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Radioactive Waste Disposal by Nuclear Transmutation
核変換による放射性廃棄物処理について

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Radioactive waste can be reduced by transmuting radioactive nuclides into stable nuclides through nuclear reactions. As target nuclides, ^{90}Sr, ^{99}Tc, ^{129}I, and ^{137}Cs are selected and the difference of transmuting efficiency among several neutron spectrum fields is investigated in this study. Numerical results show that neutron spectrum of thermal reactor is the best to reduce the radioactivity of ^{99}Tc and ^{129}I, and D-T neutron spectrum is the best to reduce the radioactivity of ^{90}Sr and ^{137}Cs by larger transmutation of the target nuclide through (n,2n) reaction than that of the neutron spectra of fission reactors.

1. Introduction

Radioactive waste can be reduced through the use of nuclear reactions such as capture, fission, and (n,2n) etc. by changing element and/or atomic mass. This transmutation efficiency of the target nuclide depends on its cross section which has a remarkable dependency on the irradiated neutron energy and also different value among nuclides. Therefore the recommended neutron spectrum to annihilate the radioactive waste is supposed to be different for different target nuclides. In this manuscript, the cause of the efficiency difference by changing neutron spectrum fields and the target nuclides is shown with numerical results.

The target nuclides in this survey are ^{90}Sr, ^{99}Tc, ^{129}I and ^{137}Cs: these are major radioactive waste (fission product:FP) from fission reactors and also to be annihilated, and the neutron spectrum fields used in this survey are from two fission reactors and one fusion reactor: Pressurized Water Reactor (PWR), Fast Breeder Reactor (FBR), and D-T neutron (DT).

2. Neutronic Parameter

Neutronic parameters to affect the transmutation efficiency are cross section (XS) of each nuclide and neutron spectrum. In addition to these, burnup chain which shows the transmuting relation among nuclides is also to be considered to discuss the results. In this section, those data are briefly described for the sake of easy understanding.

2.1 *Cross Section*

The major reaction types to be considered in the transmutation are capture and (n,2n) for FP. Fig. 1 shows XS in barn unit as a function of incident neutron energy for the nuclides: ^{90}Sr, ^{99}Tc, ^{129}I and ^{137}Cs. It is noted the XS of (n,2n) reaction type is almost the same value and shape for all nuclides.

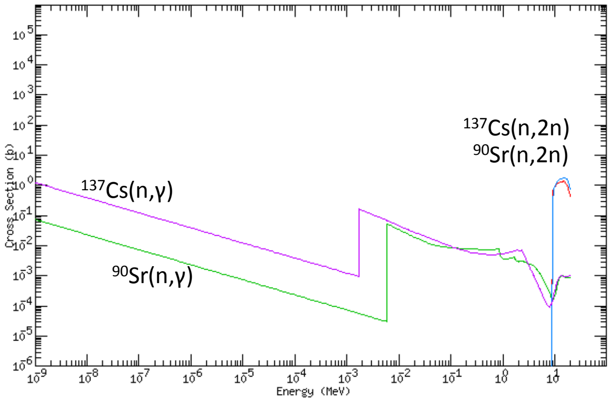

Fig.1 (a) Cross sections of ^{90}Sr and ^{137}Cs

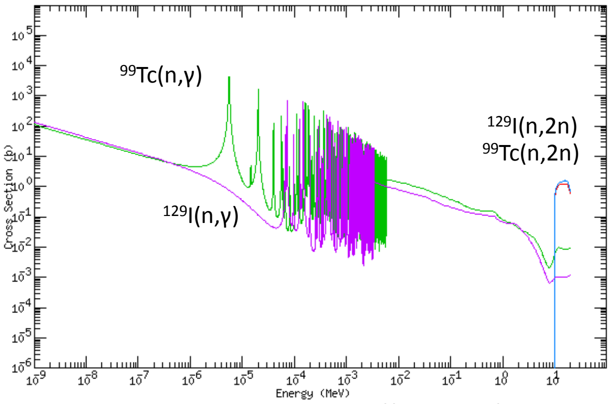

Fig.1 (b) Cross sections of ^{99}Tc and ^{129}I

2.2 *Neutron Spectrum*

Neutron spectrum is also important to consider the transmutation because of the remarkable energy dependence of XS. The neutron spectra used in this survey are shown in Fig. 2. The amount of the transmutation is evaluated as the proportional value to the product of XS and neutron flux.

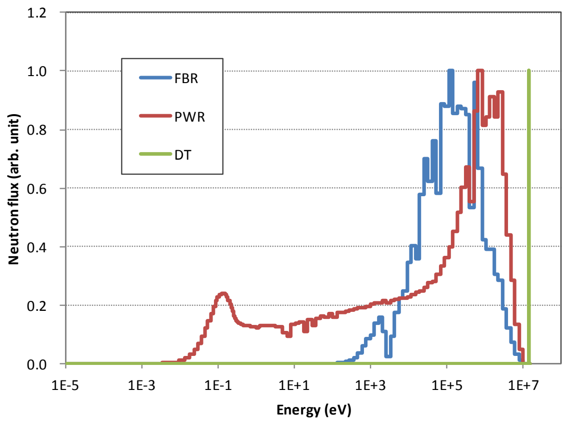

Fig.2 Neutron spectrum in PWR, FBR and DT

2.3 *Burnup Chain*

Radioactive nuclide transmutes as the results of nuclear reaction and also radioactive decay. The relation among the nuclides is called burnup chain. Fig. 3 shows a piece of burnup chain related to the ^{90}Sr. Exact burnup chain includes the relation (n,p), (n,3n), alpha-decay etc. and all the relation is considered in the following results.

