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Potential of Thorium-based Fuel Cycle for 900MWe PWR Core to Constrain Pu and to Reduce Long-term Toxicity

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Abstracts

An IAEA CRP on the use of thorium as nuclear fuel titled "Potential of Th-based Fuel Cycles to Constrain Pu and Reduce the Long-term Waste Toxicities" has been performed. The CRP has been divided into three stages: benchmark calculations, optimization of the incineration of plutonium in various reactor types, and assessment of the resulting impact on the waste toxicity. Two benchmark calculations, pin cell and a single assembly, for thorium fuel were performed with HELIOS. We has investigated the potential of thorium-based fuel in the form of ThO_2-PuO_2 in PWR type reactor currently operated in Korea as a part of CRP activity. The mass balance of plutonium isotope for thorium fuelled-core was calculated and compared with that for MOX fuelled-core. The toxicity calculation for thorium fuel cycle was also performed in terms of the radioactivity, the ingestion hazard, and the inhalation hazard.

Thorium fuelled core can consume plutonium 2.2 or 2.4 times larger than MOX core and the fissile plutonium fraction change in thorium fuel is also twice or three times larger than in MOX fuel. The toxicity of thorium fuel is rather higher than that of conventional UO_2-fuelled PWR due to higher content of Cm-244 for the near-term (~10^2 years) after discharge. For the mid-term (10^2~10^5 years) after discharge, the toxicity of thorium system is lower up to a factor of two than that of conventional UO_2-fuelled PWR due to the effect of plutonium incineration. For the long-term (10^5~10^6 years) after discharge, the toxicity of combined system with $(Th+Pu)O_2$ unit becomes higher than that of conventional UO_2-fuelled PWR due to the daughter isotopes of U-233.

1. Introduction

In the framework of IAEA activities on the use of thorium as nuclear fuel, a CRP (Coordinated Research Program) titled "Potential of Th-based Fuel Cycles to Constrain Pu and

Reduce the Long-term Waste Toxicities" has been performed[1]. This CRP examined the different fuel cycle options in which plutonium can be recycled with thorium to incinerate plutonium. Each participant has chosen his own cycle, and different cycles are compared through certain predefined parameters. The toxicity accumulation and the transmutation potential of thorium-based cycles for current, advanced and innovative nuclear power reactors are investigated. The CRP has been divided into three stages: benchmark calculations, optimization of the incineration of plutonium in various reactor types, and assessment of the resulting impact on the waste toxicity.

We has investigated the potential of thorium-based fuel in the form of ThO_2-PuO_2 in PWR type reactor as an activity of IAEA CRP. A 900MWe PWR currently operated in Korea was adopted as a reference plant. The conceptual core was assumed to be fully loaded with thorium fuel. The conceptual core with PuO_2-UO_2 (MOX) was also investigated for the comparison with thorium core. Even though the fully-loaded ThO_2-PuO_2 or MOX core concept needs to change the control rod and soluble boron systems to satisfy the current design limit and technical specification, any system design change to meet current design limit was not considered in this study.

Both the reactor- and weapon-grade plutonium composition were applied to the conceptual cores. The changes in quantity and composition of plutonium isotopes due to fuel burnup were analyzed. The neutronic characteristics of conceptual cores such as power distribution, soluble boron concentration, reactivity parameters, control rod worth etc. were also calculated.

In order to evaluate the long-lived waste toxicity of thorium-based fuel cycle, a combined system model with conventional UO_2- and with $(Th+Pu)O_2$-fuelled reactor was applied and the toxicity calculation was performed in terms of the radioactivity, the ingestion hazard, and the inhalation hazard.

2. Benchmark Calculation

In order to get a comparison of the effect of different methods and data bases applied in the countries participating in the CRP, two benchmark calculations had to be performed before the start of the actual fuel cycles studies : cell burnup and assembly burnup calculations. The contents of plutonium dioxide in the fuel pellet for both the benchmark problems are about 5 weight percent. We used HELIOS-1.4 with a designed library for these benchmark calculations[3]. The comparison of the results achieved by the participants are listed in Figures 1 and 2 and Table I. The k-infinite and reactivity parameters calculated with HELIOS show good agreement with other participants

3. Construction of Conceptual PWR Core

The typical design data for Korean 900MWe PWR were adopted for the conceptual plutonium cores. The reactor core is consisted of 157 fuel assemblies which have 17×17 fuel array. The rated thermal power is 2775MWth and the system pressure is 150 bars. As for fuel material data, the typical plutonium composition of PWR spent fuel having burnup of 33GWd/MtU is used for reactor-grade plutonium. Isotopic composition of plutonium in reactor-grade ThO_2-PuO_2 and PuO_2-UO_2 (MOX) fuel is 1.8, 59.0, 23.0, 12.2, and 4.0w/o for ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu, respectively. The composition of weapon-grade plutonium isotopes is 0.0, 94.0, 6.0, 0.0, and 0.0w/o for ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu, respectively.

The plutonium contents of thorium and MOX fuel were determined so that conceptual cores have similar cycle length as uranium core currently being operated with longer than annual fuel cycle scheme. In this study, three types of fuel composition, the thorium and reactor-grade plutonium, the thorium and weapon-grade plutonium, and MOX fuel with reactor-grade plutonium, were studied. The total plutonium contents of 7.5, 5.0 and 5.62 w/o were decided for the thorium fuel with reactor-grade plutonium, the thorium fuel with weapon-grade plutonium, and MOX fuel with reactor-grade plutonium, respectively.

