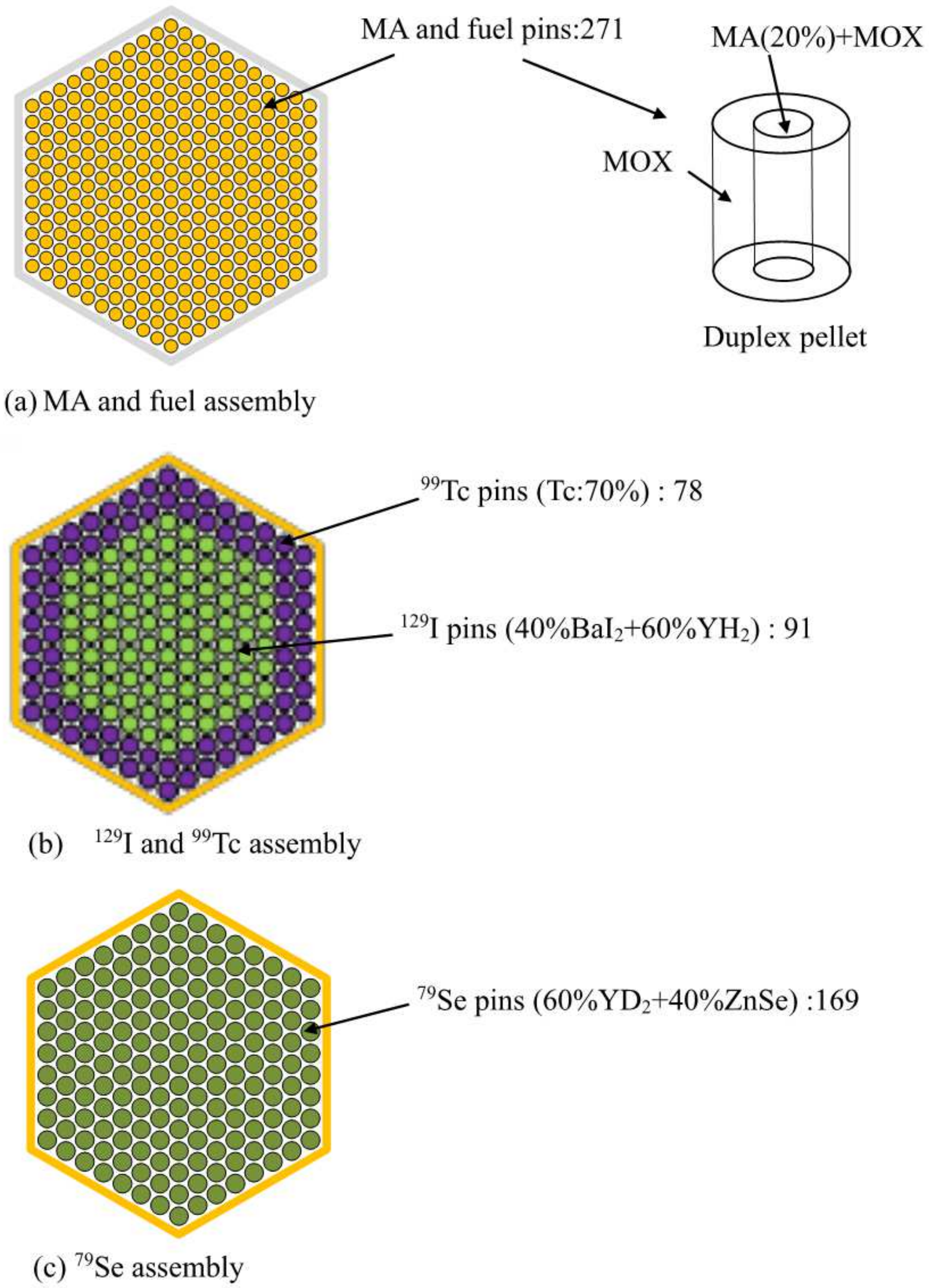

Fig.1 Configurations of MA fuel assembly, ^{129}I and ^{99}Tc assembly, and ^{79}Se assembly

(a) the structure of the MA-containing MOX fuel assembly. The number of MA fuel pins in the MA-containing fuel assembly is 271. (b) the arrangement of the ^{99}Tc pins and the ^{129}I pins in the assembly. Eighty-six ^{99}Tc and ^{129}I assemblies are loaded in the first layer of the blanket region. (c) 169 pins in the form of mixed ZnSe and YD_2 are arranged in the assembly. Ten ^{79}Se assemblies are placed in the first layer of the blanket region.

