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Doctoral Dissertation

Study on seed and blanket ADS for transuranic elements transmutation using thorium-reprocessed fuel and its application in Vietnam

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July, 2014

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Abstract

From the viewpoint of reducing the greenhouse effect and providing sufficient supply against increasing power demand, nuclear power is inevitably a choice for not only the developed countries but also the developing countries. Most operating nuclear power plants use several-percent-enriched uranium fuel while the uranium resource limitation will be an obstacle to the expand of nuclear power due to the development of the world economy. Moreover, the accumulating of spent nuclear fuel from the nuclear power plants holds the potential risk for the environment. Accelerator driven system (ADS) with its inherent safety feature and waste transmutation potential is recognized as a promising system to annihilate the radioactivity of nuclear waste. Besides, with its capability of converting fertile fuel to fissile fuel, ADS is also able to utilize thorium resource which is abundant in the nature in order to produce energy. In this thesis, a conceptual design of seed and blanket ADS for efficient TRU transmutation and power generation using thorium-reprocessed fuel and its application in the case of nuclear power installation in Vietnam is presented.

Chapter 1 provides an overview of energy production problems and the background of nuclear waste transmutation and thorium utilization potential. This chapter also points out the purpose of this thesis.

Chapter 2 describes the design parameter and core configuration of the seed and blanket ADS for efficient TRU transmutation using thorium-reprocessed fuel. In this chapter, the innovative point of the design compared to other designs is explained. In order to achieve the efficient transmutation, the TRU transmutation performance and the safety features of the system with different design parameters are examined. The ADS core with high transmutation rate and safe operation is chosen.

Chapter 3 provides the multi-cycle design concept of the seed and blanket ADS in order to achieve the high utilization of thorium fuel assemblies for power production. Instead of removing all the fuel assemblies out of the core after burning for one cycle (430 days), the reprocessed fuel assemblies shuffled for a second burnt while thorium fuel assemblies are maintained inside the reactor core until the end of core life.

TRU transmutation rate and thorium utilization of the multi-cycle core is compared with the single one to show the improvement in thorium utilization and TRU transmutation of the multi-cycle concept.

Chapter 4 discusses the application of the seed and blanket ADS in the case of nuclear power installation in Vietnam. The nuclear power employment in Vietnam in the future is described. The amount of spent nuclear fuel of future reactors in Vietnam is calculated and the benefit of the installation of seed and blanket ADS in that situation is analyzed.

In Chapter 5, an additional study is performed in order to verify the thorium cross section data using the experimental and calculated data. The impact of thorium cross section uncertainty on seed and blanket ADS calculation is discussed whether the current thorium cross section data is reliable for the ADS core design and safety evaluation.

The obtained results are summarized and the main conclusions are drawn in the Chapter 6.

Chapter 1: **Introduction**

1.1 Motivation

1.1.1. Nuclear power and the energy security issue

As energy is the main "fuel" for social and economic development, with the rapid development of the world, the energy consumption will increase significantly. As reported in Table 1.1, the energy demand and the resource consumption will rise rapidly in about 20 years (from 1993 to 2020) [1].

Table 1.1. Key indicators for 1993, 2011 and 2020 [1]

Energy Types	**1993**	**2011**	**2020**	**% Growth 1993-2011**
Coal (Mt-mega ton)	4 474	7 520	10 108	68%
Oil (Mt-mega ton)	3 179	3 973	4 594	25%
Natural Gas (bcm-billion cubic meters)	2 176	3 518	4 049	62%
Nuclear (TWh)	2 106	2 386	3 761	13%
Hydro Power (TWh)	2 286	2 767	3826	21%
Biomass (Mtoe-mega ton of oil equivalent)	1 1036	1 277	1 323	23%
Other renewables (TWh)	44	515	1 999	n/a

As we can notice, because of the widely availability, simple operation and relatively low cost, fossil fuel generated power and nature gas and hydro power is playing important role in delivering energy. These energy types are the main power support for the developing country. For example, as shown in Fig. 1.1, in 2011, 28.3% of electricity supply in Vietnam was from hydro, 41.3% from gas and 19.8% from coal [2]. However, not only because of limitation of the natural resources but also the poor environment credentials of these energy types, the shifting of the energy share to the "cleaner" energy such as nuclear power or renewable

energy is necessary. When the renewable energies do not yet represent a full – scale alternative, nuclear power is inevitably a choice in the future. For example, in order to improve the energy security, nuclear power is approved and will be employed from 2023 as planned in Vietnam. Due to the plan, 2000 MWe nuclear power plant at Phuoc Dinh in the Ninh Thuan province should be on line by 2023. A further 2000 MWe was planned at Vinh Hai nearby, followed by a further 6000 MWe by 2030. A high demand scenario would give 8000 MWe in 2025 and 15,000 MWe (10% of total) in 2030 at up to eight sites in five provinces [2]. By year 2025 with 8000 MWe, about hundreds of spent fuels are produced every year [3]. The accumulating of radioactive waste may cause the problem to the environment of the nuclear power installing country and this is a major drawback for the production of energy through nuclear power. Therefore, a nuclear power producing technology which reduces the nuclear waste accumulating is seeking to develop and employ in the near future.

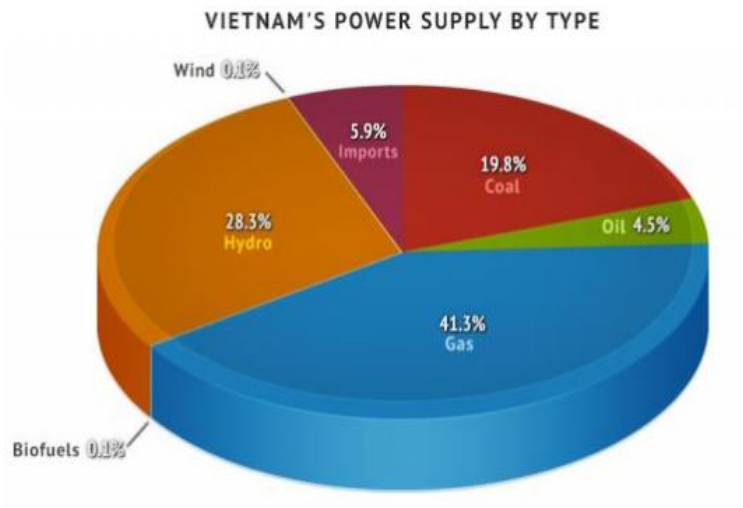


Fig. 1.1. Vietnam's power supply by type

1.1.2. Nuclear waste transmutation

As mentioned above, the problem of radioactive waste from nuclear power plants causes a big issue for the countries holding nuclear installation. Some isotopes with long half-lives will remain radioactive

hundreds of thousands of years in the future. Therefore, in order to minimize the exposing radiotoxic material to the environment, the material should either be placed safely in a geological repository or should be transmuted to convert some of the more harmful isotopes to less harmful products.

Geological repositories are designed for a safe storage of the radiotoxic material until it can decay to safe level. However, there are always risks caused by the earth crust beings and the intrusion of the water to the repository. The penetration of water into the storage area could cause the recriticalisation of waste which could have fatal consequences not only for containing unit but also for the whole repository. Not to mention the condition of the storage after centuries may change and lead to the escape of the radiotoxic waste [4]. Moreover, that area can't permanently be re-used for other purposes.

On the other hand, relatively large abundance of weapon-grade plutonium in conventional waste could be a source of future nuclear weapons proliferation.

Thus, in order to reduce the radiotoxicity, the criticality and proliferation risks and the size of the geological repository, the nuclear waste is considered to be transmuted in the dedicated facilities (Fig. 1.2). Fast spectrum systems are found to be advantageous for actinide transport from the standpoint that actinide fission-to-capture ratios are high in them. The fission-to-capture ratios of transuranic elements in fast and thermal system are given in Table 1.2 [5].

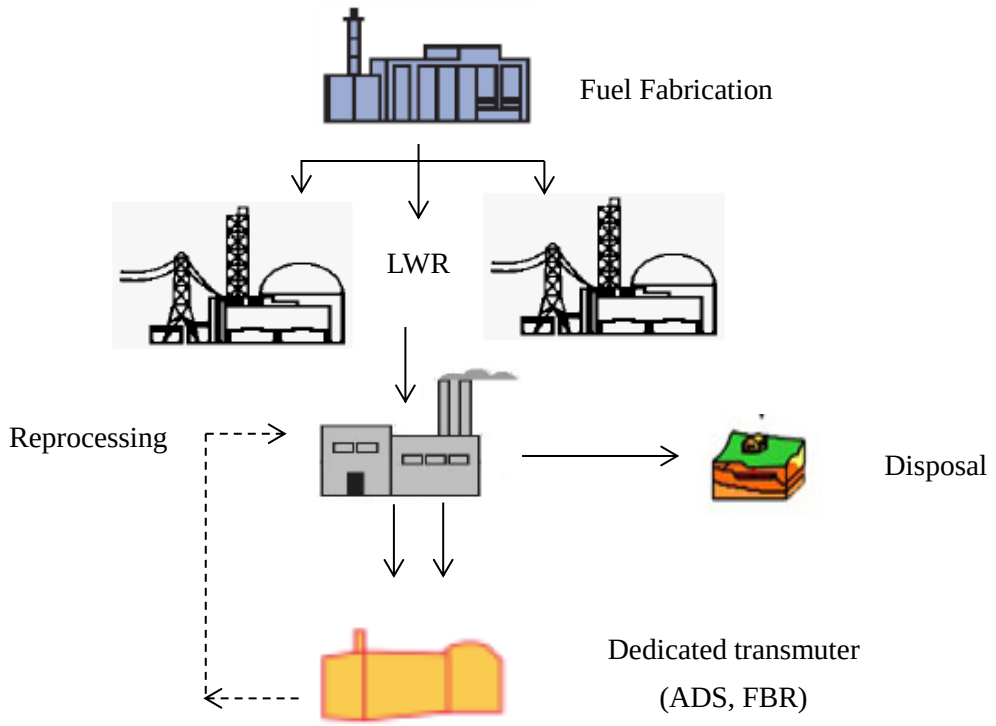


Fig. 1.2. Fuel cycle scenario into which the transmuter is introduced to transmute the nuclear waste

Table 1.2. The fission-to-capture ratios of non-uranium actinides in fast and thermal system [5]

Actinides	**Fission-to-capture ratio Thermal**	**Fission-to-capture ratio Fast**
^{237}Np	0.037	0.201
^{238}Pu	0.242	1.395
^{239}Pu	1.804	3.310
^{240}Pu	0.033	0.615
^{241}Pu	3.223	5.324
^{242}Pu	0.052	0.527
^{241}Am	0.029	0.180
^{242m}Am	5.265	9.677
^{243}Am	0.394	2.945
^{242}Cm	0.147	0.386
^{243}Cm	7.793	10.184
^{244}Cm	0.100	0.460

Currently, the technologies considered for nuclear waste transmutation are fast reactors and ADSs. Unfortunately, the fast reactors are facing to the safety problem due to the shorter lifetime of prompt neutron. Also, the fraction of delayed neutron emitted per fission for TRUs is several times smaller than that of ^{235}U and ^{238}U. TRU has little or no negative reactivity feedback due to Doppler broadening effect in the capture resonances [4]. On the other hand, ADS is considered as promising candidate to transmute nuclear waste with its flexibility with respect to fuel composition, and potentially enhanced safety. ADS systems are ideally suited to burning fuels which are problematic from the standpoint of critical reactor operation, namely, fuels that would degrade neutronic characteristics of the critical core to unacceptable level such as ^{233}U and minor actinide fuel. The enhanced safety of ADS is due to the fact that once the accelerator is turned off, the system shuts down [6]. Thus, with those advantages, ADS can be designed to have a fast neutron spectrum to be an efficient TRU transmutation facility. Moreover, with the capability of converting fertile fuel to fissile fuel, ADS also can be able to utilize thorium resource which is abundant in the nature in order to produce energy. One disadvantage of ADSs is that they depend on accelerator reliability to provide energy to a power grid because the beam must operate continuously to produce constant power [5]. Additionally, the reactivity swing due to burnup is rather large in ADSs. The larger accelerator current, which is expensive, is required to compensate the subciriticality. Hence, the reactivity swing is needed to keep small in order to reduce the expense of the ADS.

1.1.3. Thorium utilization potential

Nowadays, most operating nuclear power plants use several-percent-enriched uranium fuel. The estimated uranium resource today is considered to be exhausted in approximately 70 to 80 years if it were continued to be only utilized in a frame of current nuclear technology, namely in the light water reactors (LWRs) [7]. The increasing in demand for uranium to fuel the new reactors of the countries which increasingly rely on nuclear energy is expected. Thus, the pressure to develop new nuclear technologies as

well as to provide the alternative nuclear fuel becomes bigger and bigger. Besides, plutonium, generated from ^{238}U neutron capture, is the main source of proliferation potential and radiotoxicity. Limits on low cost uranium resources, waste, and proliferation raise concerns about long-term sustainability of nuclear energy. The use of thorium as a fertile material in nuclear fuel has been interested due to the superior neutronic properties of ^{233}U, and to the abundance of thorium in nature compared to uranium [8]. Thorium is three to four times more abundant than uranium in the earth's crust (average abundance of thorium in the earth's crust is 9.6 ppm compared to uranium at 2.7 ppm) [9]. As mentioned in the previous section about the capability of converting fertile fuel to fissile fuel of the ADS, thorium fuel can be consider to load into the ADS core to produce energy instead of uranium fuel.

Recent report [10] states that there is a world total of 5.4 million metric tons of thorium (Table 1.3). Compared with the case of uranium fuel when only 0.7% of natural uranium can be used, all of the mined thorium is potentially usable in a reactor. In other words, thorium has some 40 times the amount of energy per unit mass that could be made available, compared with uranium [11]. As it also estimated in the report, Vietnam has thorium mine with the reserves of about 10,000 tons [10]. It can be considered to utilize this amount to produce energy instead of totally relying on import nuclear fuel.

Table 1.3. Thorium reserves by country [10]

Country	Tons	% of World Total
India	846,000	16
Turkey	774,000	14
Brazil	606,000	11
Australia	521,000	10
USA	434,000	8
Egypt	380,000	7
Norway	320,000	6
Venezuela	300,000	6
Canada	172,000	3
Russia	155,000	3
South Africa	148,000	3
China	100,000	2
Greenland	86,000	2
Finland	60,000	1
Sweden	50,000	1
Kazakhstan	50,000	1
Other countries	413,000	8
World Total	**5,385,000**	

From the technical point of view, one more reason that thorium is preferred over enriched uranium is that the breeding of ^{233}U from thorium is more efficient than the breeding plutonium from ^{238}U [11]. This is so because the thorium fuel creates fewer non-fissile isotopes. Fuel-cycle designers can take advantage of this efficiency to decrease the amount of spent fuel per unit of energy generated, which reduces the amount of waste to be disposed of.

Moreover, in ^{232}Th–^{233}U fuel cycle, much lesser quantity of plutonium and long-lived Minor Actinides (MA: Np, Am and Cm) are formed as compared to the ^{238}U–^{239}Pu fuel cycle, thereby minimizing the radiotoxicity associated in spent fuel. Following the thorium decay chain in the Fig. 1.3 (the highlighted one), there are two isotopes , namely ^{233}U and ^{235}U, for which the fission cross section is much larger than capture

cross section, therefore greatly reducing the production of TRUs [4]. That is the advanced benefit of the utilization of thorium fuel in the TRU transmutation system for such as ADS. However, in the back end of ^{232}Th–^{233}U fuel cycle, there are other radionuclides such as ^{231}Pa, ^{229}Th and ^{230}U, which may have long term radiological impact.

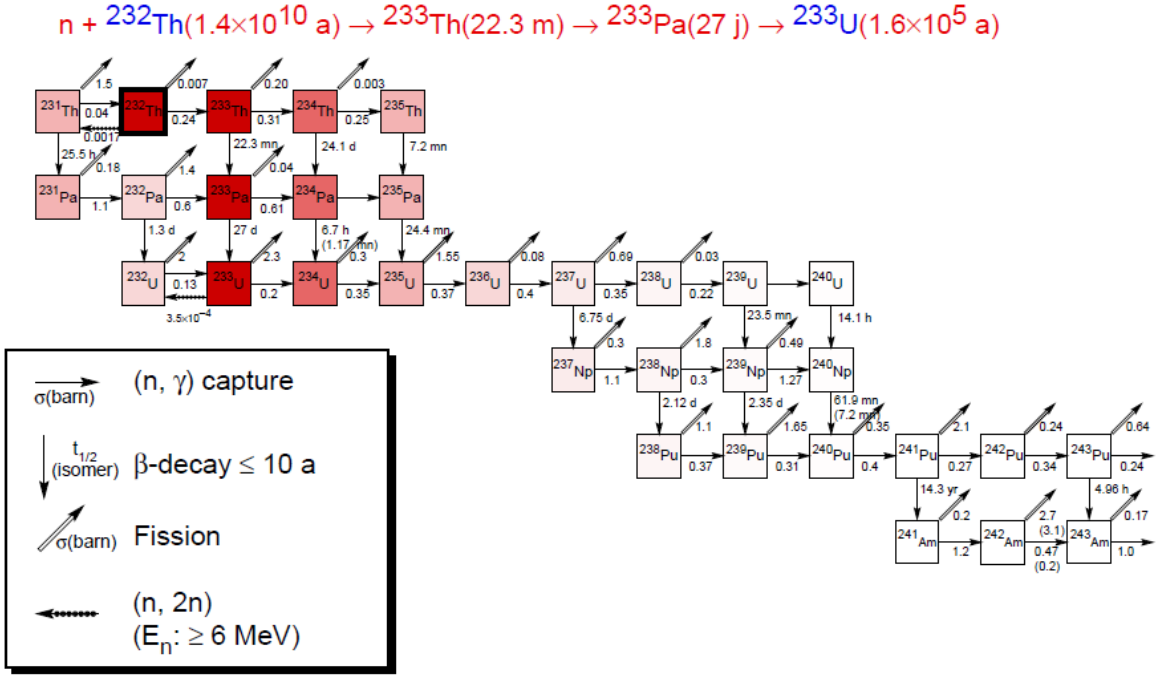


Fig. 1.3. Decay chains of different actinides in the thorium and uranium fuel cycle [12]

Other benefits of thorium fuel cycle are analyzed in the IAEA report as follows [13]:

For the 'fissile' ^{233}U nuclei, the number of neutrons liberated per neutron absorbed (represented as η) is greater than 2.0 over a wide range of thermal neutron spectrum, unlike ^{235}U and ^{239}Pu. Thus, contrary to ^{238}U–^{239}Pu cycle in which breeding can be obtained only with fast neutron spectra, the ^{232}Th–^{233}U fuel cycle can operate with fast, epithermal or thermal spectra.