The reference fuel cycle length of conceptual cores was longer than annual. Sixty-four fuel assemblies were discharged from and newly loaded into the reload core for each cycle. Some fresh fuel assemblies bear four or eight gadolinia rods as burnable poison rod to control excess core reactivity and core power distribution. The fuel cycle characteristics of thorium and MOX cores are summarized in Table II. The low-leakage loading strategy in which most of fresh fuel assemblies take inboard locations was applied. Figures 3 and 4 show the loading patterns of equilibrium cores.

4. Nuclear Characteristics Conceptual Cores

The critical soluble boron concentrations for the equilibrium cores loaded with thorium cores and MOX core were shown in Figure 5. In case of thorium core with weapon grade plutonium, the consumption of ^{239}Pu in is much larger than the conversion of fertile isotopes to fissile during core burnup, and the boron concentration was rapidly decreased as compared with the other conceptual core fuelled with reactor grade plutonium.

Key core physics parameters such as soluble boron concentration, temperature coefficients, boron worth, and control rod worth were calculated with MASTER code and are listed in Table III. The neutron spectrum of conceptual cores fuelled with plutonium are harder than that of uranium fuelled core. Since harder neutron spectrum enhances the neutron leakage from the core, the temperature coefficients of the conceptual cores are more negative than that of UO_2 core. Since boron is strong absorber for thermal neutron, boron worth is also strongly affected by neutron spectrum. The boron worth of conceptual cores are about half of nominal value of uranium fuelled core because of harder neutron spectrum. Control rod, which is also strong thermal neutron absorber, in the conceptual core has less worth than in UO_2 core.

The change in plutonium mass for thorium and MOX fuel batches is summarized in Table IV. Since each conceptual core has different fuel cycle length, the mass value in Table IV was adjusted to be equivalent to 1GW-300EFPD (Effective Full Power Day) in order to compare the mass change under the same condition.

For the thorium fuel with reactor grade plutonium, the annual charged and discharged mass of plutonium are 1708 kg and 875 kg, which means 833 kg of plutonium is incinerated annually by 300EFPD operation of one 1,000 MWe PWR. The incineration rate of plutonium for thorium core with weapon grade plutonium and MOX core are 757 kg and 351 kg per 1GWe-300EFPD.

5. Assessment of the Effect of Pu-Incineration on the Long-lived Waste Toxicity

In order to evaluate the long-lived waste toxicity of thorium-based fuel cycle, a combined system model with conventional UO_2- and with $(Th+Pu)O_2$-fuelled reactor was applied. Since the plutonium produced from the conventional UO_2-fuelled PWR can be recycled into $(Th+Pu)O_2$ core or MOX core, the combined system is consisted of conventional UO_2 core as Pu-supplier and of $(Th+Pu)O_2$ core (or MOX core) as Pu-burner. For the comparison purpose, a conventional UO_2 reactor as a reference system and an UO_2+MOX combined system were also considered. So, the toxicity of the long-lived waste from the following three scenarios were calculated and compared ;

Scenario 1 : Conventional UO_2 Only System

A typical PWR fuelled with UO_2 is adopted as conventional UO_2 system. The waste from this system is assumed to be disposed without separation of any isotopes.

Scenario 2 : Conventional UO_2 + $(Th+Pu)O_2$ (As Pu-Burner) Combined System

A combined system, which has the same size with a conventional UO_2 system, with certain fractions of UO_2 unit and of $(Th+Pu)O_2$ unit is considered. The plutonium of the spent fuel from UO_2 unit is separated and recycled into Pu-burner, $(Th+Pu)O_2$ unit, as illustrated in Figure 6. The waste of this system is the heavy metal with plutonium-separation from UO_2 unit and the spent fuel for $(Th+Pu)O_2$ unit.

Scenario 3 : Conventional UO_2 + MOX (As Pu-Burner) Combined System.

This system is the same one as Scenario 2 except that MOX unit is adopted as Pu-burner instead of $(Th+Pu)O_2$ unit in Scenario 2.

The plutonium discharge rate of one conventional UO_2-fuelled PWR is assumed to be

245kg of plutonium per one GWa (300EFPD). In the Section 4, the loading rates of plutonium are 1708kg Pu/Gwa for one $(Th+Pu)O_2$ Pu-Burner and 1346kg Pu/Gwa for one MOX Pu-Burner. Therefore, the number of the conventional UO_2 reactors required to supply the plutonium to one Pu-burner are 7.0 for a Thoria Pu-Burner and 5.5 for a MOX Pu-Burner.

The fractions of UO_2 unit and of Pu-burner unit in a combined system has to be decided to balance the plutonium between discharged from UO_2 unit and loaded into Pu-burner unit, and to have the same size with a conventional UO_2 system. So, a combined system with conventional UO_2 and with $(Th+Pu)O_2$ (Scenario 2) is composed of 0.875 UO_2 units and 0.125 $(Th+Pu)O_2$ units, and a combined system with conventional UO_2 and with MOX (System 3) is composed of 0.8462 UO_2 units and 0.1538 MOX units.

