A STUDY FOR OPTIMAL TRANSMUTATION SYSTEM

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Abstract

Couple of transmutation systems are being under investigation to design the optimal transmutation device. Several basic studies were performed for that objectives: (1) select the radioactive nuclides to be transmuted; (2) investigate the physical characteristics of each nuclide; (3) study the most favorable neutron energy environment for the transmutation. The existing LWR and LMFBR cores were found to be not a satisfiable ones in terms of transmutation rate itself.

Introduction

Since the late of 1980s, the development of nuclear transmutation technology has been one of the main topics in nuclear research. Much more attention is expected to be paid in near future because of the increased concerning on environment and the difficulty to have a geologic repository.

As an alternatives or supportive options for geologic isolation of the nuclear waste. Kaeri embarked in the research for nuclear transmutation. Various transmutation technologies for high level radioactive waste(HLW) are currently being proposed at many organizations in the world such as thermal/fast reactor based and accelerator driven subcritical systems[1]. No detail assessments among the technologies have been performed to date. It might be very difficult to have international consensus on those systems. Each nation(or institute) has developed her own technology on the basis of the nuclear fuel cycle and the technical strong points she has.

The technologies currently introduced are supposed to be everything the transmutation device can be in terms of technical and economical feasibility. Kaeri decided to develop her own model throughout two stage plans. In the first stage. Kaeri will set up basic model the most preferable to Korea by evaluating the proposed technologies. In the second stage. the conceptual design will be performed for the optimum transmutation model. On the way for the goal of the first stage, a couple of basic studies were done. The results of basic studies and the temporary direction Kaeri has were discussed in this paper.

Methodology for Optimal Basic Model Determination

Basic model is expected to be determined based on the procedure shown in Figure 1.

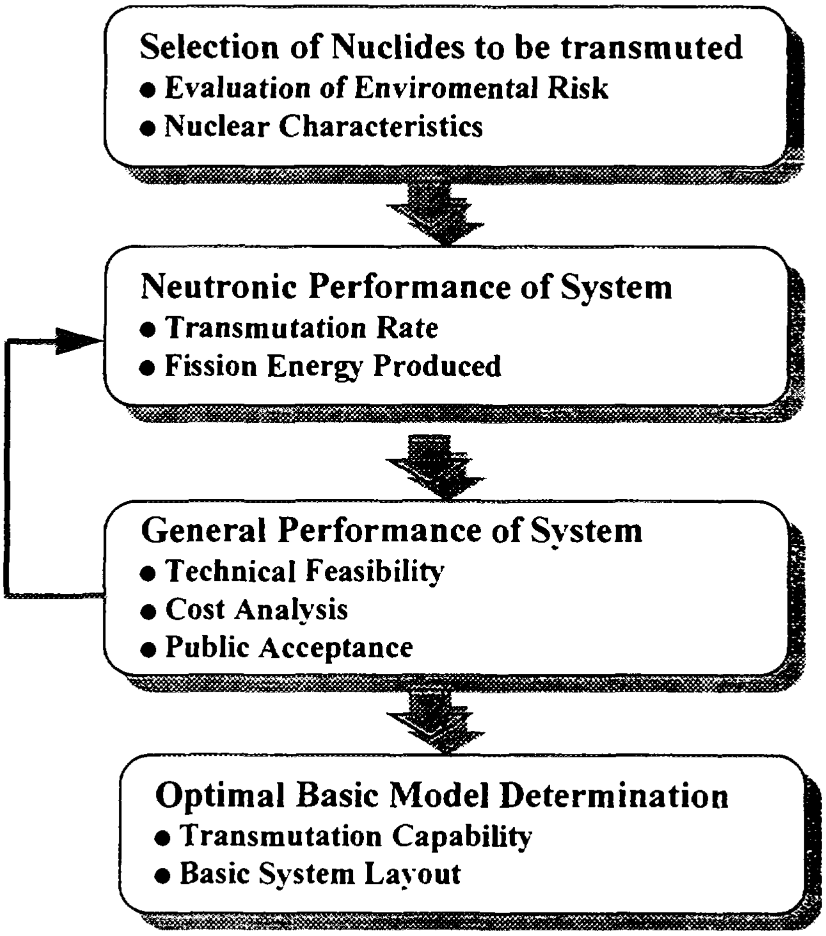

Figure 1 Optimal System Design Procedure

As it is known, one ton of LWR spent fuel includes 10 kg of actinides and 30 kg of fission products[2]. Plutonium occupying 92% of actinide is considered as an energy resource rather than waste to be eliminated. Two major factors are considered for the selection of the nuclides to be transmuted: (1) nuclear characteristics; (2) environmental risk due to the nuclide. In the evaluation of the nuclear characteristics of each radioactive nuclide, its half life and toxicity in terms of water dilution volume are included. On the other hand, solubility and retardation factor are major factors to be evaluated in the environmental risk point of view.

