Study on Nuclear Transmutation of Nuclear Waste by 14 MeV Neutrons*)

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The object of this study is to clarify the possibility of 14 MeV neutrons to decrease the radioactivity of nuclear waste, especially fission products such as Cs, Sr, Tc, and I. Simulations were performed for several fission products by Monte-Carlo code and burnup calculation code. In comparison to the spectra of thermal reactor and fast reactor, it was found that D-T neutron of 14 MeV is preferable and has the possibility to reduce the radioactivity of fission products such as Cs, Sr, Tc, and I with the magnitude of the flux more than 5×10^{15} [#/cm^2/sec] for 1000 days irradiation period. This investigation reveals the possibility and the superiority of 14 MeV neutron than thermal and fast neutron spectra to annihilate the fission products.

Keywords: nuclear waste, fission product, nuclear transmutation, 14 MeV neutron, D-T reaction

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1. Introduction

Nuclear waste disposal is a big problem not only in Japan but also all over the countries using nuclear power plants. Nuclear wastes are categorized into two materials: fission products (FPs) and minor actinides (MAs). There are some choices how to dispose them, and nuclear transmutation by neutrons with reprocessing has been investigated to minimize the burden for geological disposal. Transmutation is a phenomenon that some nuclide changes into another nuclide. Our considering scenario consists of the followings; MAs, such as Np and Am, are transmuted in Fast Reactor (FR) and/or Accelerator Driven Subcritical Reactor (ADS) as fissionable materials. FPs are vitrified after several decades storage so as to decrease their radioactivity during the period. The main isotopes of FPs radioactivity are ^{90}Sr, ^{137}Cs and their daughters for several decades, but, ^{99}Tc and ^{129}I for several millions years. The main target isotopes of this study to annihilate by 14 MeV neutrons are ^{90}Sr and ^{137}Cs in order to reduce the radioactivity and shorten the storage period. The evaluations are also performed for ^{99}Tc and ^{129}I for the comparison to ^{90}Sr and ^{137}Cs. The transmutation rate of these isotopes by neutron irradiation depends on several neutronic parameters described below, and the possibility of 14 MeV neutrons for the transmutation has been investigated in detail in this study.

2. Neutronic Parameter

2.1 Burnup chain

Figure 1 shows an example of burnup chain related to ^{90}Sr. Burnup chain shows the relation among many isotopes; ^{90}Sr is transmuted to ^{91}Sr by neutron capture reaction, ^{89}Sr by (n, 2n) reactions, ^{90}Sr decays into ^{90}Y with 29 years half-life of beta-ray emission, etc.

^{90}Sr has its half-life of 29 years, but the surrounding nuclides shown in Fig. 1 have relatively shorter half-lives than that of ^{90}Sr. This tendency shows the possibility to accelerate the reduction of storage period by nuclear transmutation, and this tendency is almost the same for ^{137}Cs (see Fig. 2).

2.2 Cross section

Each isotope has its own reaction cross section for neutrons as shown in Fig. 3. Figure 3 shows the cross sec-

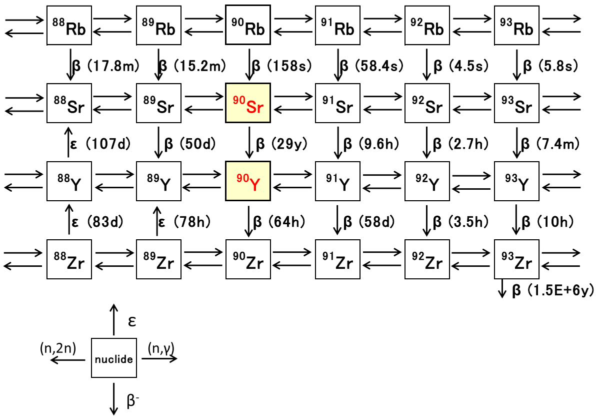

Fig. 1 Burnup chain related to ^{90}Sr.