The results of toxicity calculation for each scenarios are given in Figures 7, 8, and 9 for the radioactivity, the ingestion hazard, and the inhalation hazard, respectively. The toxicity calculation for each scenario resulted in the radioactivity, the ingestion hazard, and the inhalation hazard. Figures 10, 11 and 12 illustrate the ingestion hazard fraction of major isotope to total hazard. The dominant isotopes to the toxicity are varied with time period after discharge due to different decay time as shown in table V. The isotope mass in spent fuel from the different combined systems are given in Table VI. For the near-term ($\sim10^2$ years) after discharge, Pu-238, Pu-241, Am-241, and Cm-244 dominate the toxicity. The toxicity of combined system in this period is rather higher than that of conventional UO_2-fuelled PWR due to higher content of Cm-244. For the mid-term ($10^2\sim10^5$ years) after discharge, Pu-239, Pu-240, and Am-241 dominate the toxicity. The toxicity of combined system is lower up to a factor of 2 than that of conventional UO_2-fuelled PWR due to the effect of plutonium incineration. For the long-term ($10^5\sim10^6$ years) after discharge, Pu-239 and Th-229 are the major sources of the toxicity. The toxicity of combined system with $(Th+Pu)O_2$ unit becomes higher than that of conventional UO_2-fuelled PWR due to the daughter isotopes of U-233.

6. Results and Discussions

In order to investigate the potential of thorium-based fuel for 900MWe PWR to reduce the plutonium, the mass balance of plutonium isotope for thorium fuel was compared with that for MOX fuel. For the thorium fuel with reactor grade plutonium, the annual charged and discharged mass of plutonium are 1708 kg and 875 kg, which means 833 kg of plutonium is incinerated annually by 300EFPD operation of one 1,000 MWe PWR. The incineration rate of plutonium for thorium core with weapon grade plutonium and MOX core are 757 kg and 351 kg per 1GWe-300EFPD. Therefore, thorium fuelled core can consume plutonium 2.2 or 2.4 times larger than MOX core. The fissile plutonium fraction change in thorium fuel is also twice or three times larger than in MOX fuel.

Based on these results, it is concluded that thorium fuelled PWR core has higher potential to reduce plutonium than MOX PWR core.

The toxicity of thorium fuel is rather higher than that of conventional UO_2-fuelled PWR due to higher content of Cm-244 for the near-term (~10^2 years) after discharge. For the mid-term (10^2~10^5 years) after discharge, the toxicity of thorium system is lower up to a factor of 2 than that of conventional UO_2-fuelled PWR due to the effect of plutonium incineration. For the long-term (10^5~10^6 years) after discharge, the toxicity of combined system with $(Th+Pu)O_2$ unit becomes higher than that of conventional UO_2-fuelled PWR due to the daughter isotopes of U-233.

Acknowledgement
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1. Proposal on " Potential of Th-based Cycles to Constrain Pu and to Reduce of Long-term Toxicities, IAEA, 1995.
2. V.Arkhipov et. al., " Progress Summary of the IAEA Coordinated Research Programe on the Potential of Th-based Fuel Cycles to Constrain Pu and to Reduce the Long-lived Waste Toxicity, Proceedings of ICENES Meeting, Tel Aviv, 1998.
3. HELIOS Version 1.4, TN19/41.16.15, Scandpower, July 3, 1996.

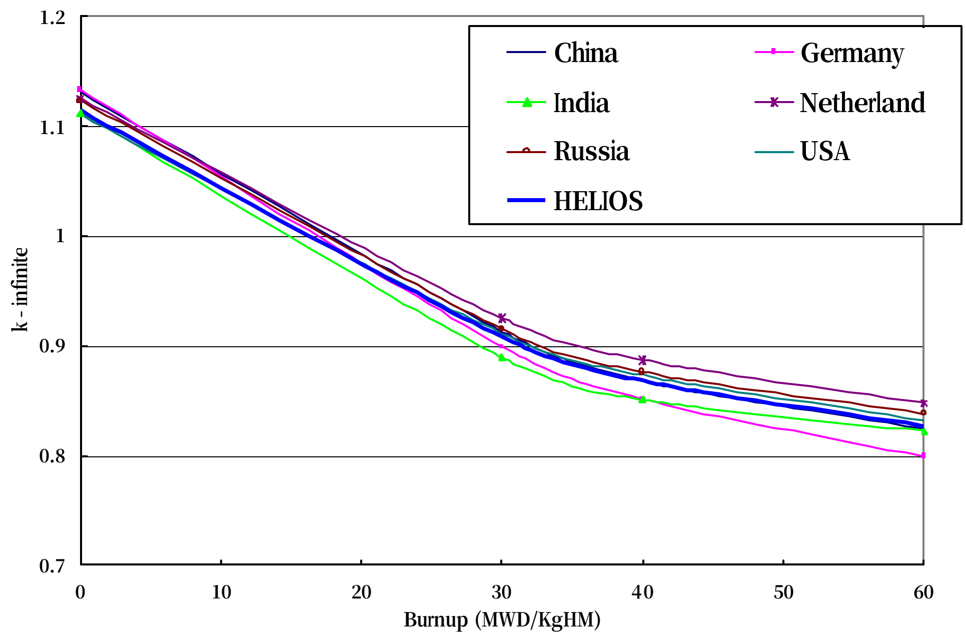

Figure 1. The Comparison of k-infinite as a Function of Burnup, Pin-Cell Calculation