Besides, thorium dioxide is chemically more stable and has higher radiation resistance than uranium dioxide. The fission product release rates for ThO_2–based fuels are one order of magnitude lower than that of

UO_2. ThO_2 has favorable thermophysical properties because of the higher thermal conductivity and lower co-efficient of thermal expansion compared to UO_2. Thus, ThO_2–based fuels are expected to have better in–pile performance than that of UO_2 and UO_2–based mixed oxide. ThO_2 is relatively inert and does not oxidize unlike UO_2, which oxidizes easily to U_3O_8 and UO_3. Hence, long term interim storage and permanent disposal in repository of spent ThO_2–based fuel are simpler without the problem of oxidation.

Compared with U – based fuel, Th–based fuels and fuel cycles have intrinsic proliferation-resistance due to the formation of ^{232}U via (n, 2n) reactions with ^{232}Th, ^{233}Pa and ^{233}U. The half-life of ^{232}U is only 73.6 years and the daughter products have very short half-life and some like ^{212}Bi and ^{208}Tl emit strong gamma radiations. From the same consideration, ^{232}U could be utilized as an attractive carrier of highly enriched uranium (HEU) and weapons grade plutonium (WPu) to avoid their proliferation for non-peaceful purpose.

Beside these potential benefits, the utilization of thorium fuel has to face with some challenges as listed in the same report. One of those is that the melting point of ThO_2 ($3{,}350^0C$) is much higher compared to that of UO_2 ($2{,}800^0C$). Hence, a much higher sintering temperature ($>2{,}000^0C$) is required to produce high density ThO_2 and ThO_2–based mixed oxide fuels. Admixing of 'sintering aid' (CaO, MgO, Nb_2O_5, etc) is required for achieving the desired pellet density at lower temperature. On the other hand, ThO_2 and ThO_2–based mixed oxide fuels are relatively inert and, unlike UO_2 and (U, Pu)O_2 fuels, do not dissolve easily in concentrated nitric acid. Addition of small quantities of HF in concentrated HNO_3 is essential which cause corrosion of stainless steel equipment and pipings in reprocessing plants. The corrosion problem is mitigated with addition of aluminium nitrate. Boiling THOREX solution [13 M HNO_3+0.05 M HF+0.1 M $Al(NO_3)_3$] at ~393 K and long dissolution period are required for ThO_2–based fuels. The irradiated thorium or thorium – based fuels contain significant amount of ^{232}U, which has a half-life of only 73.6 years and is associated with strong gamma emitting daughter products, ^{212}Bi and ^{208}Tl with very short half-life. As a result, there is significant buildup of radiation dose with storage of spent thorium –based fuel or separated ^{233}U, necessitating remote and automated reprocessing and refabrication in heavily shielded hot cells and increase in the cost of fuel cycle activities.

In the conversion chain of ^{232}Th to ^{233}U, ^{233}Pa is formed as an intermediate, which has a relatively longer half-life (~27 days) as compared to ^{239}Np (2.35 days) in the uranium fuel cycle thereby requiring longer cooling time of at least one year for completing the decay of ^{233}Pa to ^{233}U. Normally, Pa is passed into the fission product waste in the THOREX process, which could have long term radiological impact. It is essential to separate Pa from the spent fuel solution prior to solvent extraction process for separation of ^{233}U and thorium.

Moreover, the three stream process of separation of uranium, plutonium and thorium from spent (Th, Pu)O_2 fuel, though viable, is yet to be developed.

Since the database and experience of thorium fuels and thorium fuel cycles are very limited, as compared to UO_2 and (U, Pu)O_2 fuels. Thorium data is needed to be verified and its lack and knowledge are needed to be aware of before large investments are made for commercial utilization of thorium fuels and fuel cycles.

Even though much more efforts have to be made to eventually utilize thorium fuel to produce energy but it shows a great potential to be a fuel candidate to replace uranium fuel. In this study, thorium is chosen as based fuel of the ADS system and its utilization is investigated.

1.2 Purpose of this thesis

In order to find the solution for the spent nuclear fuel problem and energy security using the natural abundant resource, the purpose of this research work is to propose a conceptual design of the seed and blanket ADS for efficient TRU transmutation and power generation using thorium-reprocessed fuel and its application in the case of nuclear power installation in Vietnam. Since the seed and blanket ADS is multi-purpose facility, the design process is done step by step. Therefore, a specific goal is set for each step.

At first, the purpose of the investigation is is to provide the design of the seed and blanket ADS for efficient TRU transmutation thorium-reprocessed fuel and investigate its safety features.

In Chapter 2 of this thesis, the design parameter and core configuration of seed and blanket ADS for efficient TRU transmutation using the thorium-reprocessed fuel are described. In this chapter, the innovative point of the design compared to other designs is explained. In order to achieve the efficient transmutation, the TRU transmutation performance and the safety features of the system with different design parameters are examined. The ADS core with high transmutation rate and safe operation is chosen.

Since the utilization of thorium fuel in the seed and blanket ADS after one cycle is limited, therefore:

The multi-cycle seed and blanket ADS is proposed in order to achieve the high utilization of thorium fuel for energy production.

The multi-cycle design concept of the seed and blanket ADS in order to achieve the high utilization of thorium fuel assemblies for power production is provided in Chapter 3. Instead of removing all the fuel assemblies out of the core after burning for one cycle (430 days), the reprocessed fuel assemblies is shuffled for a second burnt while thorium fuel assemblies are maintained inside the reactor core until the end of core life. TRU transmutation rate and thorium utilization of the multi-cycle core is compared with the single one to show the improvement in thorium utilization and TRU transmutation of the multi-cycle concept.

After the TRU transmutation and thorium utilization potential of seed and blanket ADS using thorium-reprocessed fuel are examined, its application in a certain circumstance is needed to be considered.

Hence, the application of the seed and blanket ADS in the nuclear power installation in Vietnam is investigated.

The content of this study is presented in Chapter 4. In this chapter, the nuclear power employment in Vietnam in the future is described. The amount of spent nuclear fuel of future reactors in Vietnam is calculated and the benefit of the installation of seed and blanket ADS in that situation is also evaluated.

However, considering the limitation of thorium cross section accuracy and its impact on the seed and blanket ADS evaluation data, an additional study was done. The aim of this step is:

To verify the thorium cross section data using the experimental and calculated data and investigate the impact of thorium cross section uncertainty on seed and blanket ADS reactivity calculation.

The performance of the additional study is described in Chapter 5 of this thesis. The impact of thorium cross section uncertainty on seed and blanket ADS calculation is discussed whether the current thorium cross section data is reliable for the ADS core design and safety evaluation.

Finally, the obtained results are summarized and the main conclusions are drawn in the Chapter 6.

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Chapter 2: **Seed and blanket ADS using thorium-reprocessed fuel: parametric survey on transmutation performance and safety characteristics**

2.1. Introduction

Recently, ADS with its inherent safety feature and waste transmutation potential has attracted more and more attention in the reactor physics study. There are active programs in many countries to develop, demonstrate and exploit ADSs technology for nuclear waste transmutation and power generation [1]. The first detailed design of a transmutation facility using thermal neutrons was published by C. Bowman's Los Alamos group in 1991 introducing a common name "The Accelerator Transmutation of Waste" (ATW) [2&3]. The subcritical core with the TRUs loading only has the large reactivity swing due to burnup. A frequent refueling is required during operation for such core. In 1993, the basic concept of a so-called "Energy Amplifier" which is a subcritical nuclear system based on thorium cycle intended to generate energy with small amount of MAs and long life fission fragments (LLFFs) was presented by a group of CERN scientists [4]. Thorium and TRUs are mixed and served as fuel in the ADS. The disadvantage of the Th-Pu fuel cycle is the back end issue. There is the lack of experience and know-how on back end issue of the Th-Pu fuel cycle [5]. Thorium fuel and reprocessed fuel is better to separate for easier in-core fuel management and spent fuel management. Moreover, it was found that by spatially separating the fertile and fissile materials, ideally, the competition for neutron absorption between them can be eliminated, thus, the capture rate in thorium fuel would be maximized and ^{233}U conversion ratio in the blanket could be enhanced [6]. Besides, the heterogeneous core design will help to reduce the power peaking factor due to burnup since the power contribution of the fertile blanket will increase with time of radiation. Therefore, in this thesis, the seed and blanket ADS design with separated reprocessed fuel assemblies and thorium fuel assemblies for TRU transmutation and energy production is presented. Since the seed and blanket ADS is multi-purpose facility, the design process is done

step by step. In this Chapter, the design parameter and core configuration of the seed and blanket ADS for efficient TRU transmutation using thorium-reprocessed fuel are described. In order to achieve the efficient transmutation, the TRU transmutation performance and the safety features of the system with different design parameters are examined. The ADS core with high transmutation rate and safe operation is chosen.

2.2. Seed and blanket ADS design and calculation tool

2.2.1. ADS for nuclear waste transmutation

The accelerator-driven transmutation system includes 3 major components: the neutron generated accelerator, the spallation target and the subcritical reactor core as shown in the schematic view in the Fig. 2.1. As illustrated in the figure, chain reaction inside the subcritical core is maintained by the spallation neutrons produced from the spallation reaction of the accelerated protons and the target. k_{eff} less than 1.0 is intrinsic safety feature of the ADS. The core will shut down when the accelerator shuts down. Moreover, it allows the system to burn the fuels that would degrade neutronic characteristics of the critical core to unacceptable level such as ^{233}U and minor actinide fuel. The heavy materials are commonly used as the spallation target of the ADS, such as lead-bismuth eutectic or tungsten. The spallation neutrons emitted from the target are used to transmute the actinides loaded in the blanket surrounding the target [7].

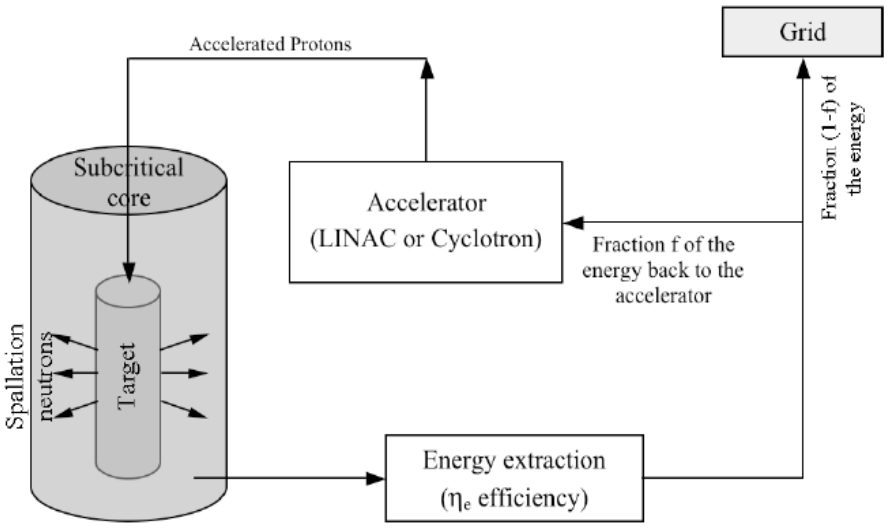


Fig. 2.1. Schematic view of main components of an ADS

2.2.2. Design parameter of seed and blanket ADS.

In this study, LBE is chosen as the target of ADS. Neutrons are produced by the spallation reaction of 1 GeV proton beam bombarded to the LBE cylindrical target placed in the center of the system. Since the ADS design has k_{eff} at beginning of cycle about 0.97 and k_{eff} at the end of cycle about 0.90, the neutron beam current is increased from 13 mA to 30 mA at EOC in order to compensate the reducing of k_{eff} through burnup. The fast spectrum ADS is designed to yield higher actinide fission-to-capture ratios, so more TRU can be transmuted compared to the thermal system. Thus, the tight pitch configuration is employed for assemblies of the ADS core. The model of ADS using in this simulation was conducted from the typical fast neutron spectrum, lead-bismuth accelerator - driven transmutation system in H. R. Trellue's research [7]. The innovation of the core design in this study compared to the previous designs is that the subcritical core is separated with individual seed and blanket assemblies. The fuel reprocessed from LWRs is employed as the seed to start fission reaction inside the core. Thorium fuel with its abundance in nature and less TRUs producing feature is very favorable to be the based fuel of ADS in terms of energy producing and eliminating

TRUs. Thorium fuel is served as blanket in the seed and blanket ADS design.

In subcritical core of reference seed and blanket ADS core configuration for the preliminary study, 96 seed assemblies of reprocessed fuel and 84 blanket assemblies of thorium fuel were inserted into the cylindrical core of 220 cm diameter. The vertical and horizontal sectional views of the reference the seed and blanket ADS is shown in Fig. 2.2.

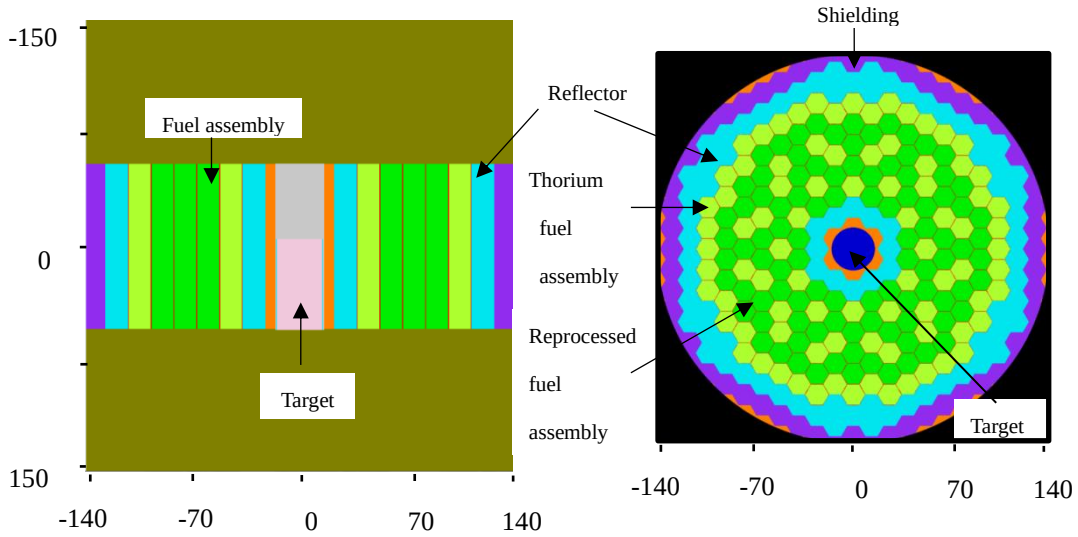


Fig.2.2. Vertical and horizontal sectional views of the thorium – reprocessed fuel ADS design

(Scale is given in cm)

The benefits of heterogeneous core design are simplifying the assembly fabrication, in-core fuel management and spent fuel management. It also helps to reduce the power peaking factor problem through burnup since the power contribution of the blanket is increased with time of radiation. Moreover, by spatially separating the fertile and fissile materials, ideally, the competition for neutron absorption between them can be eliminated, thus, the capture rate in thorium fuel would be maximized and ^{233}U conversion ratio in the blanket could be enhanced. Besides producing energy, the breeding ^{233}U from thorium compensates the burnt TRUs in reprocessed fuel, thus, reducing the reactivity swing. The thorium blanket assemblies are placed at the periphery of the core in order to improve the neutron economy.

Reprocessed fuel is recovered from the reprocessing scheme assumed in K. Tsujimoto research [8]. From that, the spent PWR fuel of 45 GWd/t burnup was reprocessed after 7 years cooling and MA and Pu were recovered using PUREX and MA is enriched for enhanced MA burning. Thorium and reprocessed fuels are introduced into the core in the oxide form. The initial content of thorium and reprocessed fuel is determined in order to achieve the initial k_{eff} of about 0.97. This value of multiplication coefficient was chosen in order to avoid the criticality conditions, with adequate safety margins [4]. The isotopic composition of thorium and reprocessed fuel is shown in Table 2.1.