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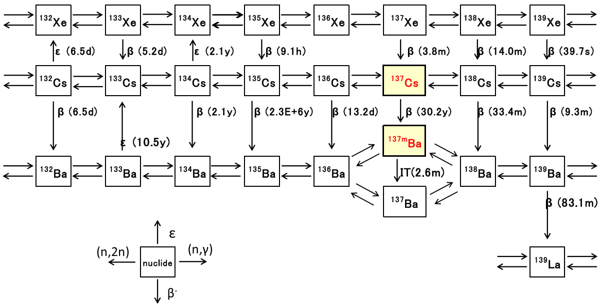

Fig. 2 Burnup chain related to ^{137}Cs.

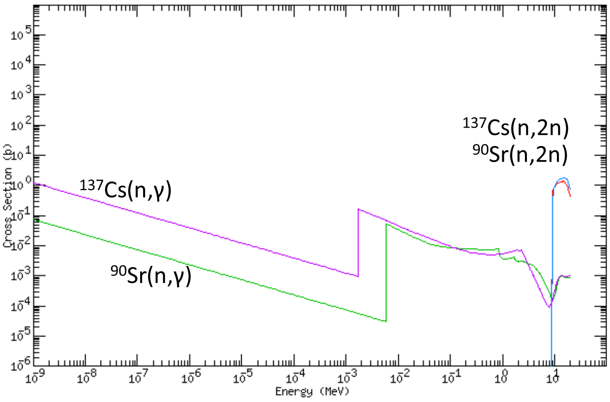

Fig. 3 Cross sections of ^{90}Sr and ^{137}Cs.

tions of ^{90}Sr and ^{137}Cs for capture and (n, 2n) reactions. The value of cross section depends on the energy of irradiated neutron, and the dependency in remarkably different among the reaction types such as capture and (n, 2n) reactions. Most of all isotopes have the same tendency for capture reaction cross section which is larger in lower energy of irradiating neutron, and for (n, 2n) reaction cross section which has the threshold around 10 MeV. This shows that (n, 2n) reaction is remarkable if 14 MeV neutron is used for the irradiation, and this point is the characteristics of this study from the former investigations.

In this investigation, three kinds of neutron spectrum are used as shown in Fig. 4. Those are the spectra of thermal reactor named PWR, fast reactor named FBR, and 14 MeV neutron named D-T.

3. Calculation Conditions

The magnitude of neutron flux affects the transmutation, as easily expected. In this study, the magnitude was changed as 5×10^{13}, 5×10^{14}, 5×10^{15} and 5×10^{16} [#/cm^2/sec] to consider the necessary magnitude for the effective transmutation.

Irradiation period was set to 1000 days, and the following cooling period was set to 3500 days to consider rapid decreasing of short-lived radioactivity produced during the irradiation. Non-irradiated case named “cooling only” was also simulated for the comparison.

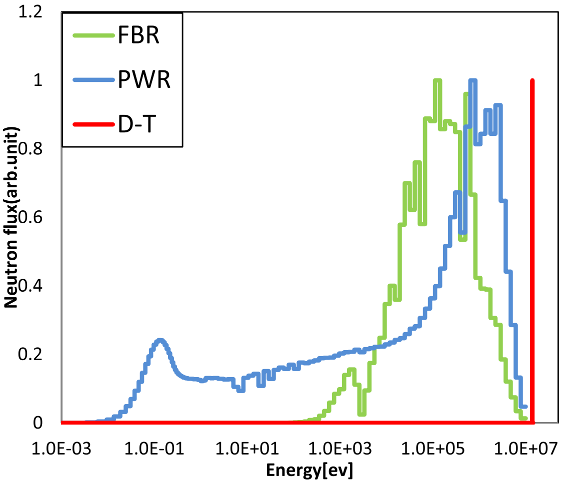

Fig. 4 Irradiated neutron spectrum.