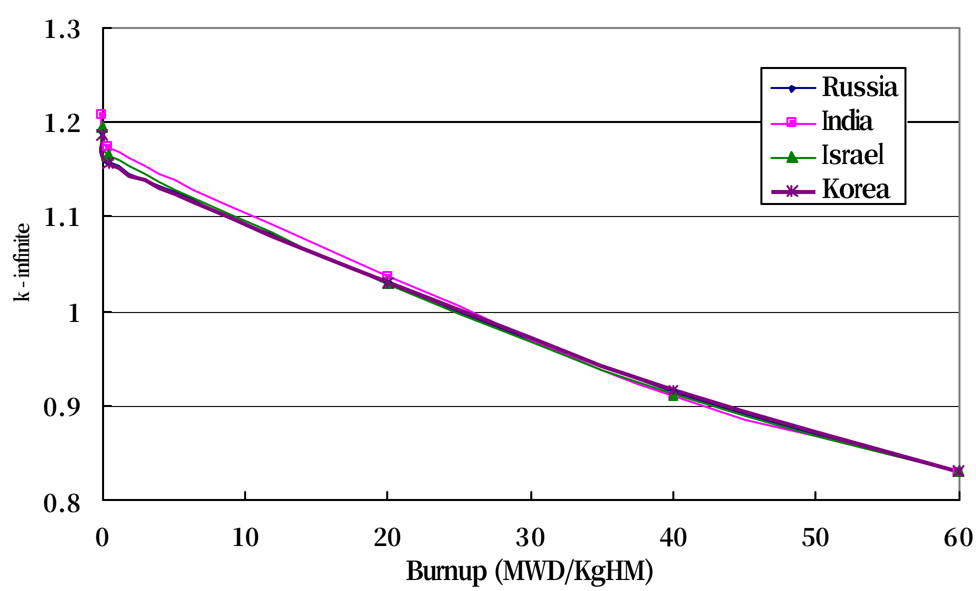

Figure 2. The Comparison of k-infinite as a Function of Burnup, Assembly Calculation

Table I. Temperature Coefficients and Boron Worth for Assembly Benchmark Problem

	0 GWD/T			60 GWD/T		
	MTC (pcm/°C)	FTC (pcm/°C)	BW (pcm/ppm)	MTC (pcm/°C)	FTC (pcm/°C)	BW (pcm/ppm)
Russia	-35.00	-2.80	-3.80	-15.0	-3.60	-11.00
Japan	-26.96	-2.83	-3.41	-9.69	-3.78	-8.64
Korea	-32.00	-3.11	-4.08	-12.89	-3.97	-11.25
Israel	-33.33	-2.92	-4.00	-11.42	-4.77	-11.19

Table II. Fuel Cycle Characteristics for Thorium and MOX Cores

Fuel Cycle / **Core Characteristics**	**Thorium Core**		**MOX Core**
	with RG Pu	**with WG Pu**	
Number of Fuel Assemblies in a Core			
Thorium or MOX Fuel Assembly	157	157	157
Number of Fresh Fuel Assemblies			
without gadolinia	32	36	32
with 4 gadolinia	12	-	12
with 8 gadolinia	20	28	20
Fuel Assembly Specification			
Total Plutonium Content in Fuel (w/o)	7.50	5.00	5.62
Fissile Plutonium Content in Fuel (w/o)	5.34	4.70	4.00
Equilibrium Cycle Length (EFPD)	401	361	393
Fuel Burnup (MWD/MtM)			
Batch Burnup	40.48	36.40	38.37
Assembly Maximum Burnup	52.68	45.80	49.58

2	1	F 8Gd	2	2	F 4Gd	1	F
1	1	1	2	F 8Gd	2	F	1
F 8Gd	1	1	F 8Gd	1	F 4Gd	F	
2	2	F 8Gd	2	1	F	1	
2	F 8Gd	1	1	F	1		
F 4Gd	2	F 4Gd	F	1			
1	F	F	1				
F	1						

2 Twice Burnt Fuel

1 Once Burnt Fuel

F Fresh Fuel

Figure 3. Loading Pattern for Equilibrium Core with Thorium or MOX Fuel with Reactor-Grade Plutonium.

2	1	F	2	2	F 8Gd	1	F
1	1	1	2	F 8Gd	2	F	1
F	1	1	F 8Gd	1	F 8Gd	F	
2	2	F 8Gd	2	1	F	1	
2	F 8Gd	1	1	F	1		
F 8Gd	2	F 8Gd	F	1			
1	F	F	1				
F	1						

2 Twice Burnt Fuel

1 Once Burnt Fuel

F Fresh Fuel

Figure 4. Loading Pattern for Equilibrium Core with Thorium Fuel with Weapon-Grade Plutonium.

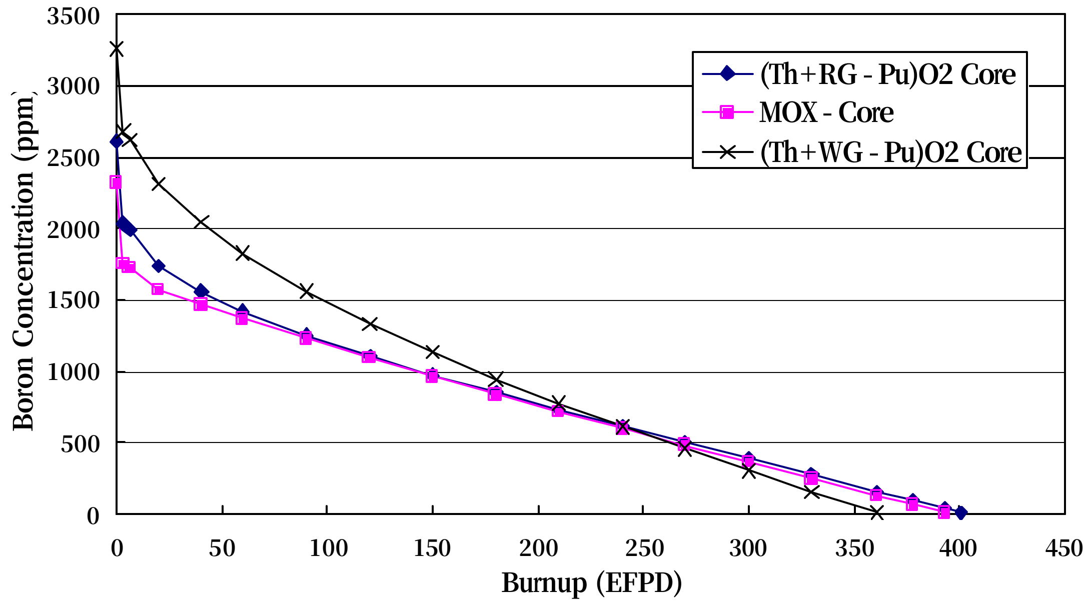

Figure 5. Critical Boron Concentration for Thorium and MOX Cores with Core Burnup.