Table 2.1. Composition of reprocessed fuel and thorium fuel

Actinides		**Number of atom density** (atom/b-cm)
Reprocessed fuel	^{235}U	4.177E-07
	^{236}U	9.462E-08
	^{237}Np	2.664E-04
	^{238}Pu	2.342E-04
	^{239}Pu	3.774E-03
	^{240}Pu	1.676E-03
	^{241}Pu	7.519E-04
	^{242}Pu	4.823E-04
	^{241}Am	1.818E-04
	^{242}Am	3.188E-07
	^{243}Am	7.214E-05
	^{243}Cm	1.677E-07
	^{244}Cm	2.074E-05
	^{245}Cm	2.448E-06
	^{246}Cm	2.622E-07
	^{16}O	1.490E-02
Thorium fuel	^{232}Th	2.1641E-02
	^{16}O	4.3282E-02

The main design parameters of the thorium – reprocessed fuel ADS is given in Table 2.2. The LBE target is bombarded with 1 GeV proton beam and creates about 30 spallation neutrons per proton. Since the LBE spallation products ^{208}Po and ^{209}Po contribute significantly to radiotoxicity, so a more inert coolant like sodium is employed as coolant of the system [7].

Table 2.2. Design parameters of seed and blanket ADS

Reactor	
Thermal power (MW_{th})	840
Fuel temperature (K)	900
Coolant temperature (K)	600
Structure material temperature (K)	600
Effective full power days (EFPDs)	430
Fuel type	Oxide
Coolant	Sodium
Thorium assemblies/reprocessed fuel assemblies	96/84
Core diameter (cm)	220
Number of pins per assembly	271
Length of pin (cm)	130
Fuel pin radius (cm)	0.36
Pitch of pin (cm)	1.03
Pitch of assembly (cm)	14.71
Accelerator	
Proton energy (GeV)	1
Currents (mA)	13~30
Spallation neutron yield (n/p)	~30

In this chapter, some parameters of the core are changed and the transmutation efficiency and safety parameters are examined. One of the survey parameter is the MA content. The MA is increased from the original composition in Table 2.1 by 2.5 and 5 times and hereafter is mentioned as 2.5 time enriched MA and 5 time enriched MA cases. Another investigation is done with the survey parameter is the core size. More fuel is

loaded into the system by increasing number of fuel assemblies of the reactor, thus enlarging the reactor radial size. Its sectional view is distinguished with the original configuration in Fig. 2.3. The reprocessed fuel composition is adjusted in order to maintain the reactivity of the system.

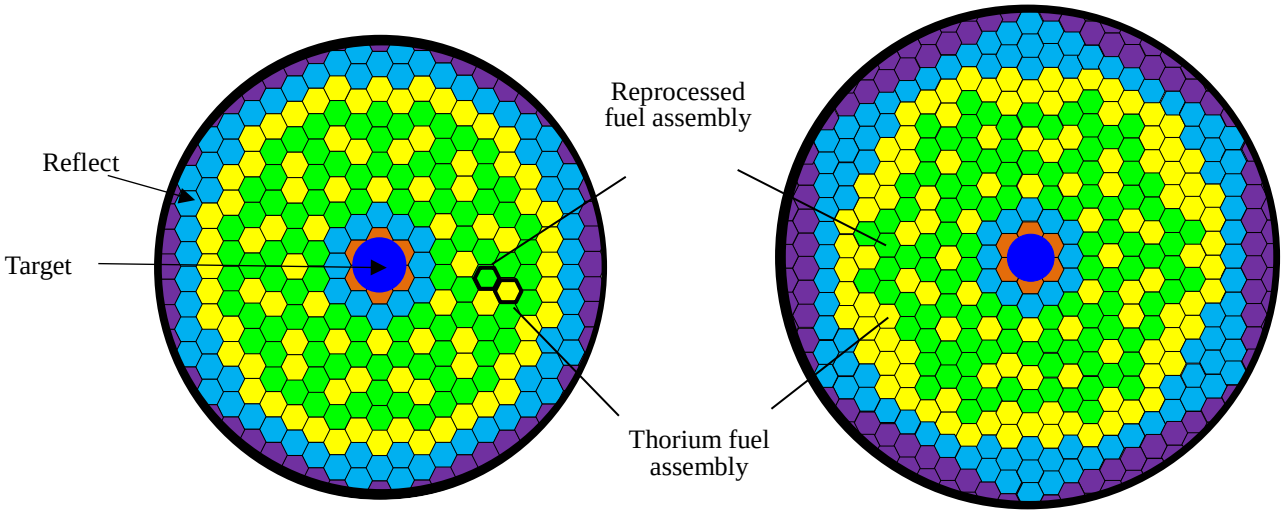


Fig.2.3. Sectional views of the seed and blanket ADS design with different core sizes:

Original core (left) and enlarged core (right)

2.2.3 Calculation method

The Monte Carlo code MCNPX 2.7.0 [9] with ENDF/B-VII.0 cross section library [10] was used for evaluation in this study. MCNP is a general purpose Monte Carlo N-Particle core that can be used for neutron, photon, electron or couple neutron/photon/electron transport [11]. Monte Carlo technique used in MCNP is a statistical method in which estimations for particle characteristics are obtained through multiple computer simulations of the behavior of individual particles in a system [7]. MCNPX is the extended version of MCNP where the major capability of LAHET code [12] and MCNP5 have been merged together. In MCNP, particle transport relies entirely on nuclear data contained in externally supplied cross section tables (E_n< 20 MeV), which are derived from evaluated nuclear data files. In LAHET, on the other hand, particle transport is accomplished by using various theoretical physics models embedded in the code, covering the energy range

up to several GeV [13]. The combination of spallation source (external source-SDEF) and kcode-mode is used for calculation flux and k_{eff} of the ADS. Burnup capability is performed via linked process involving steady-state flux calculations by MCNPX and nuclide depletion calculation by CINDER90 [14]. CINDER90 is a burnup code developed at Los Alamos National Laboratory specifically for unique applications (such as accelerators) because it tracks both fission and spallation products as a function of time. In addition, it uses multi-group (63-group) fluxes and cross section instead of the simplified one-group ones used by ORIGEN2.1 [7]. The code is unable to take into account the flux from external source during burnup calculation. However, such approximated results are reasonable as long as the external source would not be sufficient to drive the system towards criticality [15]. Thus, results can be used in the transmutation capability analysis with the change of core configuration and fuel composition. The thorium – reprocessed fuel ADS burnup calculations were done for 430 effective full power days with 500,000 numbers of histories and standard deviation for k_{eff} of 0.0006.

2.3. Performance characteristics of seed and blanket ADS

The main neutronic parameters at beginning of cycle (BOC) and the end of cycle (EOC) for the seed and blanket ADS reference configuration are summarized in Table 2.3. In this calculation, 96 thorium fuel assemblies and 84 reprocessed fuel assemblies which are equivalent to about 8.32 tons of heavy metal (HM) is loaded at BOC and burnt to produce 840 MW_{th} power. The burnup calculation is fixed for 430 effective full power days (EFPDs). The TRU transmutation rate is defined as the kilogram of transmuted TRUs per TWh. As we can see from the Table 2.3, it shows a good potential of TRUs transmutation with 39.3 kg of TRU per TWh. However, the reactivity swing due to burnup is large (0.112 Δk). The effective multiplication factor at EOC is of about 0.863. Such a deep subcriticality like this will require a very high source beam current (60 mA) from the accelerator in order to maintain the chain reaction inside the core. Nevertheless, such high proton beam current and power (60 MW) is very expensive and still under investigation. Therefore, the

reactivity swing of the system is required to be reduced in order to employ the seed and blanket ADS for TRU transmutation. The void reactivity for 40% void is -0.053 △k/k (Table 2.5). The void coefficient reduces (becoming more negative) and reaches a value of -0.164 △k/k for the case of complete void. This negative void coefficient is an important indication of stability of the system in case of loss of coolant.

Table 2.3. Performance characteristics of the reference seed and blanket ADS

Characteristics	Seed and blanket ADS
Thorium assemblies/reprocessed fuel assemblies	96/84
Initial core loading (HM/TRUs) MT	8.32/1.88
Transmutation rate (kg per TWh)	39.3
Radial PPF at BOC	2.73
Axial PPF at BOC	1.20
Radial PPF at EOC	2.25
Axial PPF at EOC	1.17
Void coefficient (at 40%) Δk/k	-0.079
k_{eff} BOC EOC	 0.9751±0.0006 0.8634±0.0006
Burnup reactivity swing (Δk)	0.112

Neutron energy spectra for different regions in ADS core is demonstrated in Figure 2.4. It clearly shows the domination of neutron in the fast energy range. The 3 keV resonance of sodium coolant is the reason to cause the flux “drop” at that region. As mentioned earlier, the fast spectrum systems are advantageous for actinide transform since the actinide fission to capture ratio is higher compared with the thermal system [7]. Therefore, it is more efficient for destructions of actinides while less minor actinide is built up.

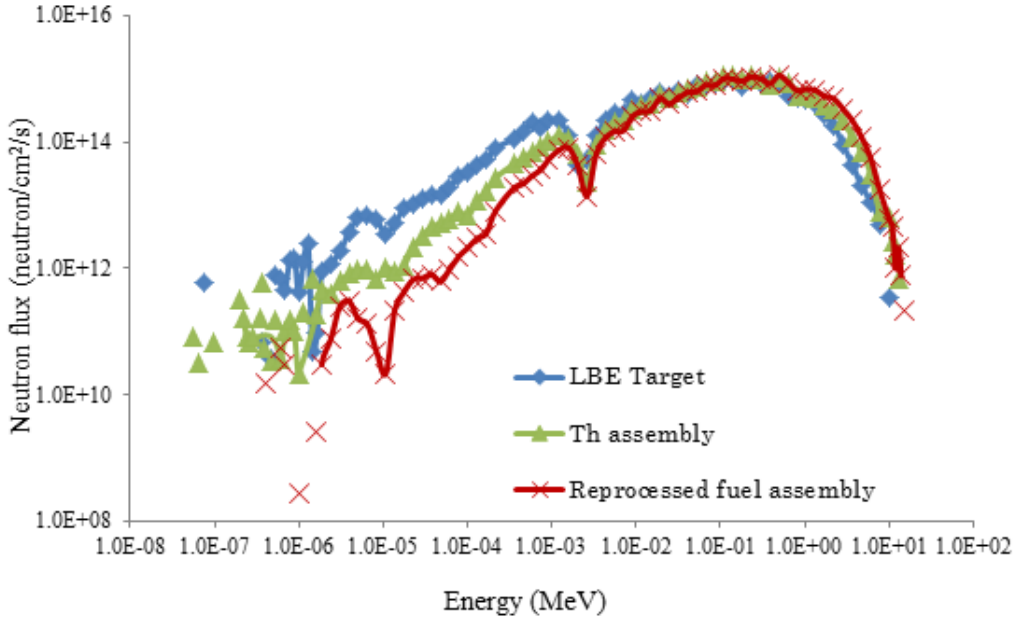


Fig. 2.4. Neutron energy spectra for different regions in ADS core

2.4. Transmutation performance and safety characteristics of the seed and blanket ADS with different parameters

2.4.1. Different MA contents

As discussed above, the calculation result of the seed and blanket ADS reference configuration with the original MA content (without enrichment) shows a large reactivity swing due to burnup which are unfavorable for the ADS system. Thus, it is necessary to reduce the reactivity swing as well as improve the transmutation capability of the system. In this parameter survey, MA is an object to be investigated. The MA content is increased from the original content in Table 2.4 by 2.5 and 5 times. The reprocessed fuel composition is adjusted in order to maintain the reactivity of the system. The number densities of isotopes of reprocessed fuel for 3 cases are shown in Table 2.4.

Table 2.4. Number densities of isotopes of reprocessed fuel with different MA enrichments

Actinides	**Original MA content** (atom/b-cm)	**2.5 time enriched MA content** (atom/b-cm)	**5 time enriched MA content** (atom/b-cm)
^{235}U	4.177E-07	4.177E-07	4.177E-07
^{236}U	9.462E-08	9.462E-08	9.462E-08
^{237}Np	2.664E-04	6.483E-04	1.332E-03
^{238}Pu	2.342E-04	2.066E-04	1.605E-04
^{239}Pu	3.774E-03	3.329E-03	2.587E-03
^{240}Pu	1.676E-03	1.479E-03	1.149E-03
^{241}Pu	7.519E-04	6.633E-04	5.154E-04
^{242}Pu	4.823E-04	4.255E-04	3.306E-04
^{241}Am	1.818E-04	4.546E-04	9.092E-04
^{242}Am	3.188E-07	7.970E-07	1.594E-06
^{243}Am	7.214E-05	1.804E-04	3.607E-04
^{243}Cm	1.677E-07	4.192E-07	8.384E-07
^{244}Cm	2.074E-05	5.185E-05	1.037E-04
^{245}Cm	2.448E-06	6.120E-06	1.224E-05
^{246}Cm	2.622E-07	6.555E-07	1.311E-06
^{16}O	1.490E-0.2	1.490E-0.2	1.490E-0.2

The performance of the cores is shown in Table 2.5.

Table 2.5. Performance characteristics of the seed and blanket ADS with different MA contents

Characteristics	**Seed and blanket ADS Original MA**	**Seed and blanket ADS 2.5 time enriched MA**	**Seed and blanket ADS 5 time enriched MA**
Transmutation rate (kg per TWh)	39.3	40.0	41.5
Radial PPF at BOC	2.73	2.64	2.50
Axial PPF at BOC	1.20	1.20	1.21
Radial PPF at EOC	2.25	2.11	1.99
Axial PPF at EOC	1.17	1.19	1.20
Void coefficient (at 40%) Δk/k	-0.079	-0.064	-0.043
Thorium utilization (%)	3.10	3.45	3.82
k_{eff}			
BOC	0.9751±0.0006	0.9682±0.0006	0.9591±0.0006
EOC	0.8634±0.0006	0.8716±0.0006	0.8983±0.0005
Burnup reactivity swing (Δk)	0.112	0.096	0.065

As shown in Table 2.5, with larger amount of MA, the TRU transmutation rate is increased and the void reactivity becomes less negative. With increasing amount of MA, more ^{242}Pu is converted from ^{242}Am (via the electron capture reaction), the Pu transmutation becomes less efficient and consequently, the Am annihilation is increased (Fig. 2.5). As we can notice the inventory change, contribution of Np transmutation to total TRU transmutation also becomes more important. As a result, the total TRU transmutation increases slightly with more MA introduced into the system. As we can notice from the Table 2.5, with the original MA content, only 3.1% of ^{232}Th is converted to ^{233}U and produces power. Its utilization increases with the increasing of MA amount. The larger converted Pu from Am and more breeding ^{233}U from utilized ^{232}Th help to reduce the reactivity swing of the system. In case of the void coefficient evaluation, when the void fraction increases, it hardens the neutron spectrum inside the reactor which is favorable to the ^{241}Am fission because it has threshold energy of fission at several hundred keV. Thus, with the increasing of ^{241}Am content, the fission of ^{241}Am inside the reactor increases and reduce the negative void reactivity. The large negative reactivity

effects is quite favorable for the ADS system, thus, the ratio of MA to heavy metal (HM) should be noticed in order to maintain the enough negative void reactivity for the ADS. In summary, MA with even small fraction in the weight is needed to be noticed because it can affect the safety feature of the system such as void reactivity remarkably. Besides, the radial and axial power peaking factors (PPF) at beginning of cycle (BOC) and end of cycle (EOC) at assembly-wise of the ADS with different MA contents are also calculated. As shown in the Table 2.5, the larger the MA content, the smaller the PPF which is preferable as core characteristics.

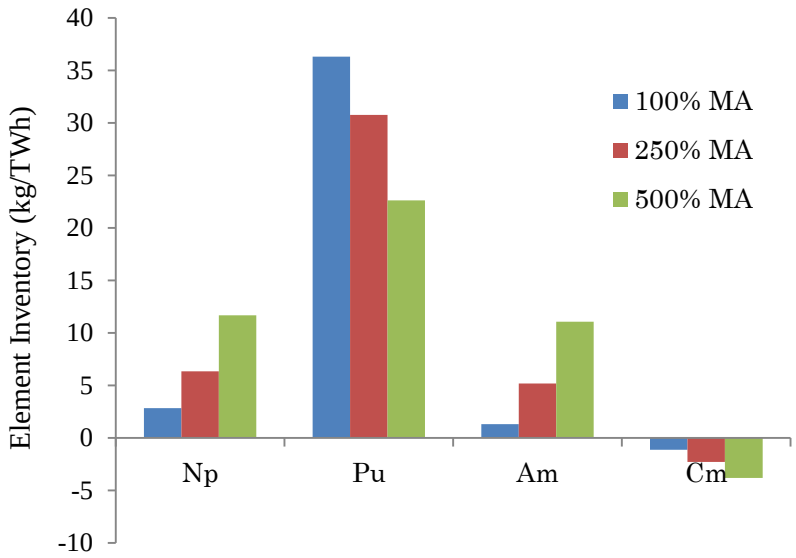


Fig.2.5. TRU inventories of seed and blanket ADS with different MA contents

2.4.2. Different core sizes

Another approach to reduce the reactivity swing of the ADS design is increasing the thorium fuel to reprocessed fuel ratio inside the system. In order to increase the thorium fuel to reprocessed fuel ratio and also maintain the reactivity of the system, more thorium and reprocessed fuel assemblies are inserted into the core, thus, increase the radial size of the ADS core. The number of thorium fuel assemblies is increased from 84 to

114 assemblies and the number of reprocessed fuel assemblies from 96 to 108 assemblies. The TRU content used in this calculation is same as it of the case MA enriched 5 times. The transmutation and the neutronics performance of the core is calculated and compared with the original core. The result is shown in Table 2.6.