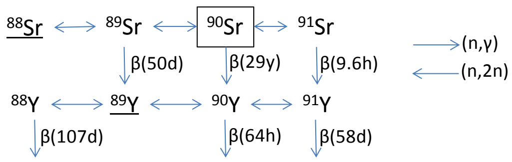

Fig. 3 Burnup chain related to ^{90}Sr

3. Calculation and Results

In this study, calculations were performed by MVP-II[1] and ORIGEN2[2]. MVP-II is used to evaluate effective XS to be used in ORIGEN2 burnup (irradiation and cooling) calculation. The amount of neutron flux is set to 5×10^{15}[#/cm^{2}sec] during irradiation period of 1000 days, and cooling period after irradiation is set to 3500 days. Non-irradiation (only cooling) case is also performed to be clear the transmutation efficiency by the irradiation.

Results are summarized in Table I with half-lives of each nuclide, where the mass of target nuclide and total radioactivity of all transmuted nuclides are normalized by initial value.

4. Discussions

Table I shows that mass of the target nuclides decrease for all cases and the transmutation efficiency remarkably changes by changing the spectrum fields. However, the spectrum field is different to get the best efficiency: DT is the best for ^{90}Sr and ^{137}Cs, and PWR is the best for ^{129}I and ^{99}Tc. Although total radioactivity increase at the end of irradiation (1000day) because of the production of short half-life nuclides, the radioactivity becomes smaller than the initial value at the end of cooling (4500day) except for the case of ^{129}I in PWR. The increment in PWR is caused by the accumulation of ^{134}Cs. However, the half-life of ^{134}Cs is 2.1y and this is remarkably shorter than the target nuclide. In addition, by considering the smaller mass of ^{129}I after irradiation, the radioactivity decreases rapidly compared to the non-irradiated case. It should be mentioned that to irradiate ^{137}Cs and ^{90}Sr in FBR and PWR is meaningless, and DT usually shows good performance to reduce the radioactivity. These differences mainly come from the facts that DT is likely to cause (n,2n) reaction compared to FBR and PWR, and the XS of (n,2n) reaction at 14(MeV) is higher than that of capture reaction especially for ^{137}Cs and ^{90}Sr, and almost all the transmuted nuclides are stable or have shorter half-life compared to the target nuclides.

Table 1 Summary of the transmutation efficiency

		Mass Ratio				Total Radioactivity Ratio			
		0	1000	2000	4500	0	1000	2000	4500
^{137}Cs 30y	FBR	1.00	0.93	0.88	0.75	1.00	1.04	0.87	0.74
	PWR	1.00	0.93	0.87	0.74	1.00	1.04	0.87	0.74
	DT	1.00	0.45	0.42	**0.36**	1.00	3.60	0.42	**0.36**
	cooling_only	1.00	0.94	0.88	0.75	1.00	0.94	0.88	0.75
^{90}Sr 29y	FBR	1.00	0.93	0.87	0.74	1.00	1.01	0.87	0.74
	PWR	1.00	0.93	0.88	0.74	1.00	0.99	0.88	0.74
	DT	1.00	0.52	0.49	**0.41**	1.00	2.94	0.49	**0.41**
	cooling_only	1.00	0.94	0.88	0.75	1.00	0.94	0.88	0.75
^{129}I 1.6e+7y	FBR	1.00	0.84	0.84	0.84	1.00	1.28E+6	0.94	0.85
	PWR	1.00	0.10	0.10	**0.10**	1.00	2.81E+6	1.67E+3	1.70E+2
	DT	1.00	0.52	0.52	0.52	1.00	4.57E+6	0.53	**0.52**
	cooling_only	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
^{99}Tc 2.1e+5y	FBR	1.00	0.75	0.75	0.75	1.00	2.43E+4	0.75	0.75
	PWR	1.00	0.03	0.03	**0.03**	1.00	1.40E+4	0.03	**0.03**
	DT	1.00	0.54	0.54	0.54	1.00	4.83E+2	0.56	0.56
	cooling_only	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

5. Conclusions

To annihilate the radioactive waste by neutron irradiation, the difference of neutron spectrum causes remarkable difference in transmutation efficiency even in the same neutron fluence. DT is found to be the best for ^{137}Cs and ^{90}Sr, and PWR is the best for ^{99}Tc and ^{129}I by considering the shorter half-life of ^{134}Cs than that of ^{129}I.

References

[1] Y. Nagaya, K. Okumura, T. Mori et al, JAERI 1348, (2005).

[2] A. G. Croff, ORNL/TM-7175, July 1980.