Table III. Key Core Physics Parameter for Thorium and MOX core

Fuel Cycle / Core Characteristics	Thorium Core		MOX Core
	with RG Pu	**with WG Pu**	
Boron Concentration (ppm)			
To control at HZP, ARO, (k=1.0)	3259	3704	2853
To control at HZP, ARI, (k=1.0)	1405	1948	1141
To control at HFP, ARO, (k=1.0)			
0 EFPD, No Xenon	2609	3258	2318
6 EFPD, Eq. Xenon	1992	2617	1724
Moderator Temp. Coefficient at HFP (pcm/°C) at BOC/ EOC	-36.2/-67.2	-20.5/-62.5	-44.2/-79.8
Isothermal Temp. Coefficient at HZP (pcm/°C) at BOC	-13.6	-2.5	-19.6
Fuel Temp. Coefficient at HFP (pcm/°C) at BOC/ EOC	-3.74/-3.87	-3.50/-3.78	-3.04/3.20
Boron Worth at HFP (pcm/°C) at BOC/ EOC	-3.05/-4.18	-3.63/-5.50	-3.50/-4.52
Total Control Rod Worth at HFP (pcm) at BOC/EOC	6618/7491	7290/7576	7048/7855

Table IV. Plutonium Mass Balance

	Mass (Kg)		
	Thorium Core		MOX Core
	with RG Pu	with WG Pu	
Plutonium Charged	1708	1264	1346
Plutonium Discharged	875	507	995
Plutonium Burned	833	757	351
Fissile Fraction for Plutonium Charged	72 %	94 %	72 %
Fissile Fraction for Plutonium Discharged	51 %	60 %	61 %