Table 2.6. Performance characteristics of seed and blanket ADS with different core sizes

Characteristic	**Seed and blanket ADS Original**	**Seed and blanket ADS Larger core**
Thorium assemblies/reprocessed fuel assemblies	96/84	114/108
Initial core loading (HM/TRUs) MT	9.07/2.63	11.6/2.81
Transmutation rate (kg per TWh)	41.5	41.1
Radial PPF at BOC	2.50	2.60
Axial PPF at BOC	1.21	1.17
Radial PPF at EOC	1.99	2.38
Axial PPF at EOC	1.20	1.15
Void coefficient (at 40%) Δk/k	0.043	0.039
Thorium utilization (%)	3.10	2.74
k_{eff} BOC EOC	 0.9591±0.0006 0.8983±0.0005	 0.9555±0.0006 0.9070±0.0006
Burnup reactivity swing (Δk)	0.065	0.049

When increasing the thorium fuel to reprocessed fuel ratio by adding more thorium and reprocessed fuel assemblies into the core, the more breeding ^{233}U from thorium fuel compensates the burnt TRU and reduce the reactivity swing from 0.065 to 0.049. In order to maintain the reactivity and the PPF of the system, the core arrangement are changed compared to the original core arrangement. This causes the slight reduction in TRU transmutation rate of the larger core compared with it of the original core (Table 2.6). The PPFs of the system with different core sizes are shown in Table 2.6. For void reactivity effect, it comes from both the changing spectrum and the neutron leakage during voiding. The void effect is decreased because of the large amount of thorium is presented in the larger core configuration while the more leakage came from by the

flatten core configuration cause more negative reactivity. It results in the slight reduction of the void effect when the reactor core is enlarged.

2.5. Conclusions

The seed and blanket ADS core concept using thorium-reprocessed fuel is preliminarily investigated in this chapter. With the reference configuration of the ADS, it shows a great potential of transmuting TRUs but the reactivity swing and the void coefficient is large and is an obstacle for employing this core concept technically. Hence, the parametric survey on seed and blanket fuel ADS was done in order to improve the core performance characteristics. It was found out that:

1. In the seed and blanket ADS, the larger amount of MA introducing into the core, the more TRUs are transmuted. The smaller reactivity swing in the system can be achieved by getting more efficient thorium utilization. Besides, the increasing MA composition also reduces negative void reactivity of the system.
2. Increasing the core size by introducing more thorium and reprocessed fuel assemblies into the system reduces the reactivity swing as well as the void reactivity of the system. It also slightly reduces the TRU transmutation efficiency of the system.

In multi-purpose seed and blanket ADS design, it is important to balance the transmutation efficiency and thorium utilization ability of the system with the safety margin is reserved. Since the starting reactivity of the core is about 0.97, the reactivity swing of the core should be about 0.07 in order to avoid the deep subcriticality situation which requires too high source beam current from the accelerator. Besides, the void reactivity should be kept negative enough by adjusting the amount of MA. That is, in order to match the safety feature during operation, the original seed and blanket ADS configuration using reprocessed fuel with enriched MA should be employed for efficient TRU transmutation and thorium utilization in the safe operation.

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Chapter 3: **Seed and blanket ADS using thorium-reprocessed fuel: multi-cycle approach for higher thorium utilization and TRU transmutation**

3.1. Introduction

As discussed in the previous chapter, the seed and blanket ADS using thorium – reprocessed fuel was investigated, illustrating its great potential for TRUs transmutation and in-core fuel management. Besides, thorium fuel has been utilized as based fuel instead of uranium fuel showing its advantages in terms of low actinide production and fuel availability. It is important implication of the ADS system for TRU transmutation and energy producing design purpose. However, for single cycle seed and blanket ADS, the utilization of thorium is very limited, only about 3% of thorium fuel is converted into ^{233}U and burnt in the ADS system. Instead of wasting the breeding ^{233}U from the thorium fuel assemblies, this fissile ^{233}U should be utilized to produce energy using the ADS system. Besides, it is taken into account that the once burnt ADS fuel can be recycled and burnt in a deeper level. Therefore, multi – cycle ADS is proposed for more efficient TRU transmutation and thorium utilization extension. According to the proposal, the reprocessed fuel assemblies are shuffled and burnt twice while the thorium assemblies are remained inside the reactor until the end of the reactor life. The thorium utilization, transmutation rate, and other neutronic parameters of the core at equilibrium cycle and the radiotoxicity at discharge are also investigated and presented in this chapter.

3.2. Multi-cycle seed and blanket ADS

Different with the single cycle seed and blanket ADS, the subcritical core of the multi-cycle ADS is divided into three individual regions: thorium fuel assemblies (TFA), inner reprocessed fuel assemblies (IFAs) and outer reprocessed fuel assemblies (OFAs). Inner and outer reprocessed fuel assemblies have the same

composition at the first cycle (Fig.3.1a). After the first cycle, thorium assemblies are remained inside the core until it is mainly utilized. After being burnt for one cycle, the inner reprocessed fuel assemblies are removed and the outer reprocessed fuel assemblies are shuffled into the inner fuel region for further radiation (Fig.3.1b). The fuel assemblies shuffle scheme is illustrated in Table 3.1. In order to compensate for the lower fission-to-capture rate caused by the decreasing of fissile materials due to fission and only the small amount of ^{233}U breeding after first cycle, the enrichment of the reprocessed fuel at the second cycle is higher compared with the subsequent ones. No other than that, the enrichment for subsequent cycle is consistent.

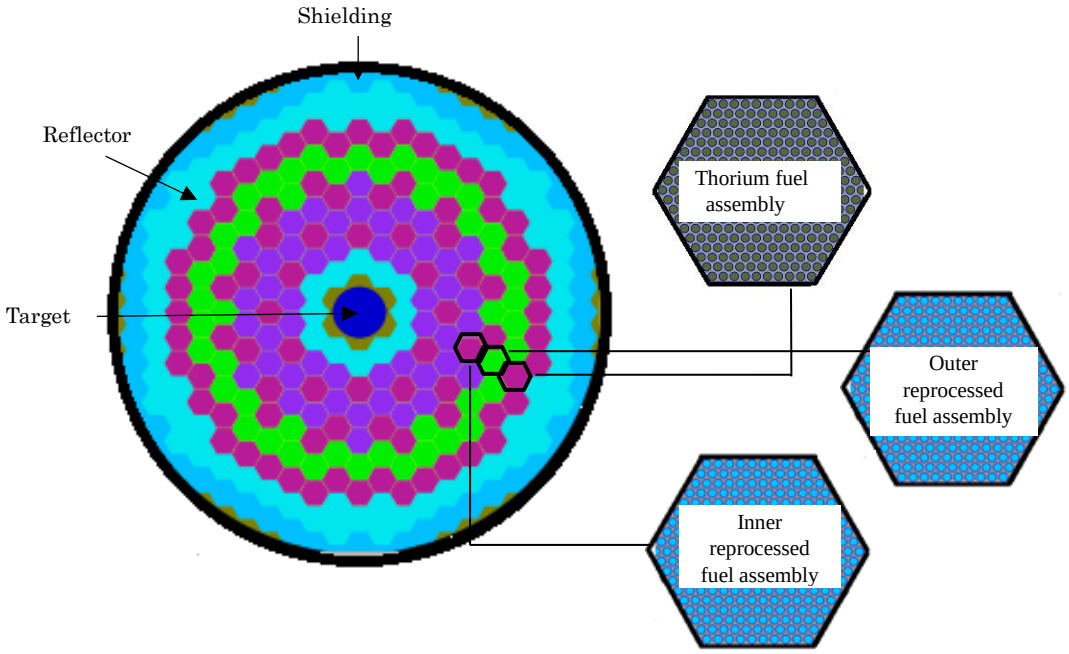


Fig 3.1a. Layout of seed and blanket ADS core at startup cycle

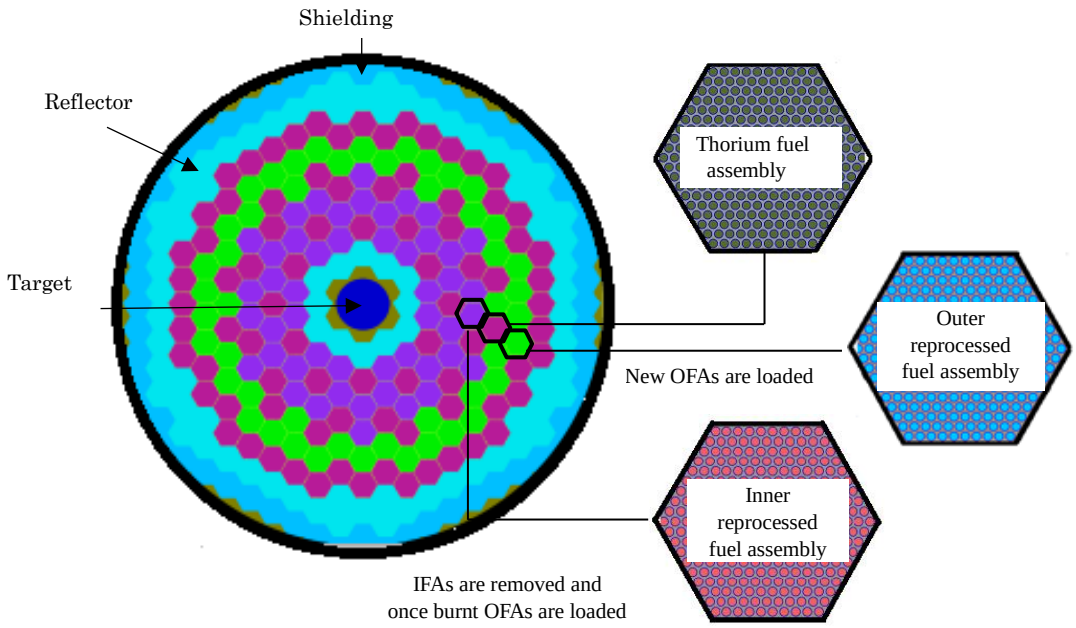


Fig 3.1b. Layout of seed and blanket ADS core at equilibrium state

Table 3.1. Fuel assemblies shuffle scheme for seed and blanket ADS for first 15 cycles.

Cycle	IFA	OFA	TFA
1	OFA1	OFA1	TFA1
2	OFA2	OFA1	TFA2
3	OFA2	OFA1	TFA3
4	OFA2	OFA1	TFA4
5	OFA2	OFA1	TFA5
6	OFA2	OFA1	TFA6
7	OFA2	OFA1	TFA7
8	OFA2	OFA1	TFA8
9	OFA2	OFA1	TFA9
10	OFA2	OFA1	TFA10
11	OFA2	OFA1	TFA11
12	OFA2	OFA1	TFA12
13	OFA2	OFA1	TFA13
14	OFA2	OFA1	TFA14
15	OFA2	OFA1	TFA15

3.3. Calculation tool

The calculations have been done using code MONTEBURNS code [1] with ENDF/B-VII.0 cross section library [2]. MONTEBURNS code is the package developed at Los Alamos National Laboratory that links the MCNP code or the related code MCNPX and the depletion code ORIGEN2 to conduct the burnup calculation. MONTEBURNS consists of a UNIX command file that call a FORTRAN77 program to process input and output file for MCNP and ORIGEN2 [3]. The MONTEBURNS calculation algorithm is illustrated in Fig. 3.2. ORIGEN2 is an isotope generation and depletion code used for performing radioactive decay and burnup analyses in a material. ORIGEN2 calculates material compositions halfway through each burnup step with initial cross section and flux information and these compositions are used to generate one-group cross section and flux in MCNP that represent the entire burnup step. ORIGEN2 then uses these cross sections and fluxes to calculate the material compositions at the end of each burnup step before proceeding to the next step. Having the capability to get detailed, system-dependent results and the flexibility to do automated feed and removal of material in multi-cycles is one of the major benefits of MONTEBURNS [4]. It is especially important for multi-cycle core design like this research because actinide cross section of the fuel varies as a function of burnup. The 840 MWth system was burnt 15 cycles using 20,000 number of histories. Each cycle (and also burnup step) is 430 days.

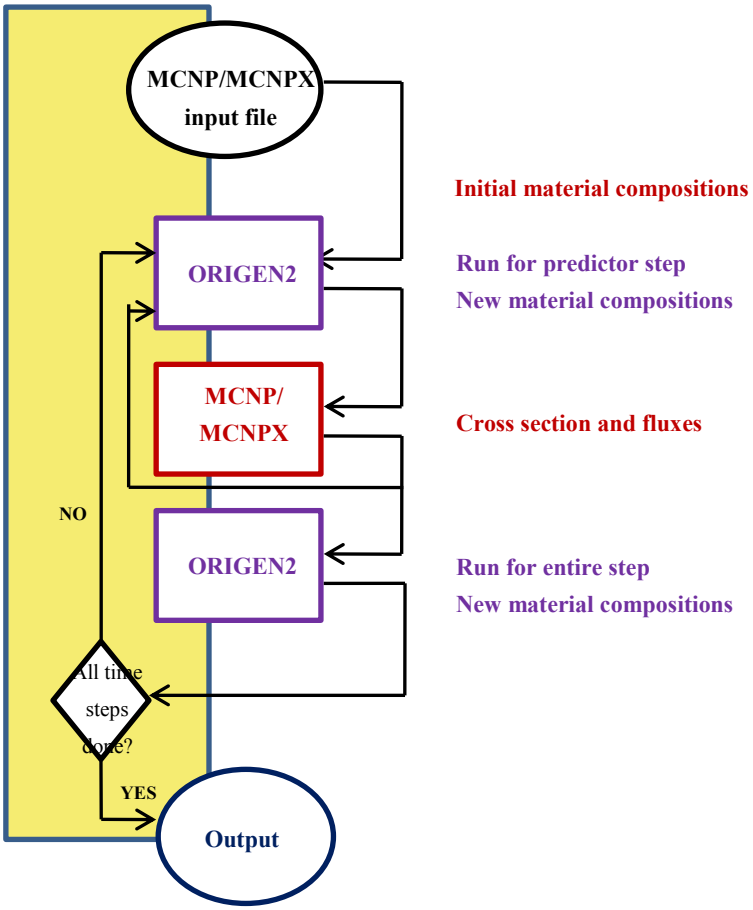


Fig. 3.2. MONTEBURNS calculation algorithm chart

3.4. Results of multi-cycle seed and blanket ADS core compared with the single cycle case

The seed and blanket ADS designed for transmuting the TRU waste in the reprocessed fuel is burnt 430 days each cycle. Between cycles, the thorium fuel assemblies are remained and the reprocessed fuel assemblies are shuffled. The once burnt reprocessed fuel assemblies are shuffled into the inner fuel region and new reprocessed fuel assemblies are loaded into the outer fuel region. After first several cycles, the core performances (such as effective multiplication factor, power distribution, TRU waste transmutation rate) are nearly unchanged until the end of reactor life (Fig. 3.3). In another word, the quasi-equilibrium state is attained and hereafter the quasi-equilibrium state will be mentioned as the equilibrium state and the cycle at

the quasi-equilibrium state will be mentioned as the equilibrium cycle of the seed and blanket ADS core.

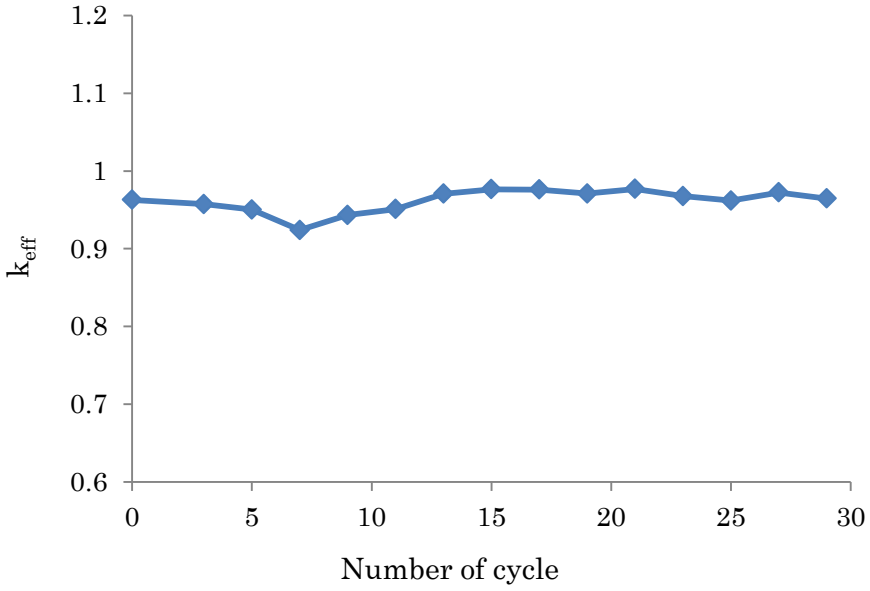


Fig. 3.3. Change in reactivity as function of time for the seed and blanket ADS

3.4.1 Thorium utilization

Thorium was proposed to be the based fuel in the seed and blanket ADS [5]. It shows the benefit in balancing the power peaking factor of the core due to burnup and the great potential of producing energy. The low concentration of TRUs when using thorium fuel in the ADS system is an important implication for transmutation designed purpose system [6]. In a single cycle core investigation, it was found that, at the end of cycle, only 3% of thorium is converted into ^{233}U and contributes about 15% to the power production of the whole core. If the thorium assemblies are removed from the reactor after one cycle, almost of thorium fuel hasn't been utilized and the back end process is required for bred ^{233}U in the spent thorium assemblies. Therefore, thorium and its breeding ^{233}U are considered to be remained inside the core for higher utilization of thorium and more power production. As shown in Table 3.2, during the equilibrium state, the contribution to the total power of the core of thorium fuel assemblies is much improved (about 35.7%) compared to it of the

startup cycle (15%). Moreover, at the end of reactor life (about 30 years), about 50% of thorium fuel is utilized.