Table 1 Composition of the target.

	mass ratio		radioactivity ratio
	Nuclear waste	FP	
U, Pu	97.04%		21.6%
Np, Am, Cm	0.10%		0.5%
Cs (Ba)	0.27%	9.36%	**38.8%**
Sr (Y)	0.09%	3.00%	**27.2%**
Tc	0.08%	2.87%	0.0%
I	0.02%	0.71%	0.0%
others	2.41%	84.06%	11.9%

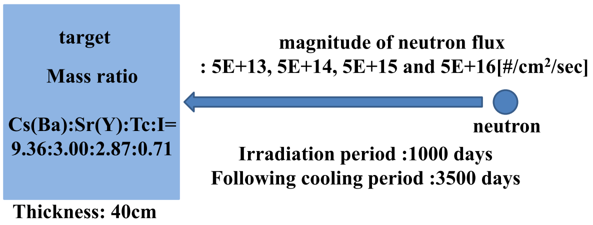

Fig. 5 Calculation conditions.

The target elements are Cs(Ba), Sr(Y), Tc, and I without considering isotope separation, and the mass composition of the target material is (Cs(Ba) : Sr(Y) : Tc : I) = (9.36 : 3.00 : 2.87 : 0.71) as shown in Table 1. This mass ratio was provided from PWR spent fuel (33 GWd/tU) cooled for 7 years. The mass ratio of 4 elements in nuclear waste is trivial but the radioactivity of them is remarkable as also shown in Table 1. 14 MeV, PWR and FBR neutrons are irradiated to the target materials as shown in Fig. 5. The irradiated neutron spectra of PWR or FBR are also simulated to check the effectiveness of 14 MeV neutrons. Although the neutron spectrum of fusion reactor is much softer than 14 MeV mono energetic spectrum, this study concentrated on revealing the effectiveness of 14 MeV neutrons on nuclear transmutation of nuclear waste.

Calculations were done by using continuous energy Monte Carlo code named MVP-II [1] to evaluate reaction rates of each nuclide, and burnup simulation code named ORIGEN2 [2] to evaluate the change of the target composition during the irradiation and/or cooling period. The cross section library used in this study is based on JENDL-4.0 [3].

4. Results and Discussions

Tables 2 and 3 show the change in radioactivity ratio to the original (before irradiation) for ^{137}Cs/^{137m}Ba and ^{90}Sr/^{90}Y, by changing spectrum and magnitude of neutron flux, respectively. These tables show that the neutron spectra of PWR and FBR are not effective to annihilate the radioactivity even if the magnitude of the flux is 5×10^{15} [#/cm^2/sec] for 1000 days irradiation. This means that the reduction ratio is almost the same to the case of cooling only. On the other hand, the D-T is only effective to annihilate if the magnitude of the flux is more than 5×10^{15} [#/cm^2/sec].

In case of PWR and FBR, nuclear transmutation is mainly caused by capture reaction for the target nuclide, and the magnitude of capture cross section shown in Fig. 2 is 1 or 2 order smaller than that of (n, 2n) cross section. This smaller cross section brings the smaller nuclear transmutation than the case of D-T, where main nuclear transmutation is caused by (n, 2n) reaction.

In the following, we focus on the result of D-T, because PWR and FBR are found to be not effective for transmutation of the target nuclides.

Table 4 shows the change in radioactivity ratio to the original (before irradiation) of each nuclide at 1000 days irradiation or cooling period (cooling only case). “Cooling only” case shows the value of more than 90 % except for ^{134}Cs, because only ^{134}Cs has short half-life of about 2 years. These nuclides expect for ^{134}Cs can be annihilated by the irradiation of D-T neutron flux, and the effectiveness becomes remarkable when the magnitude of the flux is more than 5×10^{15} [#/cm^2/sec]. Although there is a remarkable increase in radioactivity ratio of ^{134}Cs, which is transmuted from ^{135}Cs by (n, 2n) reaction, the radioactivity ratio decreases rapidly by relatively shorter half-life, as shown in Table 5. Table 5 shows the radioactivity ratio at 4500 days (3500 days cooling after 1000 days irradiation).