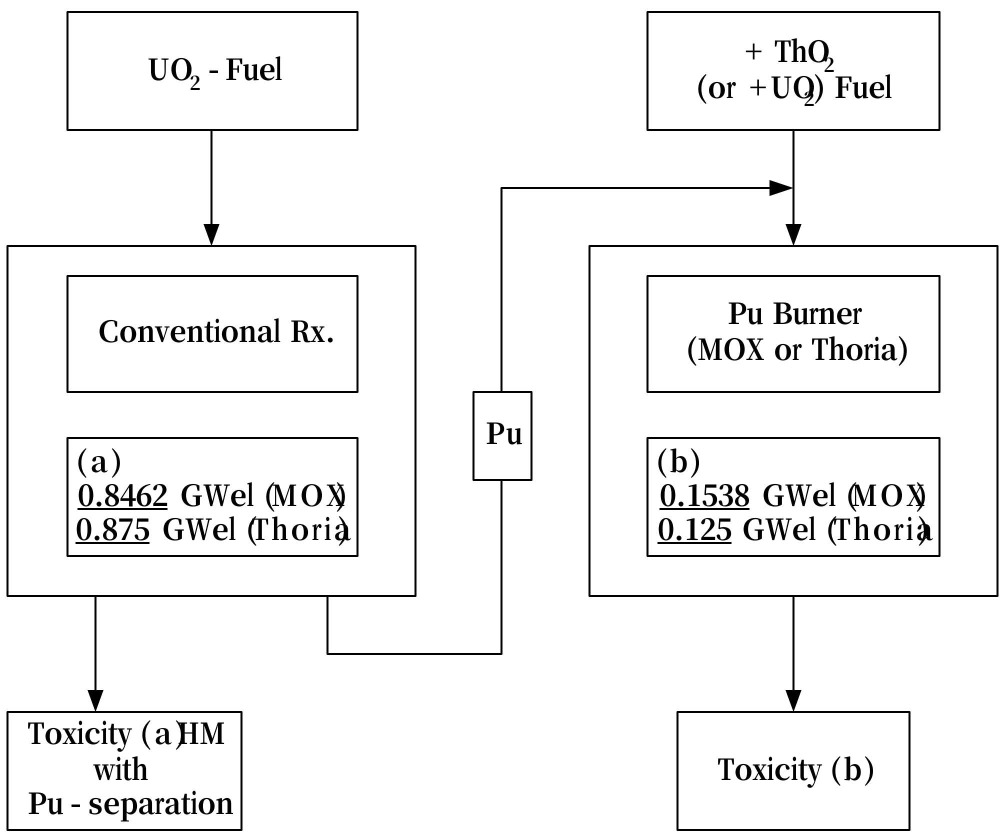

Figure 6. Diagram of Combined System

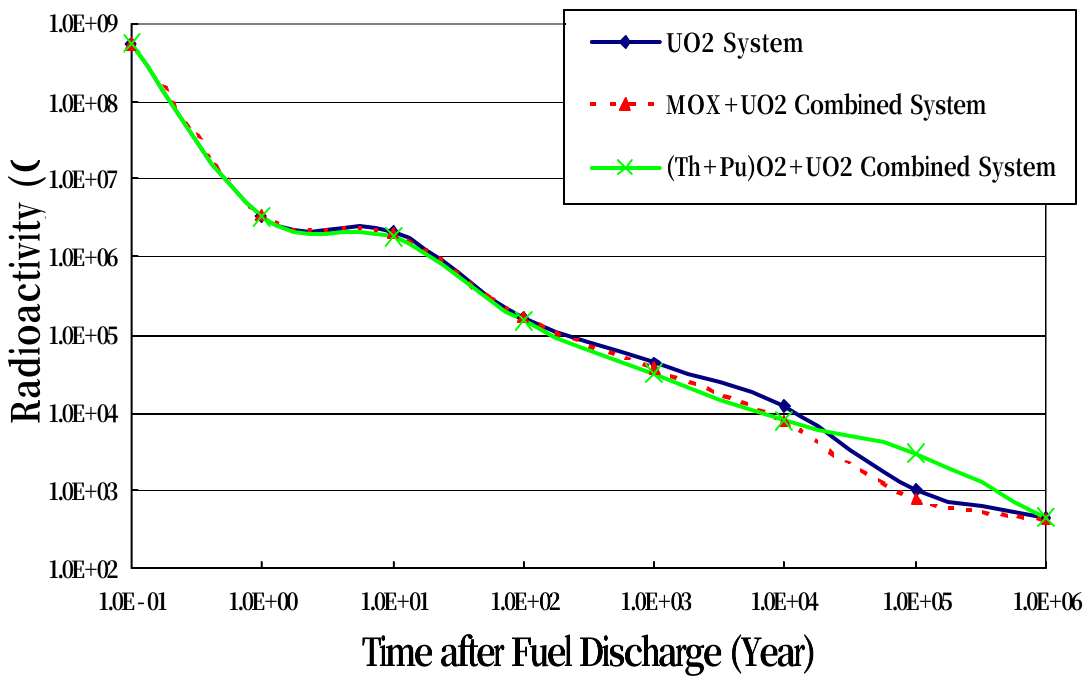

Figure 7. Radioactivity of Each Scenario, (Ci)

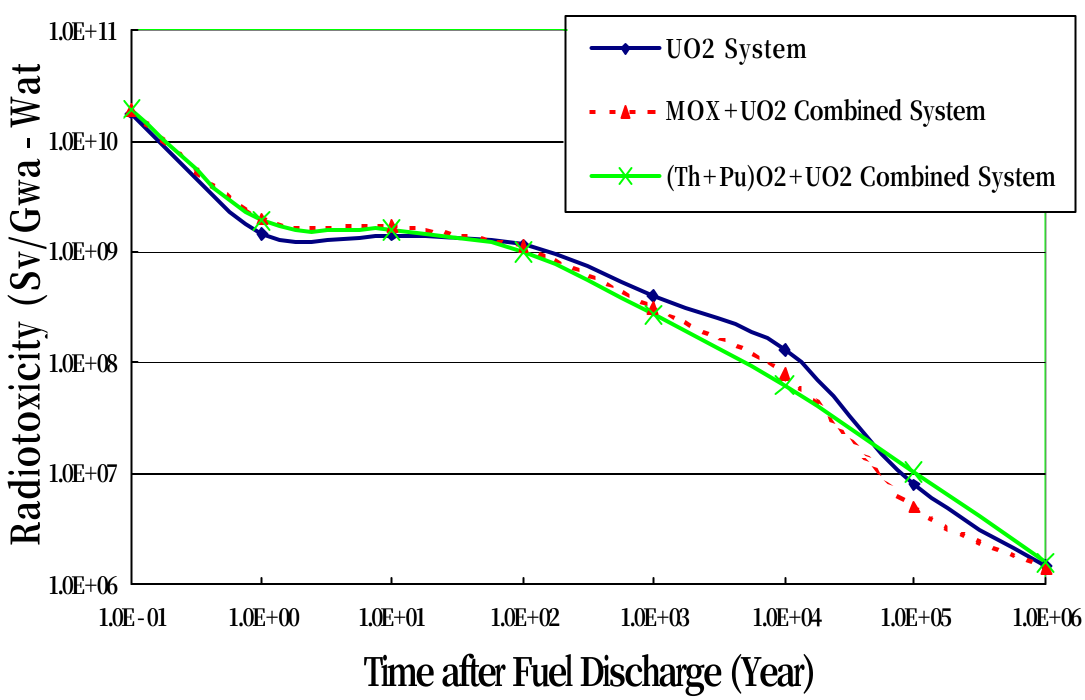

Figure 8. Ingestion Hazard of Each Scenario, (Sv/Gwa-Water)