Table 3.2. Power fraction from thorium fuel assemblies in startup and equilibrium cycles

	Power fraction from thorium fuel assemblies (%)
Startup cycle	15.0
Equilibrium cycle	35.7

3.4.2. Transmutation rate at equilibrium cycle

The TRU waste transmutation rate was estimated for both of the startup and equilibrium cycles in seed and blanke ADS core. The transmutation rate is computed in unit of kg TRU/ton of spent HM. TRU transmutation rate in startup and equilibrium cycles are presented in Table 3.3. From the table, it can be seen that the transmutation rate in the equilibrium cycle is about 124 kg and about 15 kg less than the transmutation rate in single cycle case. However, in the equilibrium cycle, the reprocessed fuel assemblies in the outer region are burnt and moved into the inner region to be burnt one more time. This helps to transmute more the TRU waste and the smaller amount of minor actinides is presented at discharge.

Table 3.3. TRU transmutation rate of startup and equilibrium cycles in the seed and blanket ADS core

	Transmutation rate (kg/ton of spent HM)
Startup cycle	139
Equilibrium cycle	124

3.4.3. Other main performance parameters at equilibrium cycle

Other key parameters of the multi-cycle core at equilibrium cycle are calculated and listed in Table 4. At equilibrium state, the reprocessed fuel assemblies in the inner region are the once burnt assemblies of the outer region, thus, its initial core loading is smaller than the startup cycle. With the same power output, the reprocessed fuel including TRU is burnt in a deeper level in the multi-cycle core. The assembly-wise power distribution of 1/6 core of the seed and blanket ADS at BOC and EOC for equilibrium state cycle are shown in Fig. 3.4 The utilization of thorium fuel is improved in the equilibrium state, so the axial PPF is much smaller compared to the startup cycle. The equilibrium cycle has larger void coefficient than the single one. The reactivity change of the system due to the variation of void fraction for equilibrium cycle at BOC and EOC is presented in Fig. 3.5.

Table 3.4. Main performance parameters of the seed and blanket ADS in startup and equilibrium cycles

	Startup cycle	**Equilibrium cycle**
Power output (MW_{th})	840	840
Cycle length (days)	430	430
Initial core loading (HM/TRU), MT	7.36/2.63	6.76/2.00
Seed – blanket fuel form	$TRUO_2/ThO_2$	$TRUO_2/ThO_2$
Axial PPF at BOC	2.50	1.42
Axial PPF at EOC	2.09	1.34
Radial PPF at BOC	1.21	1.21
Radial PPF at EOC	1.20	1.19
Void coefficient (40%) $\Delta k/k$	0.060	0.090

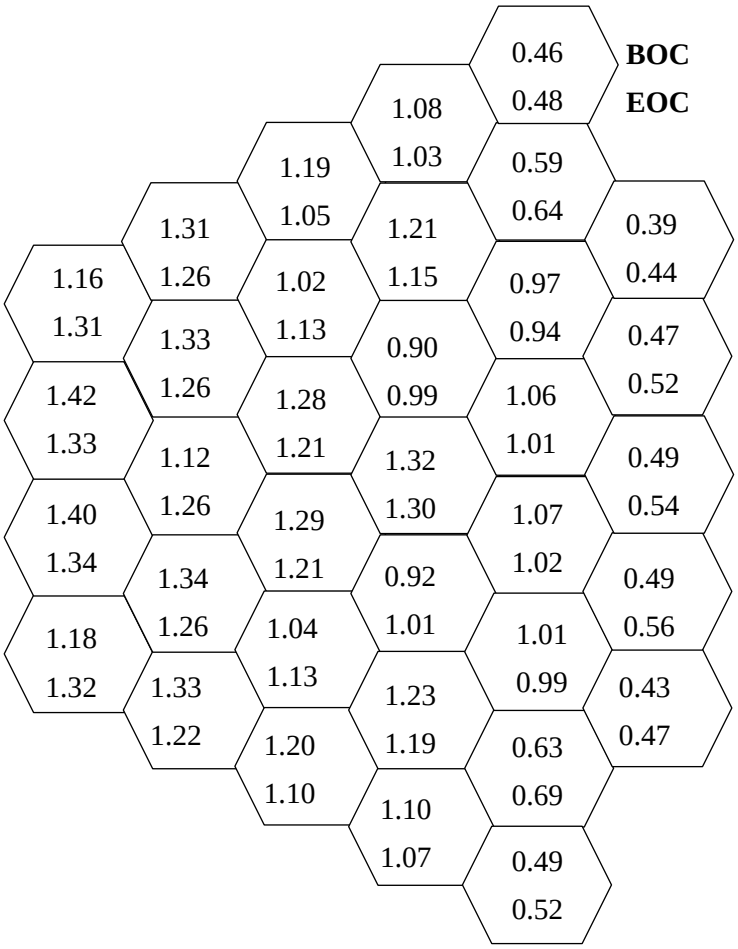


Fig. 3.4. Assembly-wise power distribution of 1/6 core of the seed and blanket ADS at BOC and EOC of equilibrium cycle

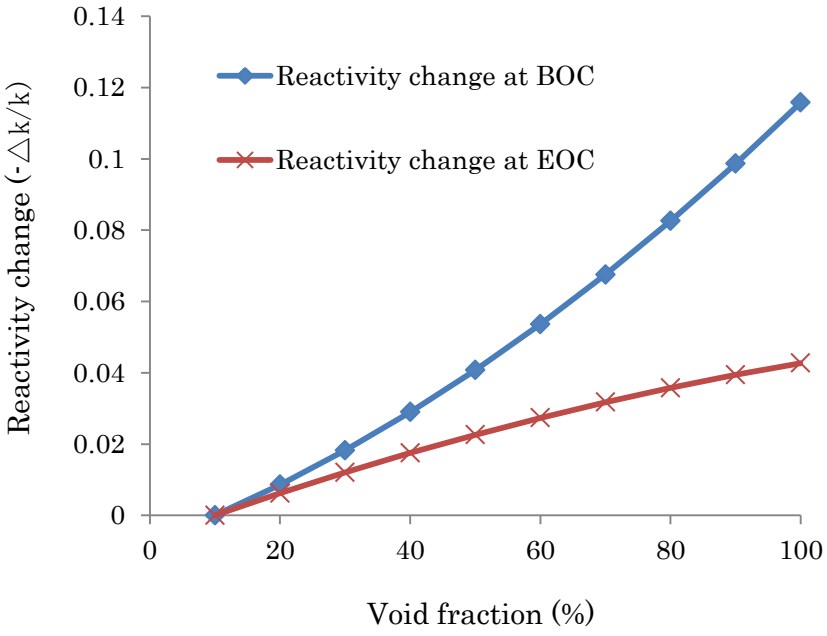


Fig. 3.5. Reactivity change with the void fraction in the coolant at BOC and EOC of equilibrium cycle

3.4.4. Radiotoxicity at discharge

The actinides inventories of the input and discharged reprocessed fuel due to decay were calculated using MONTEBURNS code. The radiotoxicity of those actinides was derived from the isotopes inventories and the dose coefficients given for intake by ingestion in adults [7]. The results for radiotoxicity are normalized per metric ton of HM input for ADS. Comparison of radiotoxicity of TRUs at input and at discharge is shown in Fig. 3.6. The TRU waste is efficiently transmuted using the seed and blanket ADS, thus 20% of the dose is reduced and mainly due to the reduction of ^{239}Pu and Am. Most of the dose is come from Pu and the radiotoxicity starts to drop after 20,000 years (half-life of ^{239}Pu). The production of curium isotopes enlarges the toxicity slightly however, it do not have large impact on the toxicity of the spent fuel at discharge.

Table 3.4. Dose coefficients for important nuclides in spent nuclear fuel given for intake by ingestion and for inhalation in adult humans [7]

Nuclide	Dose coefficient(nSv/Bq)
^{237}Np	110
^{238}Pu	230
^{239}Pu	250
^{240}Pu	250
^{241}Pu	4.8
^{242}Pu	240
^{241}Am	200
^{242m}Am	190
^{243}Am	200
^{242}Cm	12
^{243}Cm	150
^{244}Cm	120
^{245}Cm	210

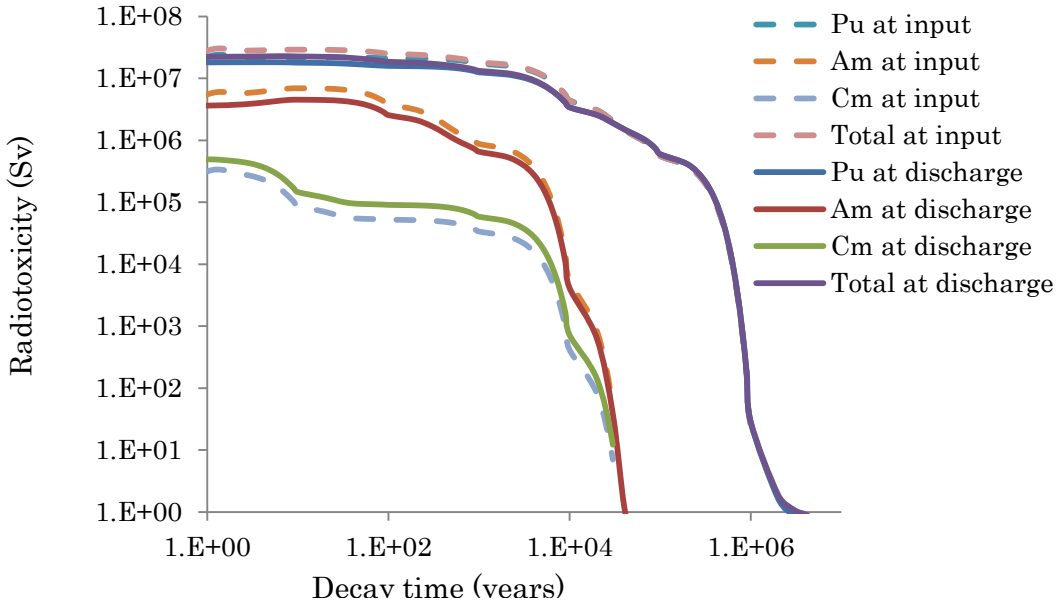


Fig. 3.6. Evolution of ingestion radiotoxicity for reprocessed fuel of seed and blanket ADS at input and discharge

3.4.5. Influence of the cross section uncertainties on the transmutation rate and radiotoxicity at discharge.

As discussed above, the reduction of the radiotoxicity at discharge is mainly due to the reduction of Pu and Am. However, regarding to the cross section uncertainties for those isotopes, it is essential to evaluate the contribution of the uncertainty of Pu and Am cross section to the transmutation rate and the radiotoxicity at discharge. For that purpose, additional calculations of sensitivity coefficient of reaction rate of ^{239}Pu, ^{241}Am, and ^{243}Am for the multi-cycle ADS core at equilibrium state were conducted by using SAGEP code [8] with 107-group cross section data attained by SRAC2006-CITATION [9]. SAGEP is the code which calculates sensitivity coefficients in a two dimensional system on the basis of generalized perturbation theory. The sensitivity calculation scheme is presented in the Fig.3.7. The sensitivity coefficient of reaction rate S_{RR} gives the relative change in the reaction rate of ^{239}Pu, ^{241}Am, and ^{243}Am due to the relative change of cross section data of those isotopes. Figure 3.8 illustrates the sensitivity coefficients of reaction rate for ^{239}Pu, ^{241}Am and ^{243}Am of ENDF/B-VII cross section library. As we can see from the figure, sensitivity coefficient of those isotopes at the fast energy range has the main contribution to the total sensitivity coefficient.

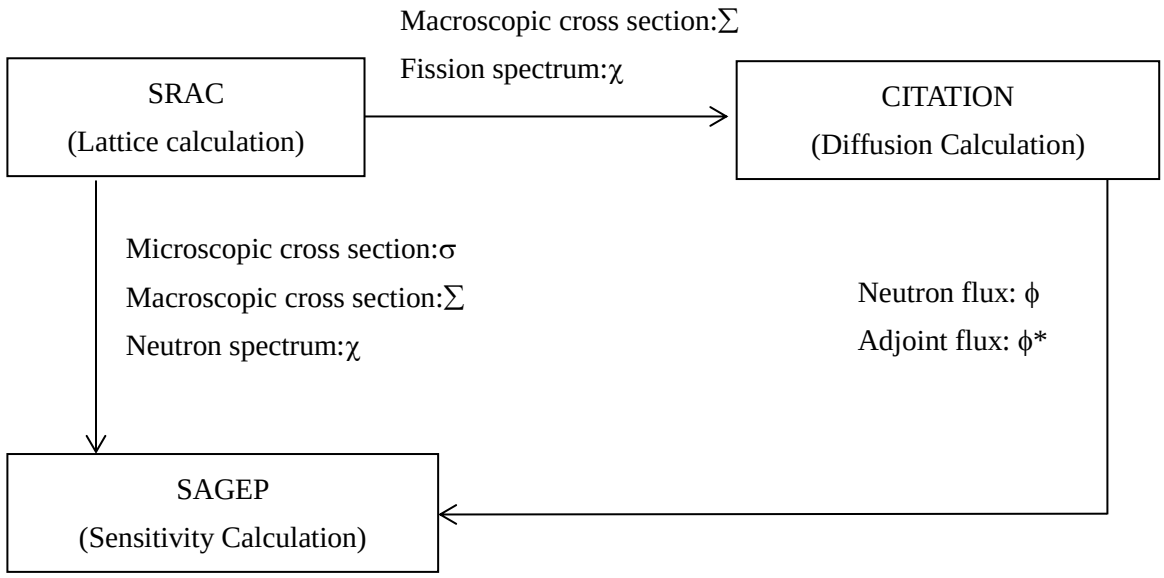


Fig. 3.7. Sensitivity calculation scheme

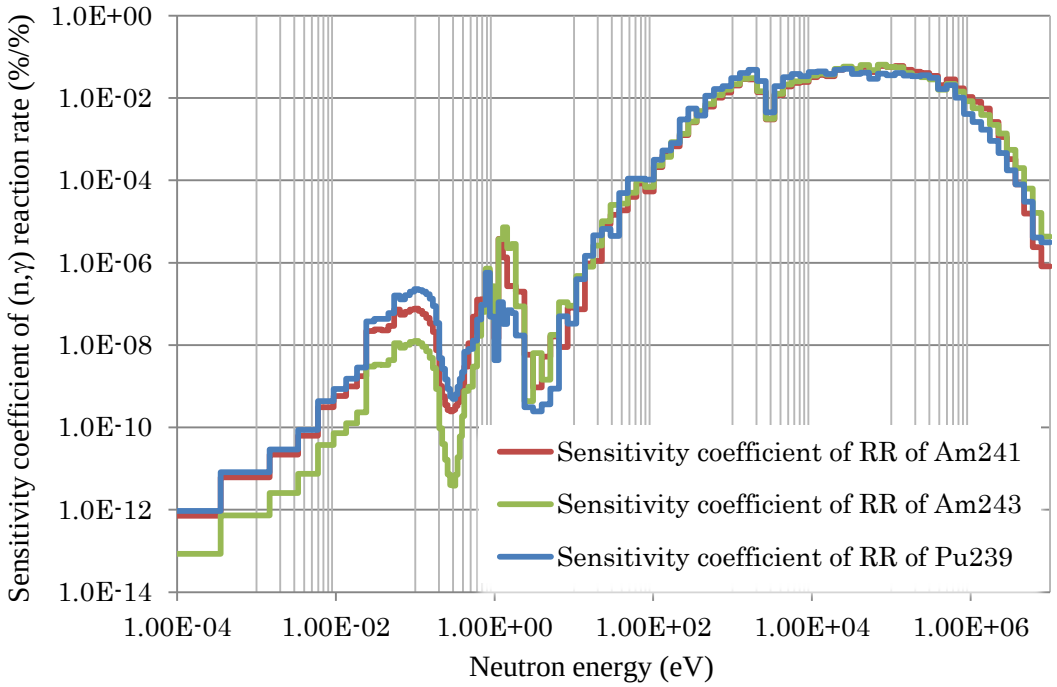


Fig. 3.8. Sensitivity coefficient of reaction rate for ^{239}Pu, ^{241}Am and ^{243}Am capture cross section

The covariance data was generated in a 107-group energy structure to map them into the same group structure as the sensitivity coefficients [10]. The cross section covariance data of ^{239}Pu, ^{241}Am and ^{243}Am were derived from the ENDF/B-VII evaluated nuclear data file using ERRORR module for NJOY99 code [11]. Figure 3.9 and 3.10 shows the covariance matrix of capture cross section of ^{239}Pu , ^{241}Am and ^{243}Am.