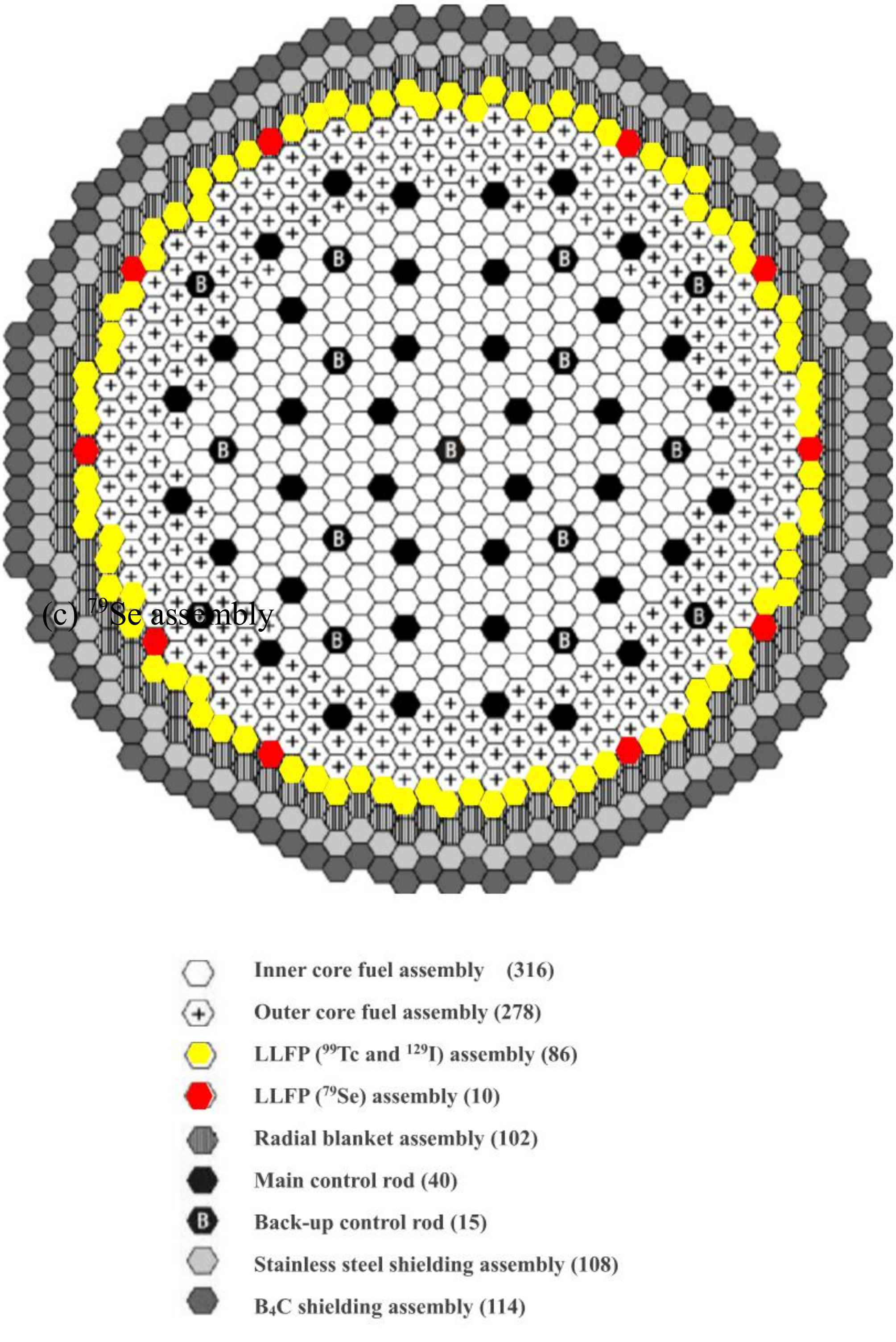

Fig.2 Core arrangement for MA and LLFP transmutation in fast breeder reactor

The core has two homogeneous zones: inner and outer cores. MA fuel assemblies are loaded in the inner and outer cores. The LLFP assemblies are loaded in the first layer of the blanket region. The blanket fuel assemblies are loaded in the second layer of the blanket region.