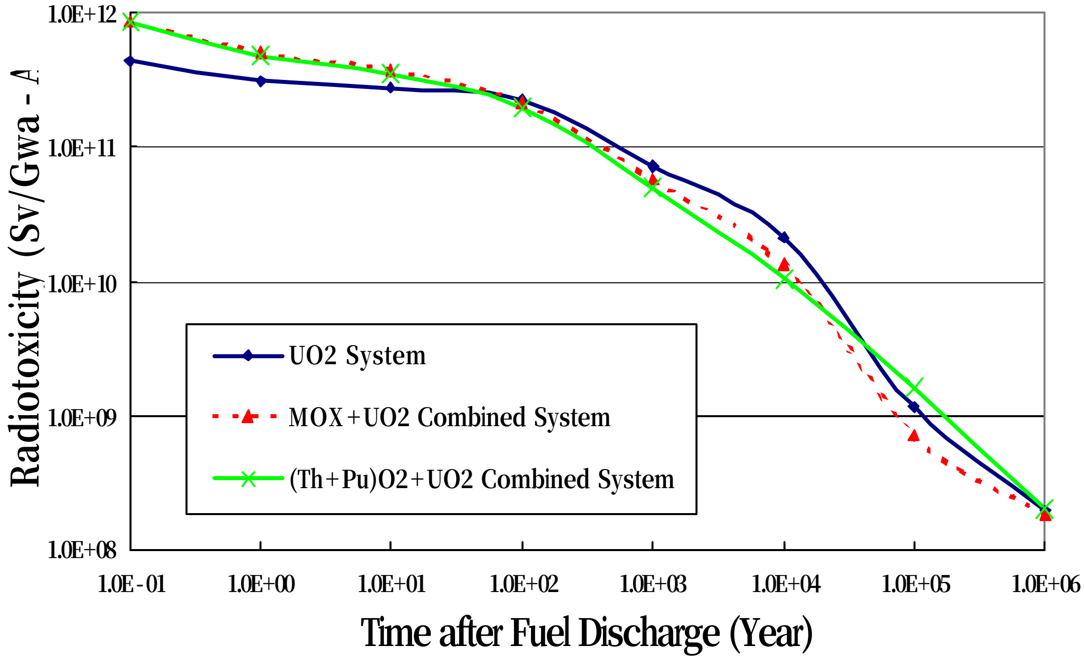

Figure 9. Inhalation Hazard of Each Scenario, (Sv/Gwa-Air)