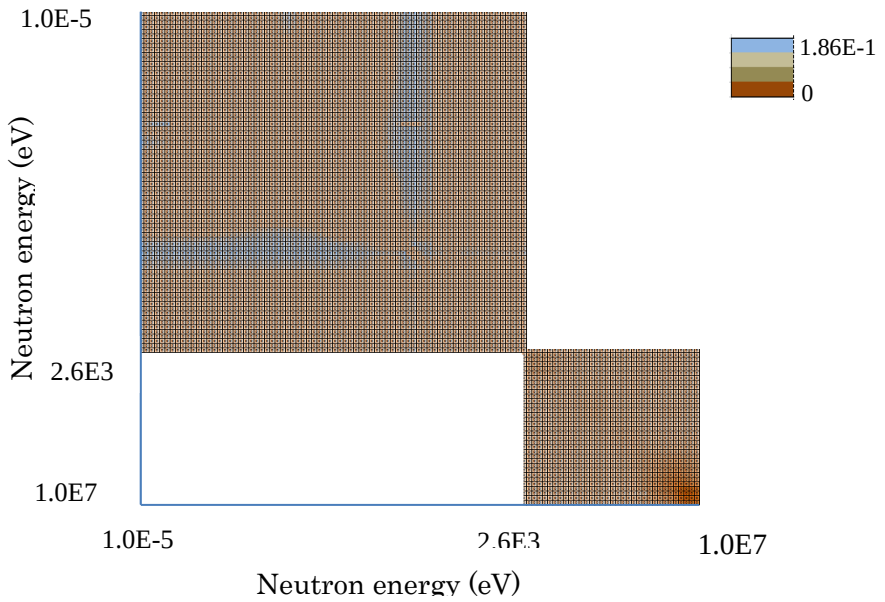


Fig. 3.9. Covariance matrix of (n,γ) reaction for ^{239}Pu capture cross section

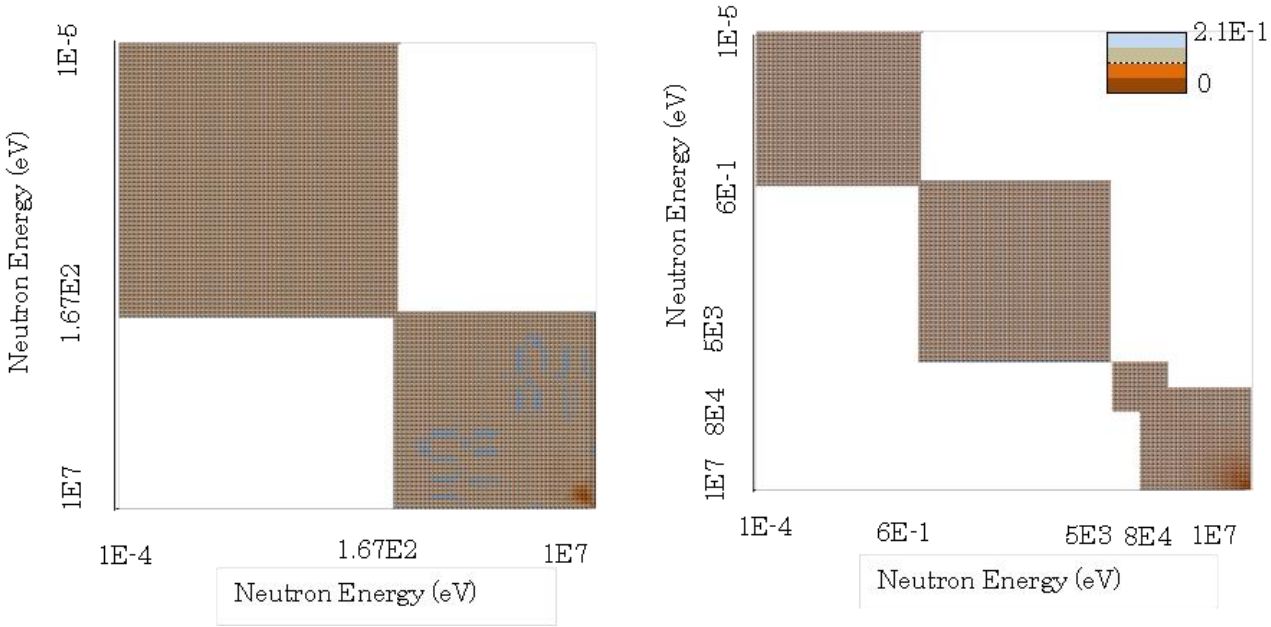


Fig. 3.10. Covariance matrix of (n,γ) reaction for ^{241}Am and ^{243}Am capture cross section

The uncertainty (δE_{rr}) is derived as in Eq. 3.1 as follows

$$\delta E_{rr} = \sqrt{S_{RR}^{T} \cdot V \cdot S_{RR}}\ , \tag{3.1}$$

where S_{RR} is sensitivity coefficient of reaction rate in a 107-group energy structure obtained by SAGEP code and the superscript T means transposition,

$$S_{RR} = \begin{bmatrix} S_{RR1} \\ S_{RR2} \\ \vdots \\ S_{RR107} \end{bmatrix},$$

and V is the covariance matrix for (n,γ) reaction of each isotope in a 107-group energy structure obtained by NJOY99 code:

$$V = \begin{bmatrix} V_{1,1} & V_{1,2} & \cdots & V_{1,107} \\ V_{2,1} & V_{2,2} & \cdots & V_{2,107} \\ \vdots & \vdots & \ddots & \vdots \\ V_{107,1} & V_{107,2} & \cdots & V_{107,107} \end{bmatrix}.$$

As a result, the reaction rate calculation results have uncertainty about 5.9% for ^{239}Pu. For ^{241}Am and ^{243}Am, the reaction rate calculation results have uncertainty about 2.3% and 7.1%. Taking into account the weight ratio of those isotopes in the total radiotoxicity reduction at discharge, the uncertainty caused by the isotopes cross section variance is about 6%. That means, the radiotoxicity reduction of the seed and blanket ADS is about (20±1)%.

3.5. Conclusions

The multi-cycle seed and blanket ADS core for higher thorium utilization and TRU waste transmutation is investigated. Instead of wasting thorium fuel and its breeding ^{233}U, thorium fuel assemblies are remained inside the core and utilized to produce energy by employing multi-cycle approach. At the

equilibrium state, the thorium fuel contribution to total core power is improved (35.7%) compared to the startup cycle. It also has benefit in reducing the axial power peaking at BOC from 2.50 at startup cycle to 1.42 at equilibrium state. Compared to the startup cycle, the TRU transmutation rate of the equilibrium cycle reduces about 15 kg per ton of spent HM. However, the once burnt reprocessed fuel assemblies are shuffled and transmute more for a second time, thus, the TRUs are transmuted into a deeper level. At discharge, the reduction of radiotoxicity from the TRU waste is about 20% with 1% uncertainty, mainly due to the reduction of Pu and Am.

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Chapter 4: **Transmutation strategy using seed and blanket ADS for future reactors in Vietnam**

4.1. Introduction

Demand for power is rising as Vietnam's economy expands. Electricity demand growth has been 14% per annum and is expected to be 15% per annum to 2015, then slowing to 2020, though other figures suggest 10% per annum. In order to improve the energy security, the government has approved the nuclear power development plan in Vietnam. The planed and proposed nuclear power reactors to 2030 are shown in the Table 4.1 [1]. Due to the plan, 2000 MWe nuclear power plant at Phuoc Dinh in the Ninh Thuan province should be on line by 2023. A further 2000 MWe was planned at Vinh Hai nearby, followed by a further 6000 MWe by 2030. A high demand scenario would give 8000 MWe in 2025 and 15,000 MWe (10% of total) in 2030 at up to eight sites in five provinces. As demonstrated in detail later, from 2025 with 8000 MWe, about 170 tons of spent fuels (of which 3 tons are TRUs and long life fission products (LLFFs) are produced every year. It will extend to 120 tons of TRUs and LLFFs after 40 years of operation. The problem of radioactive waste from nuclear power plants causes a big issue for the countries holding nuclear installation. Some isotopes with long half-lives will remain radioactive hundreds of thousands of years in the future. Therefore, in order to minimize the exposing radiotoxic material to the environmental, the material should be transmuted to convert some of the more harmful isotopes to less harmful products and the rest should be placed safely in a geological repository. With the waste transmutation potential and capability of producing energy from utilizing thorium fuel, the seed and blanket ADS can be a good candidate for next few - decade technology in Vietnam. In this chapter, the application of seed and blanket ADS in the nuclear waste transmutation strategy for future reactors in Vietnam is considered.

Table 4.1 Planned and Proposed Nuclear Power Reactors to 2030 [1]

Location	Plant (province)	Type	MWe nominal	Start construction	Operation
Phuoc Dinh	Ninh Thuan 1-1	VVER-1000/428	1060	2017 or 2018	2023
	Ninh Thuan 1-2	VVER-1000/428	1060	2018 or 2019	2024
	Ninh Thuan 1-3	VVER-1000	1000		(N/A)
	Ninh Thuan 1-4	VVER-1000	1000		(N/A)
Vinh Hai	Ninh Thuan 2-1	Japanese Gen III or Atmea1	850-1150	Dec 2015, delayed	(N/A)
	Ninh Thuan 2-2	Japanese Gen III or Atmea1	850-1150	2016, delayed	(N/A)
	Ninh Thuan 2-3	Japanese Gen III or Atmea1	850-1150		(N/A)
	Ninh Thuan 2-4	Japanese Gen III or Atmea1	850-1150		(N/A)
Central		(N/A)	1350		2028
Central		(N/A)	1350		2029
Total planned (4)			4000		
Total proposed by 2030			6700		

4.2. TRU elimination strategy for future reactors in Vietnam

4.2.1. Utilization of seed and blanket ADS for TRU elimination for future reactors in Vietnam.

The development plan of nuclear power expresses the quick expansion of nuclear power in Vietnam in the near future. By 2029, eight 1000 MWe – class reactors are planned to come on line. The large amount of nuclear waste from those nuclear power plants will bring serious radiotoxic and proliferation risks. Massive research and development (R&D) works on how to deal with these hazardous materials are needed to be done and should start from now on.

Regarding the radiotoxicity and decay heat, HLWs which includes TRUs and LLFFs are the most

hazardous materials. TRUs and LLFFs respectively occupy 1.1% and 0.6% of the conventional spent nuclear fuel from a LWR. The element masses in the waste for UOX with an initial enrichment of 4.2%, an average thermal burnup of ~50 GWd/t of heavy metal and after 5 years of cooling time is demonstrated in Table 4 .1[2]. The amount of waste for 8 GWe by 2027 is derived as illustrated in the figure. More than 3 tons of TRUs and LLFFs are produced every year. It will extend to 120 tons of TRUs and LLFFs of which 10 tons are MA after 40 years of average lifetime for a conventional nuclear reactor. TRUs are responsible for most of long-term radiotoxicity because of their long half-lives combined with complicated decay chain.

Table 4.2 Composition of nuclear waste after 50 GWd/t of heavy metal burnup [2]

Actinides	**Actinides (kg/GWe/yr) at 50 GWd/tHM**	**Element (kg/GWe/yr) at 50 GWd/tHM**	**Half-life (yr)**	**Element (kg/8 GWe/yr) in 2025**
^{234}U	5.2	20765	2.5×10^{5}	166120
^{235}U	173.0		7.0×10^{8}	
^{236}U	136.0		2.3×10^{7}	
^{238}U	20424.0		4.5×10^{9}	
^{237}Np	160	16.0	2.1×10^{6}	128
^{238}Pu	8.6	255.2	87.7	2041.6
^{239}Pu	125.3		2.4×10^{4}	
^{240}Pu	71.3		6.6×10^{3}	
^{241}Pu	30.3		14.3	
^{242}Pu	19.8		3.7×10^{5}	
^{241}Am	11.6	16.4	432.7	131.2
^{243}Am	4.8		7.4×10^{3}	
^{243}Cm	0.01	1.7	29.1	13.6
^{244}Cm	1.5		18.1	
^{245}Cm	0.1		8.5×10^{3}	
^{246}Cm	0.03		4.7×10^{3}	
LLFFs		127.2		1017.6

By taking the advantages as discussed in previous chapters, the seed and blanket ADS using thorium – reprocessed fuel is considered as a safe, clean and economical solution of waste management problem. The production of TRUs per $TW_{th}h$ at 40GWd/tHM burnup of a PWR [2] and the seed and blanket ADS is presented in Table 4.3. It shows the large efficiency in eliminating TRUs of the seed and blanket ADS. These numbers imply that each 1 GW_{th} ADS can be able to transmute the TRUs from 2 GW_{th} LWRs. By that it means, in case of Vietnam, if we want to eliminate the TRUs from the nuclear power plants corresponding to 8000 MWe, 14 units of ADS with 840 MW_{th} are required to be installed. If the thermal efficiency of the ADS is assumed to be equal to 40%, more than 4 GWe from those ADS will be added to the grid.

Table 4.3 Comparison of the actinide balance per $TW_{th}h$ for PWR and the seed and blanket ADS

Actinides ($kg/TW_{th}h$)	**PWR (UOX) 41 GWd/tHM**	**Seed and blanket ADS 40 GWd/ton of spent fuel**
Pu	+11.3	-22.6
Np	+1.18	-11.6
Am	+1.04	-10.7
Cm	+0.03	+3.9

The proposed fuel cycle with the ADS utilizing thorium and reprocessed fuel is illustrated in Fig. 4.1 Pu and MA from spent fuel after storage for 7 years to reduce the decay heat and short life radiotoxic isotopes is reprocessed and fabricated for ADS. After being burnt, the spent fuel of ADS can be recovered and put back to the system for further transmutation. However, regarding to the radiotoxicity, not only TRU waste but also LLFFs in the spent fuel of LWRs should be transmuted into stable or short-lived nuclides in order to eliminate the permanent geological repository. The LLFFs could be separated and transmuted in a dedicated facility. One of the options is that it can be loaded in the core reflector of ADS where the thermal neutron spectrum is utilized to transmute those nuclides.This approach is now under estimation and will not be discussed in the scope of this thesis.

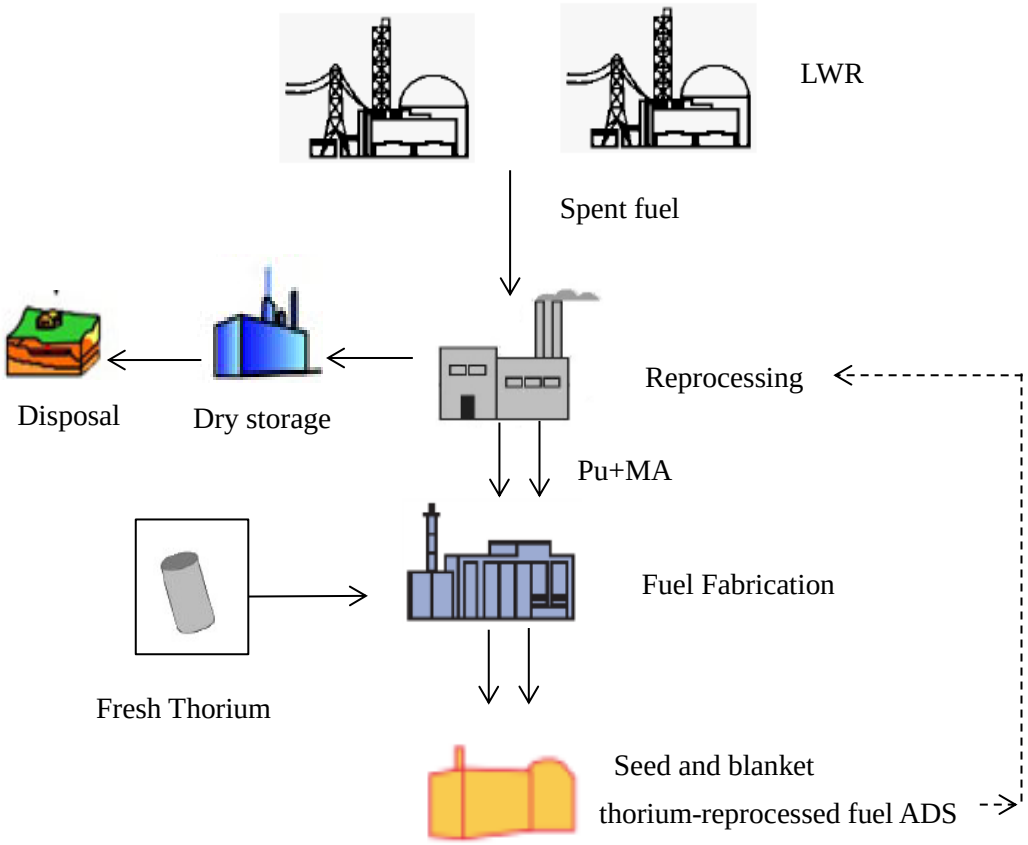


Fig. 4.1. Fuel cycle with the ADS utilizing thorium and reprocessed fuel

4.2.2. Potential benefits and challenges for installation of seed and blanket ADS

The seed and blanket ADS with great potential of TRUs transmutation and its advanced safety features can be the solution for the nuclear waste transmutation and energy security problem in Vietnam in the next few decades. Even though it is a new technology, its simplification in the safety feature of the system will not be an obstacle for the installation and operation technically.

Regarding to the thorium availability, with the estimated thorium reserve about 10,000 tons, Vietnam can consider to utilize thorium in the seed and blanket ADS to produce energy instead of totally rely on the imported nuclear fuel.

However, in order to realize the advanced benefit of the seed and blanket ADS, there are many

challenges related to the ADS technical issues and thorium fuel cycle. The most serious and unattractive disadvantage of ADS would be to couple an expensive device of high power proton accelerator on the nuclear power reactor. A rough estimate of cost of 30 MWt beam power proton linac stands same as the order of cost of reactor plant [3].

Thorium is much more abundance than uranium in nature. However, the knowledge of thorium fuel behavior inside reactor and its nuclear data is very limited compare to uranium fuel. Therefore, much more validation and verification on thorium data and thorium utilized ADS need to be done in order to provide more reliable data before apply the design in the industrial scale.

Regarding the thorium fuel fabrication, because of higher melting point of ThO_2, much higher sintering temperature is required to produce high density ThO_2 fuel. For thorium fuel back end issues, even though the seed and blanket ADS using separate thorium fuel assemblies and reprocessed fuel assemblies, but the thorium-uranium extraction (THOREX) only has been developed on laboratory or pilot plant scale [4]. It may take some more time to be applied in full scale. Moreover, for the LWR spent fuel reprocessing, because of the proliferation related issue, the reprocessing included in this cycle may have to face to strict political discussions to be approved for the peace used purpose of recovered spent fuel.