The objectives of transmutation is not just to convert the nuclide from one to another but is to reduce the toxicity to the acceptable level within the time period human can handle. The transmutation rate of each nuclide varies from system to system because a neutron cross section does from nuclide to nuclide. In order to estimate the overall transmutation efficiency of the proposed system, following performance index of neutron energy spectrum was set up,

$$PI = W_t \sum_i \omega_i \varepsilon_i + W_f \sum_j \omega_j f_j \quad \text{----------------------------------}(1)$$

where.

$W_{t,f}$: weighting factor for toxicity reduction and the amount of fission energy

ω_i : weighting factor accounting for the toxicity and the amount of nuclide i

ε_i : the ratio of the toxicity without transmutation to the toxicity with transmutation for nuclide i

f_j : amount of energy generated by fission of nuclide j

In equation (1), first term account for the toxicity reduction and second term is for the energy production by fission. By including the fission term in PI, the system which has a neutronic performance enhancing the system economy more can be preferred. The optimal system is the one maximizing PI. Table 1 shows the transmutation systems currently proposed and being investigated.

Table 1 Typical Transmutation Systems

Neutron Energy Spectrum	Transmutation System
Thermal	• LWR Based • CANDU Based • Accelerator Based(ADDT)
Fast	• LMFBR Based • Accelerator Based

General performance of a system is evaluated in terms of cost, technical feasibility, public acceptance, etc. The optimal basic model is finalized by considering the general performance as well as the neutronic performance.

Basic Studies and Results

Selection of Nuclides

As it was mentioned before, plutonium was not considered as a nuclide to be transmuted. The assessment on the nuclear characteristics and environmental risk of nuclides produces Table 2. The nuclides in Table 2 are the major radioactive materials to be eliminated.

Table 2. Long-lived Radioactive Nuclides to be Transmuted

Nuclide	Half Life(year)	Amount(gram)[#]	Water Volume(m^3)[*]
Np-237	2.1E6	440	2.4E8
Am-241 Am-243	432 7.4E3	500 90	8.6E11 5.0E10
Tc-99	2.1E5	760	8.5E7
I-129	1.6E7	180	2.9E9

: Amount per one ton of spent fuel

* : Water Volume required to dilute the toxicity of nuclide to the level of MPC(m^3/MG of Nuclide).

Neutronic Performance of the System

The transmutation capabilities of LWR and LMFBR based systems were investigated by using the neutron energy spectra of a typical LWR and LMFBR. The calculations were done by using ORIGEN2[3] and WIMS[4].

The meaningful transmutation device should be able to reduce the amount of toxicity considerably within a manageable time period. In this study, 500 year was assumed as a period man can predict. In order to see the changes in the toxicity reduction rate due to the employment of transmutation system, the toxicity was calculated for two cases: 1) no irradiation and natural decay for 500 years; 2) irradiation for 3 years and natural decay for remaining 497 years. Also the amount of the original nuclides remained after 500 years was evaluated to see the relationship between the toxicity reduction and the reduction of the original nuclide. Four different flux levels were employed to check the sensitivity of the transmutation efficiency to the flux level. Table 3 summarizes the results of the evaluation. In the table, the numerical value represents the ratio of the value obtained from the irradiated case to the one from no irradiated. Thus lower value means higher efficiency.

For the toxicity due to the fission products(I-129, Tc-99), LWR based system has higher performance than LMFBR based system. In addition, the destruction rate of original nuclide is directly reflected in the toxicity change in fission product transmutation. Very similar trend can be found in Am-241/243 transmutation. However, the case of Np-237 is totally different. Any reduction in the toxicity can be found in both of the systems although considerable amount of Np-237 itself can be destroyed. It means that most of Np-237s are destroyed by neutron capture under existing LWR and LMFBR energy spectra and those capture reactions generate lots of higher toxic actinides. It should be noted that the flux levels are ~ 10^{14} n/cm^2-sec and ~10^{15} n/cm^2-sec in existing LWR and LMFBR core, respectively. In those flux range the transmutation rate is very small except the case of Am-241/243 and the small transmutation rate loads too much burden onto the separation system. Thus present LWR and LMFBR cores are not suitable for the purpose of the transmutation.

Table 3. Comparison of transmutation capability between LWR and LMFBR based system

		Np-237		Am-241/243		I-129		Tc-99	
		MR	TR	MR	TR	MR	TR	MR	TR
LWR System	1.0E13**	0.97	9.15	0.90	0.89	0.995	0.995	0.991	0.991
	1.0E14	**0.73**	**65.5**	**0.42**	**0.39**	**0.95**	**0.95**	**0.917**	**0.917**
	1.0E15	0.05	61.7	0.05	0.05	0.61	0.61	0.421	0.421
	1.0E16	0.001	1.1	0.001	0.001	0.007	0.007	0.001	0.001
LMFBR System	1.0E13	0.998	1.4	0.99	0.976	0.999	0.999	0.999	0.999
	1.0E14	0.98	4.7	0.97	0.965	0.996	0.996	0.996	0.996
	1.0E15	**0.84**	**32**	**0.88**	**0.96**	**0.964**	**0.964**	**0.958**	**0.958**
	1.0E16	0.18	72	0.25	0.23	0.69	0.69	0.654	0.654