The radioactivity of not only Cs/Sr but also Tc/I are decreasing to almost the same value about 40 % by irradiation of D-T neutron flux. This is because of almost the same magnitude of (n, 2n) cross section for those nuclides.

Table 2 Change in radioactivity ratio to the original (before irradiation) of ^{137}Cs and ^{137m}Ba.

days		before irradiation	1000days	4500days
5×10^{13}[#/cm^2/sec]	FBR	100%	94%	75%
	PWR	100%	94%	75%
	D-T	100%	100%	75%
5×10^{14}[#/cm^2/sec]	FBR	100%	95%	75%
	PWR	100%	96%	75%
	D-T	100%	149%	70%
5×10^{15}[#/cm^2/sec]	FBR	100%	101%	75%
	PWR	100%	111%	73%
	D-T	100%	361%	36%
cooling only		100%	94%	75%

Table 3 Change in radioactivity ratio to the original (before irradiation) of ^{90}Sr and ^{90}Y.

days		before irradiation	1000days	4500days
5×10^{13}[#/cm^2/sec]	FBR	100%	94%	75%
	PWR	100%	94%	75%
	D-T	100%	97%	74%
5×10^{14}[#/cm^2/sec]	FBR	100%	94%	74%
	PWR	100%	94%	75%
	D-T	100%	129%	70%
5×10^{15}[#/cm^2/sec]	FBR	100%	101%	74%
	PWR	100%	99%	74%
	D-T	100%	294%	41%
cooling only		100%	94%	75%

Table 4 Change in radioactivity ratio to the original (before irradiation) of each nuclide at 1000 days.

	irradiation flux [#/cm^2/sec]			1000days cooling
	5E+14	5E+15	5E+16	
Cs-134	73%	358%	1317%	36%
Cs-135	99%	92%	50%	100%
Cs-137	93%	86%	45%	93%
Ba-137m	93%	86%	45%	93%
Sr-90	93%	87%	48%	93%
Y-90	93%	87%	61%	93%
Tc-99	99%	92%	44%	100%
I-129	99%	91%	41%	100%

Table 5 Change in radioactivity ratio to the original (before irradiation) of each nuclide at 4500 days.

	irradiation flux [/cm^2/sec]			4500days cooling
	5E+14	5E+15	5E+16	
Cs-134	3%	13%	48%	1%
Cs-135	99%	92%	50%	100%
Cs-137	74%	69%	36%	74%
Ba-137m	74%	69%	36%	74%
Sr-90	73%	68%	38%	74%
Y-90	73%	69%	38%	74%
Tc-99	99%	92%	44%	100%
I-129	99%	91%	41%	100%

Table 6 Change in radioactivity ratio to the original (before irradiation) at 1000 days.

	before irradiation	irradiation flux [#/cm^2/sec]			1000days cooling
		5E+14	5E+15	5E+16	
Cs-134	5%	3%	17%	62%	2%
Cs-135	0%	0%	0%	0%	0%
Cs-137	28%	26%	24%	13%	26%
Ba-137m	26%	24%	23%	12%	25%
Sr-90	20%	19%	18%	10%	19%
Y-90	20%	19%	18%	12%	19%
others	0%	12%	108%	517%	0%
Total	100%	104%	207%	626%	91%

Table 7 Change in radioactivity ratio to the original (before irradiation) at 4500 days.

	before irradiation	irradiation flux [#/cm^2/sec]			4500days cooling
		5E+14	5E+15	5E+16	
Cs-134	5%	0%	1%	2%	0%
Cs-135	0%	0%	0%	0%	0%
Cs-137	28%	21%	19%	10%	21%
Ba-137m	26%	20%	18%	9%	20%
Sr-90	20%	15%	14%	8%	15%
Y-90	20%	15%	14%	8%	15%
others	0%	0%	0%	3%	0%
Total	100%	70%	66%	40%	71%