4.3. Conclusions

Although great amount of validation and verification are needed to be done to finalize the design of the seed and blanket ADS and much more effort is needed to utilize the seed and blanket ADS, this study offers a potential solution to solve the TRU waste management in Vietnam. With the TRU transmutation capability of the seed and blanket ADS, 14 units of ADS would be required to deal with the TRUs from the future reactors to be constructed in Vietnam and add about 4 GWe (40% thermal efficiency) to the grid. The seed and blanket ADS is not only the solution for the waste management problem in Vietnam but also improve the energy security in Vietnam by utilizing the thorium resource.

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Chapter 5: **Accuracy of thorium cross section and its impact on the thorium utilized ADS reactivity calculation**

5.1 Introduction

As mentioned above, beside the advantage of thorium's abundance in nature, thorium fuel cycle with the feature that the TRUs production during an operating cycle is drastically reduced compared with the use of uranium fuel cycle is favorable for the TRUs eliminating system such as the seed and blanket ADS [1]. However, because of limitation of experimental data, the accuracy of the cross section library as well as the calculation's result might not be evaluated properly. The aim of this chapter is to verify the accuracy of thorium cross section using the experimental and calculated data. In order to achieve this goal, a series of critical experiments with various neutron spectra at Kyoto University Critical Assembly (KUCA) facility was implemented. In the fuel rod, thorium and enriched uranium plates were loaded in combination with polyethylene moderator to configure two critical cores with different neutron spectra ($H/^{235}U$=140 and 210). The criticality experimental results are compared with the calculated ones for verification purpose. Control rod worth and reactivity worth measurements of thorium fuel plates were also performed in these experiments to confirm the safety of the operation and the reliability of the verification. Sensitivity coefficients of thorium sample worth are also analyzed to evaluate impact of the inaccuracy of ^{232}Th cross section on reactivity worth. The influence of the ^{232}Th cross section on the seed and blanket ADS using thorium-reprocessed fuel will be discussed in this chapter.

5.2 Thorium cross section verification methodology

5.2.1. Description of the KUCA core configurations

The critical cores in this paper are consisted of pure thorium plates, 93% enriched uranium plates and solid polyethylene moderator and reflector. Two core configurations with the same ^{232}Th to ^{235}U atom ratio (15.2) but different H to ^{235}U atom ratios (140, 210) were adopted as follows:

First core (with H/^{235}U=140 and ^{232}Th/^{235}U=15.2) is referred as B7/8”P12ETEETEE core. Fuel unit employed in this core is composed of two 1/8 inch thick plates of pure Th, five 1/16 inch thick plates of enriched U and seven 1/8 inch thick plates of polyethylene. It is repeated twelve times and placed into fuel rod within reflectors as shown in Fig. 5.1a.

Second core (with H/^{235}U=210 and ^{232}Th/^{235}U=15.2) is referred as B10/8”P9ETEETEE core. Fuel portion here is composed of nine times of fuel unit which consists of two 1/8 inch thick plates of pure Th, five 1/16 inch thick plates of enriched U and ten 1/8 inch thick plates of polyethylene (Fig. 5.1b).

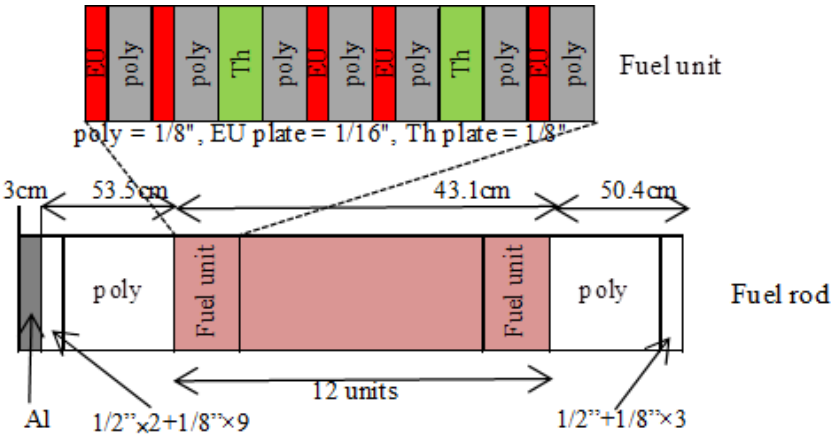


Fig. 5.1a. The B7/8”P12ETEETEE fuel unit and fuel rod

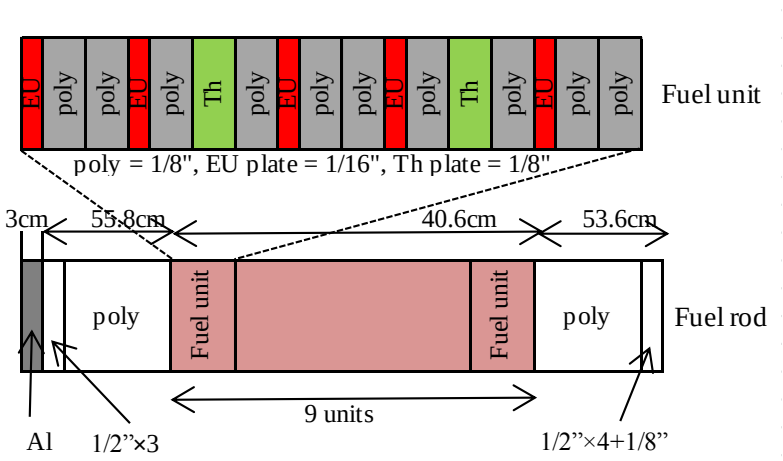


Fig. 5.11b. The B10/8"P9ETEETEE fuel unit and fuel rod

5.2.2. Experiments

Characteristics of two critical cores are investigated in this study. Due to critical experiment process, the core configurations were started with 29 fuel rods and additional fuel rods were loaded to the initial core step by step as shown in Table 5.1 until criticality was achieved.

Table 5.1. Experiment cases of B7/8"12ETEETEE and B10/8"P9ETEETEE cores

Step i	Number of fuel rods inserted in B7/8"P12ETEETEE core	Number of fuel rods inserted in B10/8"P9ETEETEE core
0	29	29
1	36	36
2	39	39
3	43	43
4	46	44
5	48	45
6	49	46
7	50	47
8	51	

Critical state was established with 51 fuel rods inserted inside B7/8”P12ETEETEE core and 47 fuel rods inside B10/8”P9ETEETEE core (Fig. 5.2a and 5.2b). Their neutron spectra are illustrated in Fig. 5.3. Neutron multiplication of all subcritical states corresponding to each step was also recorded to study impact of fuel rod insertion in order to achieve the criticality. Count rate measurements were carried out at fission chamber number 1, 2 and 3 and detector BF_3 number 1 and 2 as arranged in Fig. 5.2a and 5.2b.

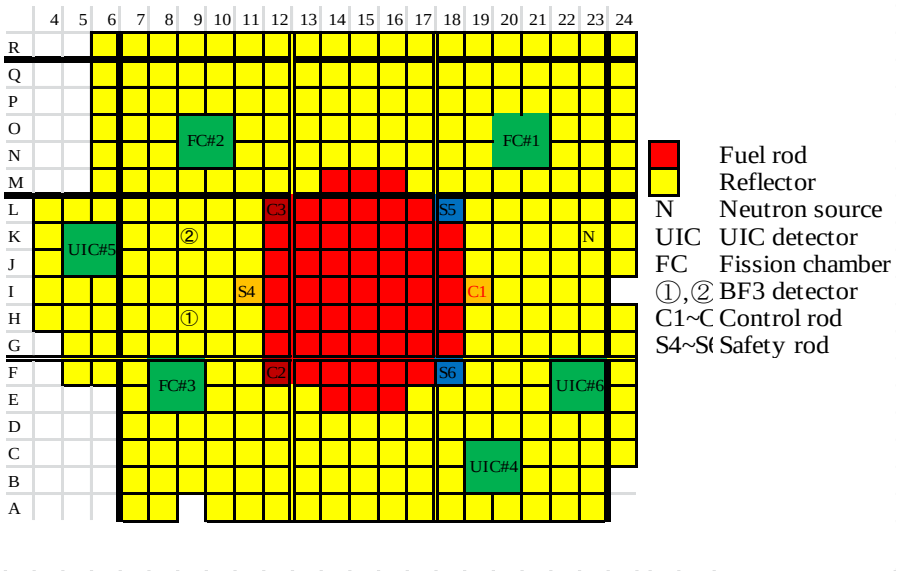


Fig. 5.2a. Top view of B7/8”P12ETEETEE critical core configuration

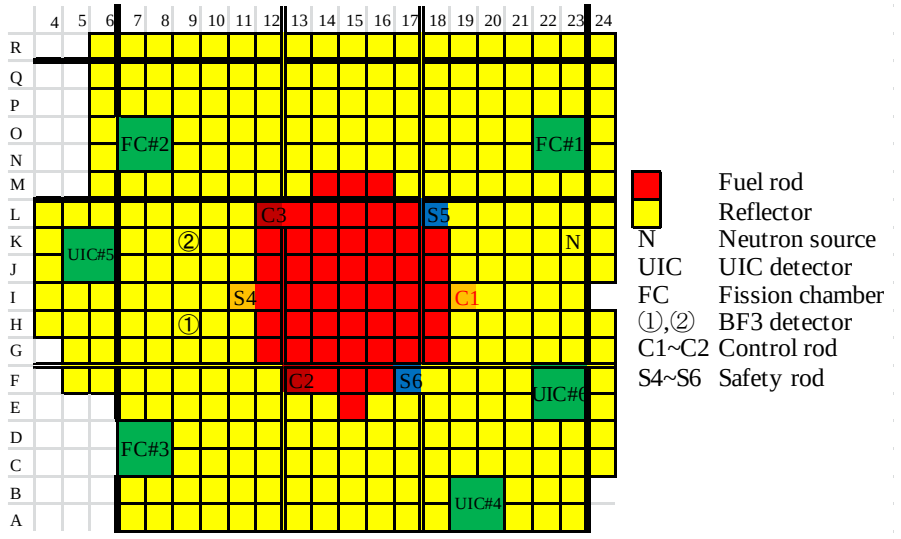


Fig. 5.2b. Top view of B10/8”P9ETEETEE critical core configuration

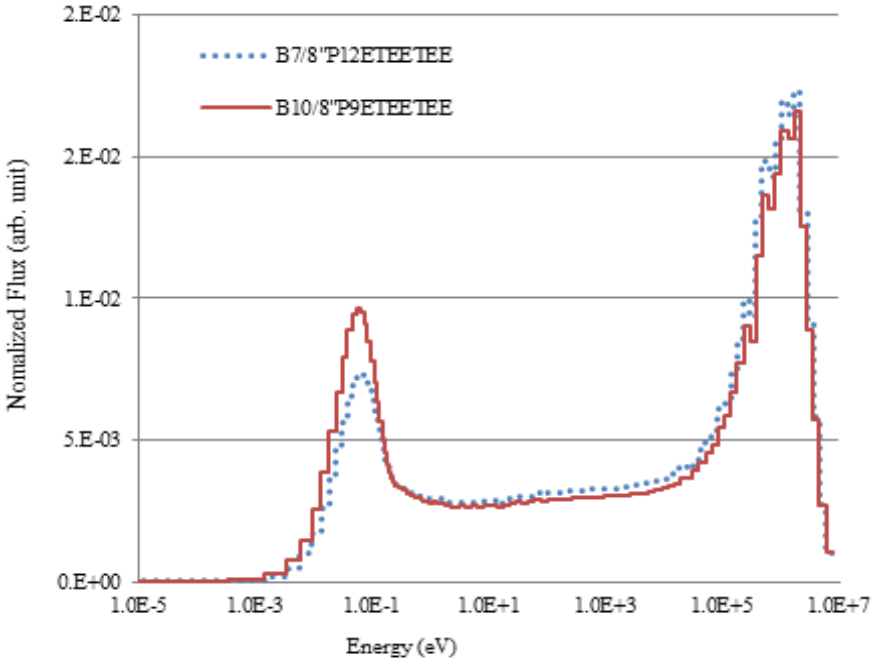


Fig. 5.3. Neutron spectra of B7/8”P12ETEETEE and B10/8”P9ETEETEE cores

Core excess reactivity and reactivity worth for each control rod of B7/8”P12ETEETEE and B10/8”P9ETEETEE cores were measured using positive period and drop methods. Period method is a method to measure a positive reactivity inserted into the core at critical state through the positive reactor period. Drop method is adopted to measure the negative reactivity of a control rod by suddenly inserting it into the core after adjusting criticality [2]. Since position of control rods and safety rods is symmetrical, only worth measurement of control rod C1, C2 and C3 was performed.

In the experiment, reactivity worth of a Th plate was also measured. The sample worth measurements were implemented by replacing Th plates by Al plates. The position of the replacement was chosen around the core center in vertical and horizontal directions. Additionally, to adjust the excess reactivity of the cores, partial fuel rods (P) were loaded at 14E and 16E positions in B7/8”P12ETEETEE core and at 15I position in B10/8”P9ETEETEE core. Because of safety regulatory that the maximum excess reactivity in KUCA core is 0.35%Δk/k, the number of thorium plates replaced by aluminum plates could only be up to 4.

5.2.3. Calculations

To perform eigenvalue calculations for 2 core configurations, MVP code is employed. MVP is a continuous energy Monte Carlo code which is developed by Japan Atomic Energy Research Institute using event-based algorithm to enhance the performance of vector and parallel supercomputers in order to improve the calculation speed [3]. The MVP calculations were executed with JENDL-4.0 cross section library [4] by 100 million histories and the statistical errors in eigenvalue were less than 0.01%. Control rod worth evaluation was using the same cross section library and history number. In cases of sample worth calculations, zero, two Al (2Al), and four Al (4Al) plate replacements were calculated and compared with the experimental data.

5.3 Comparison of experimental and calculated results and the impact of inaccuracy of ^{232}Th cross section library on the calculated results

5.3.1. Criticality calculation

In experiments, neutron flux is measured on the basis of detector response such as the count rate of the neutron detectors placed in fission chambers. Detector count rates are assumed to be proportional to neutron flux and its relation with the k_{eff} can be expressed as follows:

$$A_0 / A_i = \phi_{T0} / \phi_{Ti} = 1 - k_{eff,i}. \quad (5.1)$$

We also have:

$$1/M = 1 - k_{eff,i}, \quad (5.2)$$

where A_0 is detector count rate for initial core configuration, A_i is detector count rate for the ith fuel loading core configuration, ϕ_{T0} is neutron flux for initial core configuration, ϕ_{Ti} is neutron flux for the ith fuel loading core configuration, $k_{eff,i}$ is effective multiplication factor for the ith fuel loading core configuration and M is

neutron multiplication.

Therefore, A_0/A_i which is assigned as the inverse count rate is employed to predict the critical mass or the number of fuel rods to make the core critical [2].

The critical state was established at B7/8"P12ETEETEE core with 51 fuel rods (F51) and partial insertion of control rod C1. Criticality benchmark calculations were performed with the MCNP code [5] and its continuous-energy ENDFB/VII.0 [6] and it shows a good agreement with MVP results. The comparison of inverse count rate corresponding to every criticality approaching step of B7/8"P12ETEETEE core derived from MVP calculated and measured data by detectors is illustrated in Fig. 5.4.

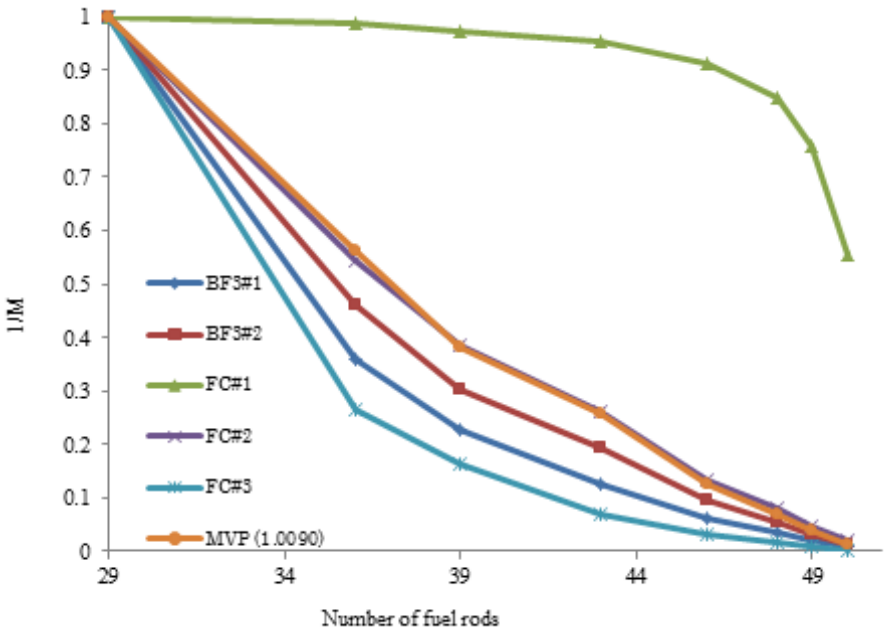


Fig. 5.4. Comparison of the inverse count rates of B7/8"P12ETEETEE core obtained by detectors and MVP calculated result

As we notice from Fig. 5.2a, compared with other detectors, FC#2 fission chamber has been judiciously placed (not quite close to neutron source like FC#1, not quite far from active region when there is only a small number of inserted fuel rods inside the core like FC#3 and BF_3#2, and having a big enough angle

facing to active region, larger than that of BF_3#1 and moreover, not affected by criticality approaching process with increasing number of fuel rods) to collect the most stable and reliable data. As illustrated in Fig. 5.4, the inverse count rate, 1/M, acquired from FC#2 is the best match and agrees well with the calculated result.