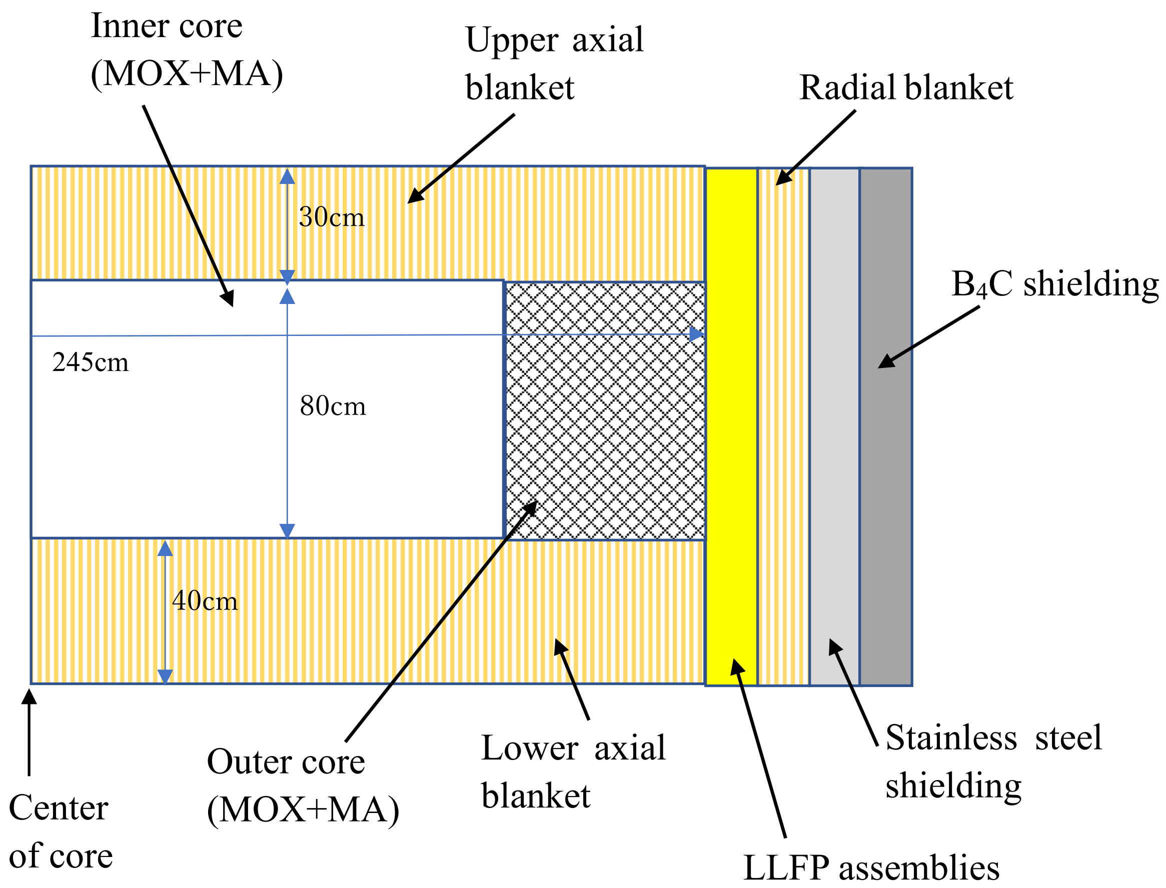

Fig.3 Axial core arrangement for MA and LLFP transmutation in fast breeder reactor

The height of the core is 80 cm. The thicknesses of the upper and lower axial blankets are 30 cm and 40 cm, respectively. The core equivalent diameter is 490 cm.

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Author Contributions

T.W. conceived the study. T.W. designed the transmutation system and computational simulations. T.W. carried out computational simulations and analyses.

Additional Information

Competing Interests: The author declares no competing interests.