In Tables 6 and 7, the radioactivity ratios of ^{99}Tc and ^{129}I are in others, because their half-life is long, and the ratio for the whole radioactivity is very small. Table 6 shows the change in radioactivity ratio of the target at 1000 days, normalized to that the original (before irradiation) radioactivity is 100 % in total. As already mentioned, if the irradiation is performed by 5×10^{14} [#/cm^2/sec], nuclear transmutation is trivial and there is no acceleration to reduce the radioactivity. The magnitude of D-T neutron flux more than 5×10^{15} [#/cm^2/sec] is necessary to reduce the radioactivity of the target nuclides. On the other hand, some nuclides such as ^{134}Cs and others shown in Table 6 increase during the irradiation period. However those nuclides decay rapidly during the following cooling period (3000 days) by relatively short half-life (see Fig. 1) and radioactivity can be reduced to about 40 % by 5×10^{16} [#/cm^2/sec] for 1000 days irradiation and the following 3500 days cooling, as shown in Table 7. The reduction rate of radioactivity for the case of 5×10^{16} [#/cm^2/sec] case is about 60 % and that of "cooling only" case is about 30 %. This means the irradiation by D-T spectrum has the possibility to accelerate the annihilation of radioactivity of the target nuclides.

It should be mentioned that the magnitude of 5×10^{16} [#/cm^2/sec] is hard to realize, but these results shows that larger flux level is preferable to accelerate the annihilation of the radioactivity of the target nuclides (Cs, Sr, Tc, I).

In this study, the irradiation period is fixed to 1000 days (about 3 years) by considering the realistic period to irradiate as the target. However if the irradiation period is enhanced to 10000 days, the change in radioactivity ratio of 5×10^{15} [#/cm^2/sec] is about the half value of 5×10^{16} [#/cm^2/sec] shown in Tables 4 to 7, for ^{137}Cs/^{137m}Ba and ^{90}Sr/^{90}Y and about the same value for ^{99}Tc and ^{129}I. This means that the total amount of irradiated neutron (fluence) is also the main parameter to consider the reduction of radioactivity of the target by nuclear transmutation. However longer irradiation period would be preferable only for the transmutation of some nuclide with remarkably long half-life such as ^{99}Tc and ^{129}I in this study.

The spectrum of the fusion reactor greatly changes by the kind of blanket and really includes a lot of low energy neutrons [4]. Transmutation by (n, 2n) reaction will decrease in case of actual spectrum of fusion reactor, because the neutron spectrum of fusion reactor is much softer than 14 MeV mono energetic spectrum. It is necessary to pay attention to this point when clearing the effectiveness of actual fusion reactor.

5. Conclusions and Future Works

The application of 14 MeV neutrons for nuclear waste transmutation was investigated to be clear the effectiveness to reduce the radioactivity of fission products. In comparison to the spectra of thermal reactor and fast reactor, it was found that D-T neutron of 14 MeV is preferable and has the possibility to reduce the radioactivity of fission products such as Cs, Sr, Tc, and I. Because the reaction to transmute the nuclide is different between D-T and PWR/FBR, and the magnitude of the cross section is smaller for PWR/FBR cases than that for D-T case.

Although the 14 MeV neutron is useful for the nuclear transmutation by (n, 2n) reaction, it was found that the irradiation by the magnitude less than 5×10^{15} [#/cm^2/sec] for 1000 days is meaningless to accelerate the reduction of the radioactivity. However if the irradiation period is extended to 10000 days, the remarkable reduction can also be obtained by the neutron flux of 5×10^{15} [#/cm^2/sec].

The magnitude of annihilation and the magnitude of fission products stored inside the D-T reactor are to be evaluated to be clear the effectiveness of D-T reactor for the annihilation of the radioactivity with consideration for the spectrum of fusion reactor which is not 14 MeV. The investigation by considering this point is planned as future work.

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