MR : Mass Ratio [M(Irrad)/M(No Irrad)]
TR : Toxicity Ratio[T(Irrad)/T(No Irrad)]
** : neutrons/cm^2-sec

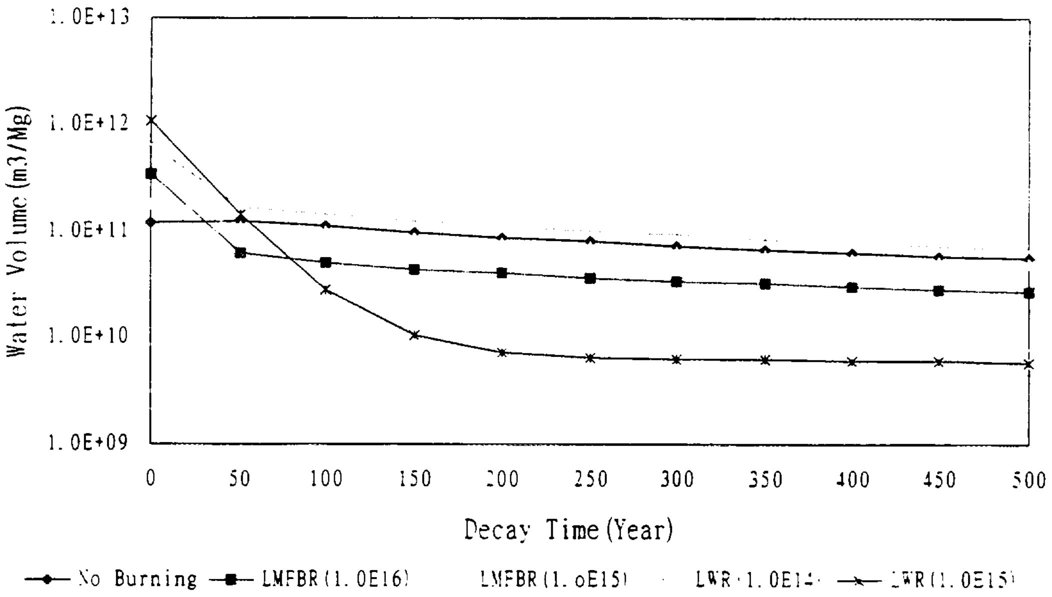

Figure 2. Plutonium Toxicity Variation after 3 year burning at Various Flux Conditions

Although plutonium is not considered as the nuclide transmuted in Kaeri, some calculations were done to examine how much toxicity can be reduced when plutonium is burnt up in existing LWR or LMFBR reactors. It was found that a considerable amount of toxicity reduction can not be expected under present flux level of LWR or LMFBR. This fact means that plutonium burning in LWR or LMFBR is not a burning of toxic actinide but is just a generation of energy. Figure 2 shows the results of the calculations.

Summary

Kaeri is currently investigating the transmutation capability of each proposed system and doing some study on its general performance such as a technical feasibility, economical gain/loss, and public acceptability.

It is possible to transmute a radioactive nuclide by using various types of particle such as proton, electron, photon. Using neutron is found to be the most effective way. It is noted that each neutron has its own production cost although the cost varies from system to system. From the economical point of view, the effectiveness of a neutron should be maximized.

It is found that the capture cross section of Np-237 is 10,000 and 100 times higher than the fission cross section in LWR and LMFBR energy range, respectively[5]. Thus more than 99% of Np-237s are destroyed by neutron capture which might produce more toxic higher actinide. Table 3 reflects this kind of phenomena. The neutron cross section study tells that Np-237 and Am-241 have a reversing point around neutron energy of 1 MeV where fission cross section starts being higher than capture cross section. Fissioning destroys the nuclide in the most efficient way in terms of toxicity reduction rate and system economy. Therefore neutron higher than 1.0 MeV is recommendable for the transmutation of the toxic actinide. On the other hand, low energy neutrons are much more effective for the transmutation of toxic fission products.

From the preliminary study, following tentative directions were obtained,

- Existing LWR and LMFBR core conditions are not appropriate for the transmutation, even for the fission products.
- The preferred neutron energy for actinide transmutation is totally in oppositessite from that of fission product transmutation.
- Although very high thermal flux level(> 10^{16} n/cm^2-sec) can transmute both of actinides and fission products altogether, such a system is not practical because of the irradiation problem of structural material and its extremely low neutron economy(high system cost).

- It is almost impossible to transmute the toxic nuclides to be selected using one single neutron energy spectrum. It might be suggested to develop a hybrid system where two extreme neutron energy spectra can coexist.

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