Figure legends

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Figures

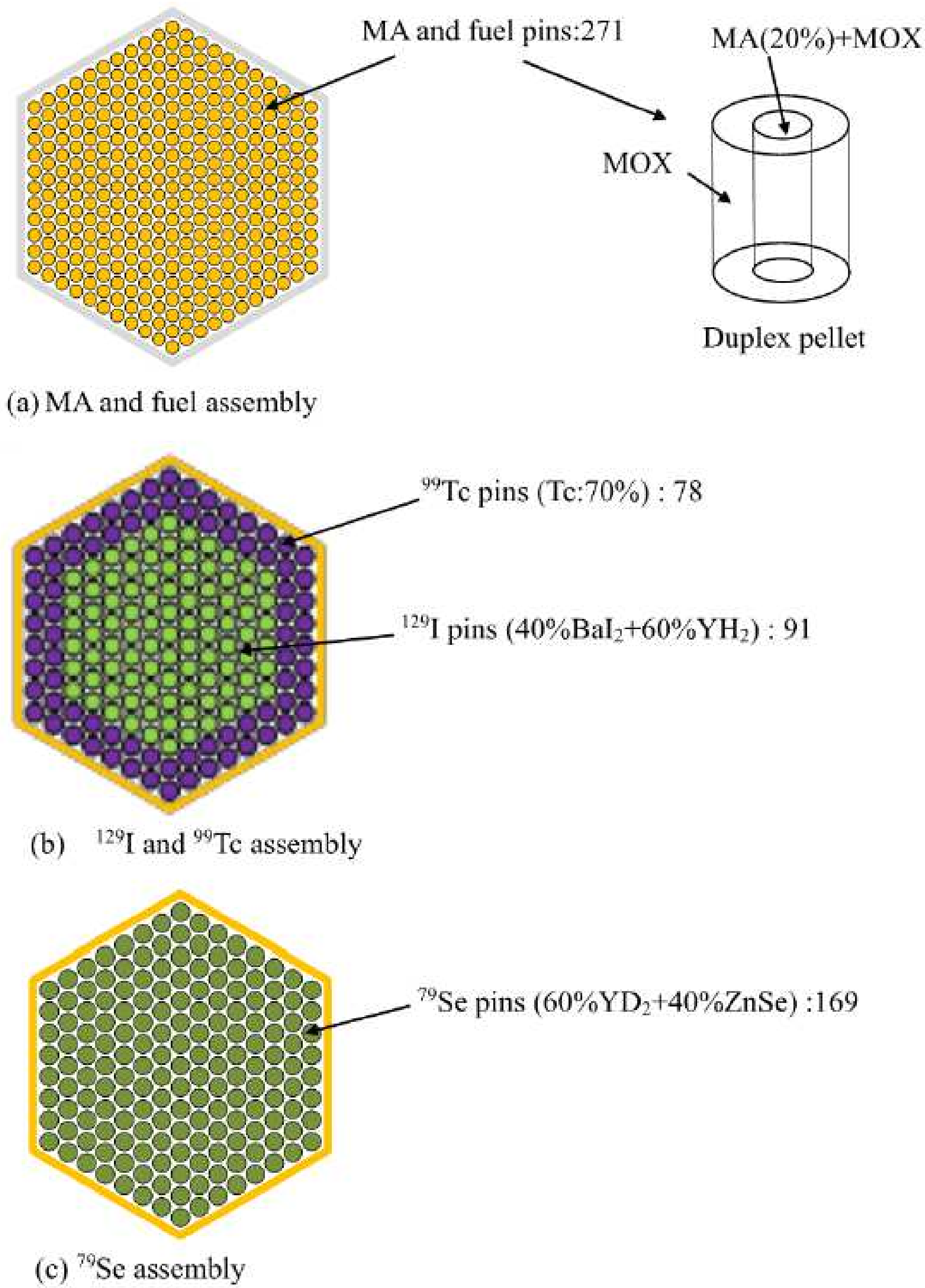

Figure 1

Configurations of MA fuel assembly, 129I and 99Tc assembly, and 79Se assembly (a) the structure of the MA-containing MOX fuel assembly. The number of MA fuel pins in the MA-containing fuel assembly is 271. (b) the arrangement of the 99Tc pins and the 129I pins in the assembly. Eighty-six 99Tc and 129I assemblies are loaded in the first layer of the blanket region. (c) 169 pins in the form of mixed ZnSe and YD2 are arranged in the assembly. Ten 79Se assemblies are placed in the first layer of the blanket region.