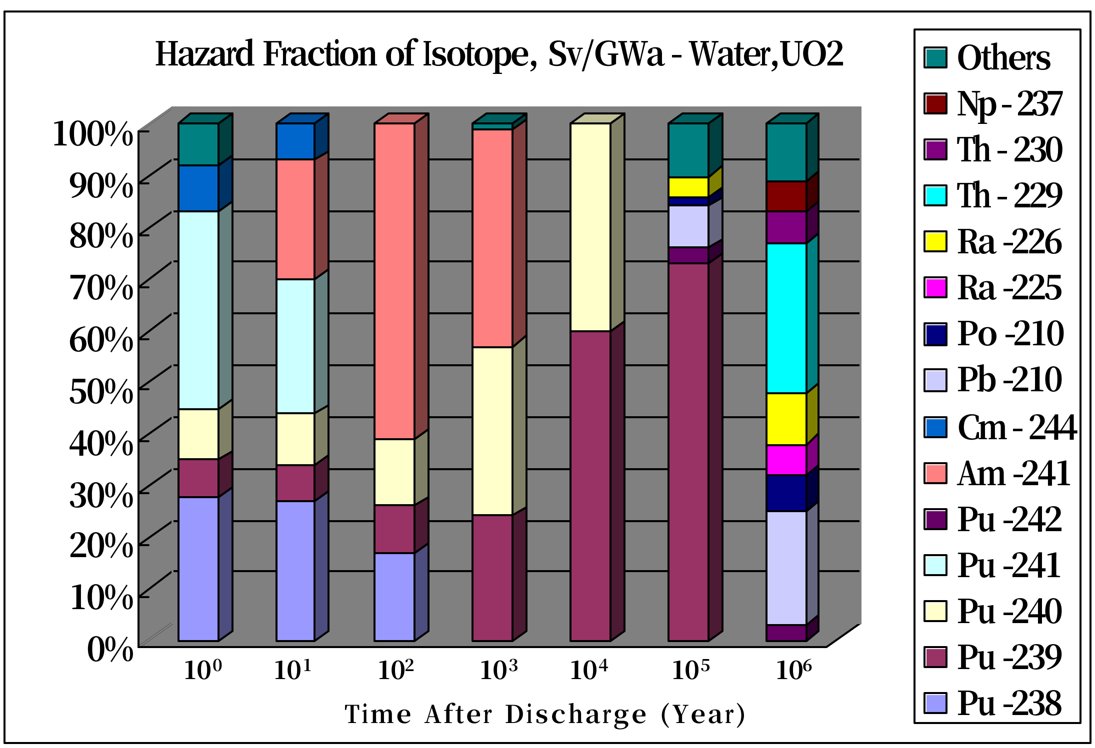

Figure 10. Ingetion Hazard Fraction of Major Isotope in Total Hazard, UO_2 Fuel

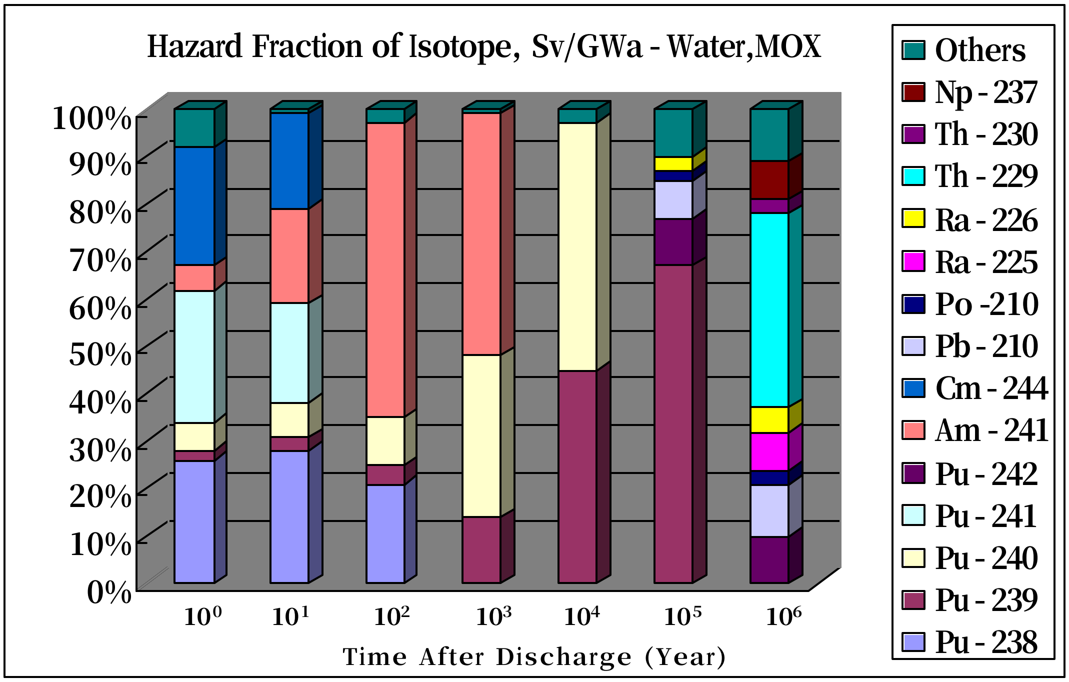

Figure 11. Ingetion Hazard Fraction of Major Isotope in Total Hazard, MOX Fuel

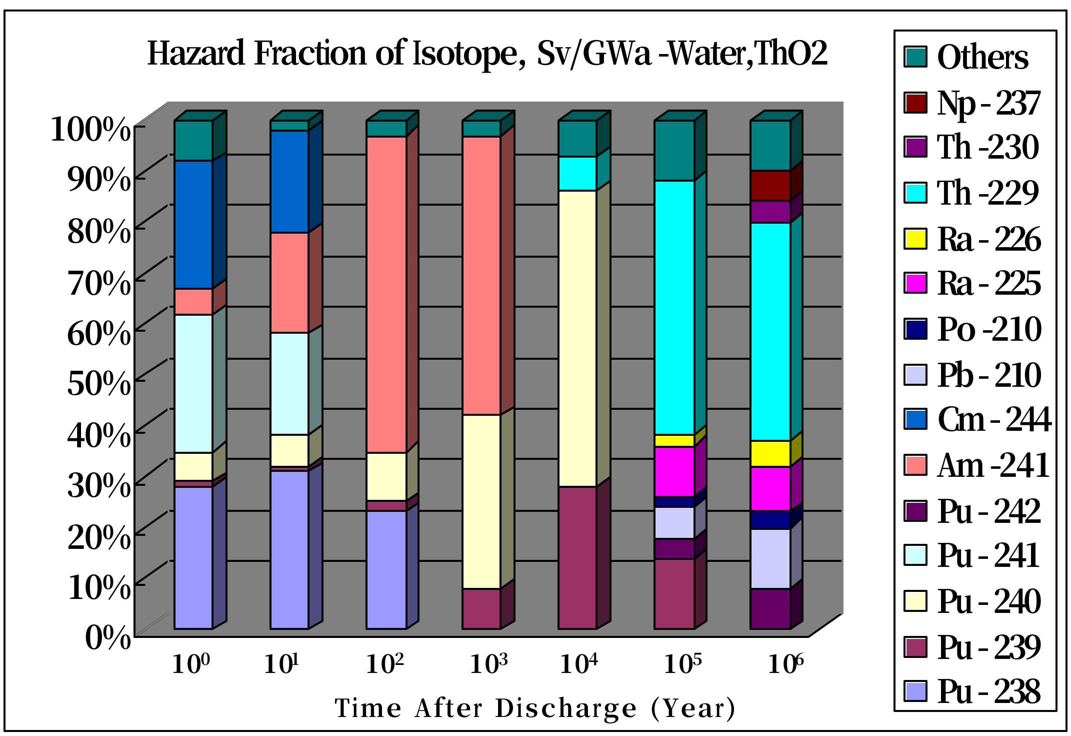

Figure 12. Ingetion Hazard Fraction of Major Isotope in Total Hazard, ThO_2 Fuel

Table V. Dominant Isotopes to Toxicity

Year after Discharge	10^0	10^1	10^2	10^3	10^4	10^5	10^6
UO_2	Pu-238 Pu-241	Pu-238 Pu-241 Am-241	Am-241 Pu-238	Am-241 Pu-240	Pu-239 Pu-240	Pu-239	Th-229 Pb-210
$(Th+Pu)O_2$	Pu-238 Pu-241 Cm-244	Pu-238 Pu-241 Am-241 Cm-244	Am-241 Pu-238	Am-241 Pu-240	Pu-239 Pu-240	Pu-239 Th-229	Th-229 Pb-210
MOX	Pu-238 Pu-241 Cm-244	Pu-238 Pu-241 Am-241 Cm-244	Am-241 Pu-238	Am-241 Pu-240	Pu-239 Pu-240	Pu-239	Th-229 Pb-210

Table VI. Isotope Mass in Spent Fuel from the Combined System (Kg/Gwa)

	Pu-238	Pu-239	Pu-240	Pu-241	Am-241	Cm-244
UO_2	3.12	143	57.8	31.1	0.9	0.5
$(Th+Pu)O_2$	3.79	33.9	42.3	28.5	3.1	2.2
MOX	3.89	74.1	51.5	32.9	3.2	2.4