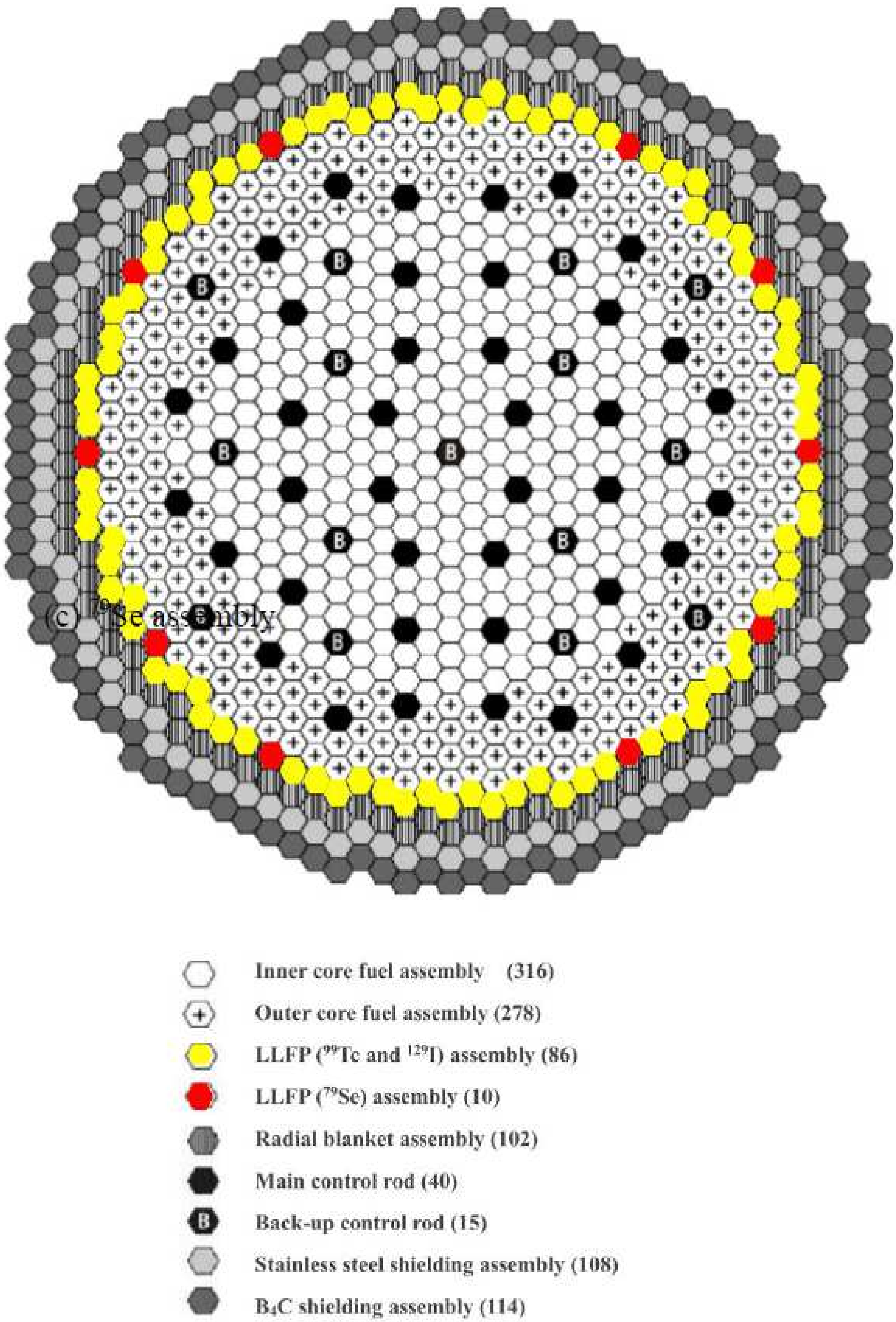

Figure 2

Core arrangement for MA and LLFP transmutation in fast breeder reactor The core has two homogeneous zones: inner and outer cores. MA fuel assemblies are loaded in the inner and outer cores. The LLFP assemblies are loaded in the first layer of the blanket region. The blanket fuel assemblies are loaded in the second layer of the blanket region.

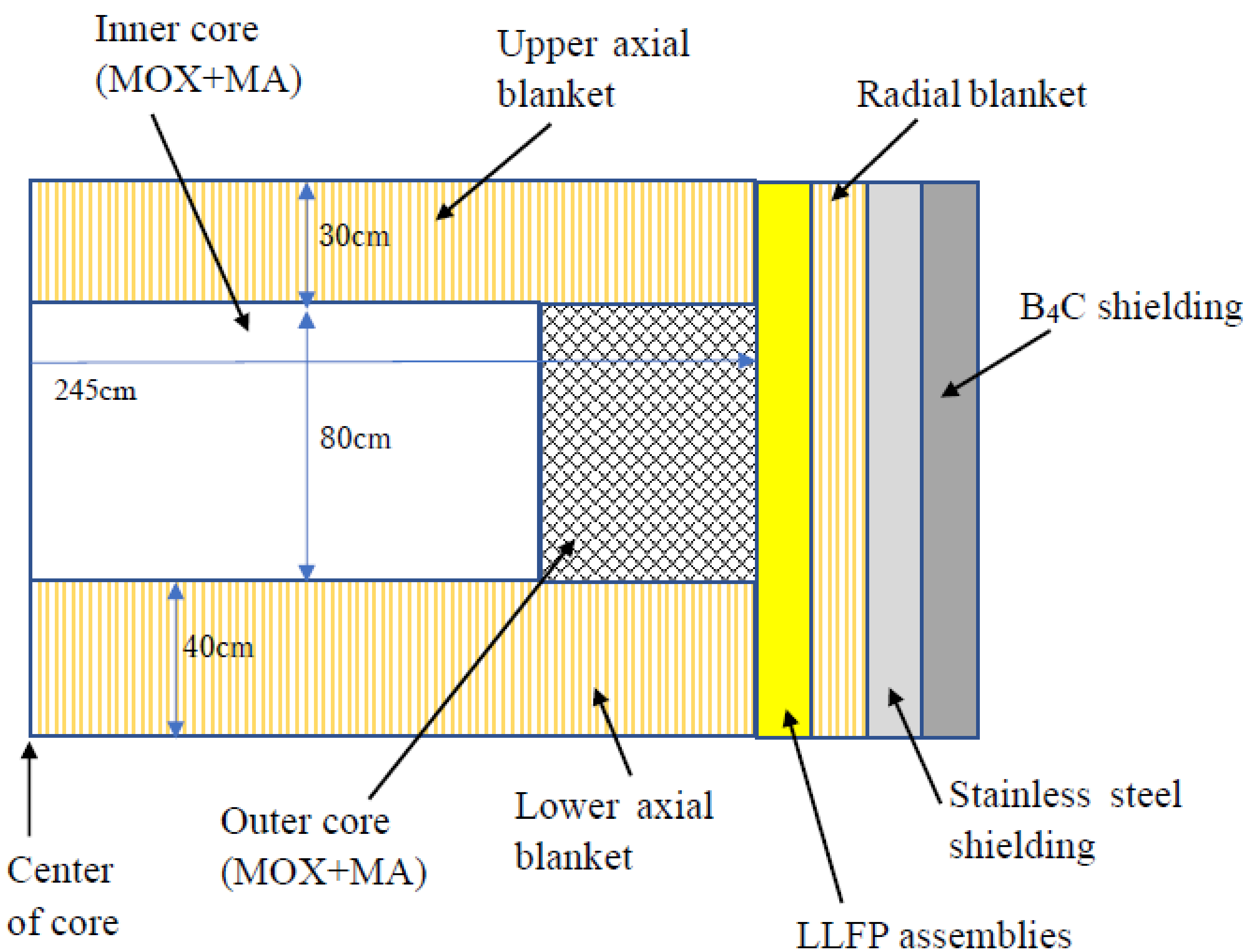

Figure 3

Axial core arrangement for MA and LLFP transmutation in fast breeder reactor The height of the core is 80 cm. The thicknesses of the upper and lower axial blankets are 30 cm and 40 cm, respectively. The core equivalent diameter is 490 cm.