Similarly, B10/8"P9ETEETEE core achieved criticality with 47 fuel rods in core and partial insertion of control rod C1. The inverse count rate obtained from FC#2 is in good agreement with the calculated one.

With F51 core configuration, B7/8"P12ETEETEE core has the measured excess reactivity of (1.47±0.04)%∆k/k (the uncertainty of excess reactivity is estimated by considering both experimental and parameter errors in the evaluation process). Computed k_{eff} value executed by MVP is 1.01046±0.008%. Critical value calculated by MVP is derived from that excess reactivity and k_{eff} by using Eq. (5.3) and (5.4) as follows:

$$\rho_{ex} = \frac{1}{k_{cri}} - \frac{1}{k_{eff}}, \tag{5.3}$$

$$k_{cri} = \frac{k_{eff}}{1 + \rho \times k_{eff}}, \tag{5.4}$$

where ρ_{ex} is excess reactivity, k_{eff} is effective multiplication factor for F51 calculated by MVP and k_{cri} is MVP multiplication factor to acquire criticality in core.

k_{cri} is equal to 1.0090±0.0001. That means by using JENDL-4.0 cross section library, the MVP evaluation has overestimated effective multiplication factor for B7/8"P12ETEETEE core about (0.90±0.01)%. For B10/8"P9ETEETEE core configuration which has softer neutron spectrum (Fig. 5.3), the calculation overestimates effective multiplication factor for critical core about (0.99±0.01)%. Analyses which had been done for critical cores with enriched uranium only has found the largest overestimation among different neutron spectra of calculated results compared to experimental data about (0.21±0.05)% (H. Unesaki, personal contact). Thus, limitation of ^{232}Th cross section library is concluded to contribute about (0.78±0.06)% to the overestimation of the calculation in thorium core.

5.3.2. Control rod

Experimental reactivity worth of control rods of B7/8”P12ETEETEE and B10/8”P9ETEETEE cores obtained by drop method is shown in Table 5.2.

Table 5.2. Measured reactivity worth of control rods

Control rod	**ΔρExp. (%) B7/8”P12ETEETEE core**	**ΔρExp. (%) B10/8”P9ETEETEE core**
C1	0.306±0.0005	0.2720±0.0001
C2	0.323±0.0009	0.3650±0.0005
C3	0.321±0.0040	0.3250±0.0010

Not only the total rod worth but also the control rod worth as a function of rod position is very important for safe operation of a reactor and for carrying the further reactivity measurement experiments [2] such as reactivity worth measurement. Once the control rod calibration curve is built, it will be used to estimate the reactivity change corresponding to different adjusted control rod positions. Therefore, reactivity worth of the control rod was carefully measured and calibrated in the experiment. Hereafter, the reactivity worth calibration of control rod C1 of B7/8”P12ETEETEE core will be explained in detail. The experimental data used to calibrate in this part was acquired by period method. Inserted reactivity as a function of rod position, which is designated as “control rod calibration curve”, was obtained from the following formula:

$$\rho = \rho_H \left\{ \frac{x - m_3}{m_2} - \frac{1}{2\pi} \sin(2\pi \times \frac{x - m_3}{m_2}) \right\}, \tag{5.5}$$

where ρ is inserted reactivity, x is control rod position, ρ_H is C1 total rod worth, m_2 is 587.84mm and m_3 is 367.02mm. C1 total rod worth of B7/8”P12ETEETEE core, ρ_H, was attained by using drop method (Table 5.2), while active core length m_2 is the total length of the fuel portion adding the extrapolation distance at its both ends and m_3 is the bottom position of the active core which is obtained from the least square fitting of the

measured results to Eq. (5.5).

As shown in Fig. 5.5, the experimental data of reactivity worth of control rod C1 (CR worth 1 and CR worth 2), reactivity worth of the core with partial fuel rod (P), 2 Al plate replaced central rod (2Al), and partial fuel rod and 4 Al plate replaced central rod (P+4Al) are used for the calibration. As we notice, the measured data of CR worth 2 at rod position 667.74 mm is slightly off the curve and the reason for that has not yet been found. However, another experimental data was collected at the same control rod position to confirm the reliability of the fitted curve. Since the control rod worth of control rod C1 is calibrated, the calibration curve can be used to deduce the reactivity change which is the difference between the C1 total reactivity worth (in case of B7/8"P12ETEETEE core is 0.00306) and the reactivity corresponding to the adjusted rod position after introducing the sample into the core.

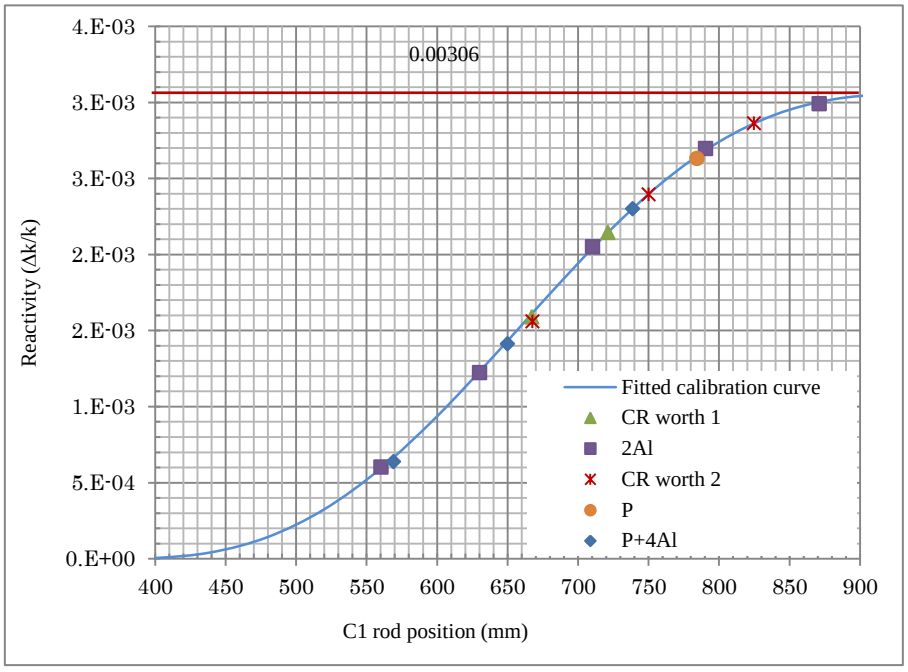


Fig. 5.5. Comparison of the control rod worth between the calibration curve and experimental data of B7/8"P12ETEETEE core

Reactivity worth of control rod C3 of B10/8”P9ETEETEE core was calibrated in the same manner and its calibration curve is also employed to obtain the reactivity change in this experiment.

5.3.3. Sample worth

Sample worth experimental data was obtained by substituting Al plates to Th plates and measuring the change of reactivity because Al is considered as transparent to neutrons. The reactivity change was experimentally deduced by using the control rod calibration curve for the cores with partial fuel rod P, P+2Al, and P+4Al. The reactivity difference between the core with partial length P and the core with P+2Al, the core with P+2Al and the one with P+4Al which equals to reactivity worth of two Th plates was measured several times to reduce the deviation and its average is shown in Table 5.3. Reactivity worth of a single Th plate is determined as a half that value. The measured and calculated reactivity worth of a Th plate for two core configurations is compared in Table 5.3. For B7/8”P12ETEETEE core, reactivity worth of one Th plate has discrepancy between the experimental and calculated results about 3.1% and “-” indicates the underestimation of Th worth of the calculation. For B10/8”P9ETEETEE core, which has softer neutron spectrum, the larger discrepancy is observed (14.2%). This tendency suggests that the ^{232}Th capture cross section is underestimated especially at the low energy range.

Table 5. 3. Comparison of experimental and calculated sample worth

Cases of measurement	**B7/8”P12ETEETEE core**			**B10/8”P9ETEETEE core**		
	Δρ Exp. (%)	*Δρ Cal. (%)*	*RD**	*Δρ Exp. (%)*	*Δρ Cal. (%)*	*RD**
P→P+2Al	0.0996±0.0005	0.099±0.012	-0.6%	0.1250±0.0010	0.122±0.012	-2.2%
P+2Al→P+4Al	0.1010±0.0012	0.095±0.012	-5.9%	0.1290±0.0006	0.105±0.012	-22.9%
Th	0.0501±0.0003	0.049±0.004	-3.1%	0.0650±0.0003	0.057±0.004	-14.2%

$$*: RD = \left(\frac{\Delta\rho Cal - \Delta\rho Exp}{\Delta\rho Cal}\right)$$

5.3.4. Impact of the inaccuracy of ^{232}Th cross section on calculated results

As discussed above, the inaccuracy of ^{232}Th cross section library is mainly responsible for the discrepancies between calculated and experimental data. Thus, it is essential to evaluate the contribution of the uncertainty of ^{232}Th cross section to the discrepancies depended on different energy ranges. For that purpose, additional calculations of sensitivity coefficient of thorium reactivity worth were conducted by using SAGEP code [7] with cross section data attained by SRAC2006-CITATION [8]. The sensitivity coefficient of reactivity worth evaluated as described in the following equation:

$$S_W = \frac{dR/R}{d\sigma/\sigma} = \frac{S_k/k - S_{k'}/k'}{1/k - 1/k'}, \tag{5.6}$$

where S_W is sensitivity coefficient of reactivity worth, R is property (reactivity worth), σ is cross section, S_k is sensitivity coefficient before substituting Al plates to Th plates, $S_{k'}$ is sensitivity coefficient after substitution, k is eigenvalue with Th sample and k' is eigenvalue without Th sample.

Sensitivity coefficients of Th reactivity worth for B7/8”P12ETEETEE and B10/8”P9ETEETEE cores is illustrated in Fig. 5.6 and standard deviation of ^{232}Th capture cross section of JENDL-4.0 is shown in Fig. 5.7. It indicates that error of the calculation mostly comes from the uncertainty of ^{232}Th cross section library at thermal range (from 0.05eV to 0.5eV) and resonance range (from 17eV to 4keV). The result at the thermal energy range shows more remarkable influence on the worth than the contribution from the resonance range and the more sensitive reactivity change due to neutron spectrum of B10/8”P9ETEETEE core than that of B7/8”P12ETEETEE core. By considering the standard deviation of ^{232}Th capture cross section of JENDL-4.0 (Fig. 5.7) at both energy ranges where the deviations have the same magnitude (about 20%), ^{232}Th capture cross section at thermal range (from 0.05eV to 0.5eV) is suggested to be adjusted. The reactivity change estimated when increasing capture cross section of ^{232}Th at the thermal energy range about 20%

indicates that the underestimation of the calculation can be compensated and the discrepancy between the calculated result and the experimental one caused by ^{232}Th cross section library can mainly be reduced.

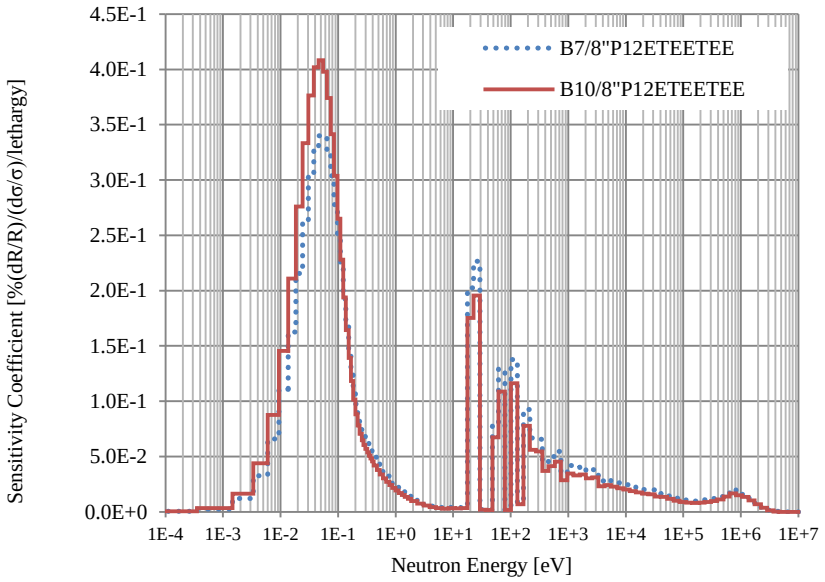


Fig. 5.6. Sensitivity coefficient of Th replacement worth of B7/8”12ETEETEE and B10/8”P9ETEETEE

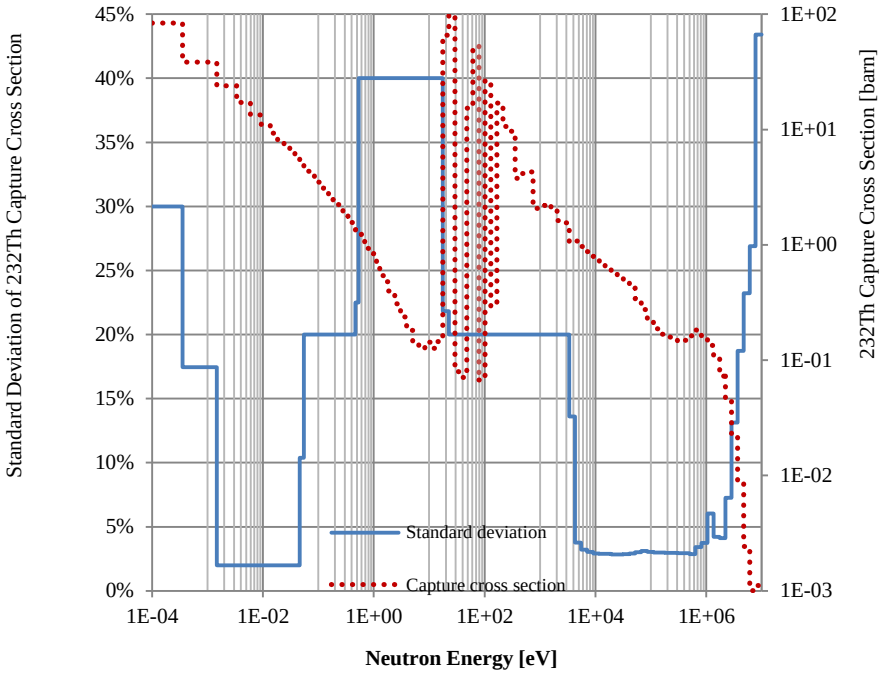


Fig. 5.7. Standard deviation of ^{232}Th capture cross section

5.4 Impact of thorium cross section discrepancy on thorium utilized ADS reactivity calculation

As explained above, the underestimation of ^{232}Th capture cross section in the thermal energy range is concluded to cause the larger effective multiplication factor in the calculation result compared to experimental result of thorium utilized core. Regarding the influence of uncertainty of thorium cross section library on the reactivity calculation on the fast system, the sensitivity calculations of effective multiplication factor of the seed and blanket ADS for ^{232}Th capture cross section of JENDL-4.0 and ENDF/B-VII library were done.

Calculations of sensitivity coefficient of k_{eff} of the seed and blanket ADS core for ^{232}Th capture cross section of JENDL-4.0 and ENDF/B-VII were conducted by using SAGEP code with cross section data attained by SRAC2006-CITATION. 2-D seed and blanket thorium-reprocessed fuel ADS model is used in the sensitivity calculation. The sensitivity coefficient S_k gives the relative change in the system multiplication factor value as a function of the relative change in the ^{232}Th capture cross section data [9]. The cross section

covariance data of ^{232}Th was derived from the JENDL-4.0 and ENDF/B-VII evaluated nuclear data file using ERRORR module for NJOY99 code.

The uncertainty (δE_k) is derived as in Eq.5.7 as follows

$$\delta E_k = \sqrt{S_k^T \cdot V \cdot S_k} \quad , \tag{5.7}$$

where S_k is sensitivity coefficient of k_{eff} and the subscript T means transposition; V covariance data for (n,γ) reaction of ^{232}Th.

Figure 5.8 illustrates the sensitivity coefficients of multiplication factor for capture reaction of ^{232}Th of JENDL-4.0 and ENDF/B-VII cross section library obtained using SAGEP code. As we can see from the figure, sensitivity coefficients of ^{232}Th capture cross section of two libraries mainly concentrate on the fast energy range and is almost the same because the difference between two libraries is very small (Fig. 5.9). The covariance data of two libraries was generated using NJOY99 code and illustrated in Figs. 5.10a and b. As we can see from the Figs. 6a and b, the correlation in the (n, γ) cross section for energy groups of JENDL-4.0 library is larger than that of ENDF/B-VII, especially in the energy range from 10 eV to 0.5 MeV and it is the source to cause the difference in the k_{eff} uncertainty between two libraries. As derived from the Eq. 1, the uncertainty of the k_{eff} caused by ^{232}Th capture cross section of JENDL-4.0 is about 1.3%. This uncertainty would cause a significant influence in reactivity calculation in the thorium utilized ADS evaluation. Thus, the uncertainty of ^{232}Th capture cross section of JENDL-4.0 is needed to be enhanced to give more reliable results on reactivity calculation for the fast system. On the other hand, the impact of uncertainty of ^{232}Th capture cross section of ENDF/B-VII is small (0.1%). Therefore, its influence on reactivity calculation of thorium fuel utilized ADS is not significant. It can be used for the fast spectrum ADS design purpose.

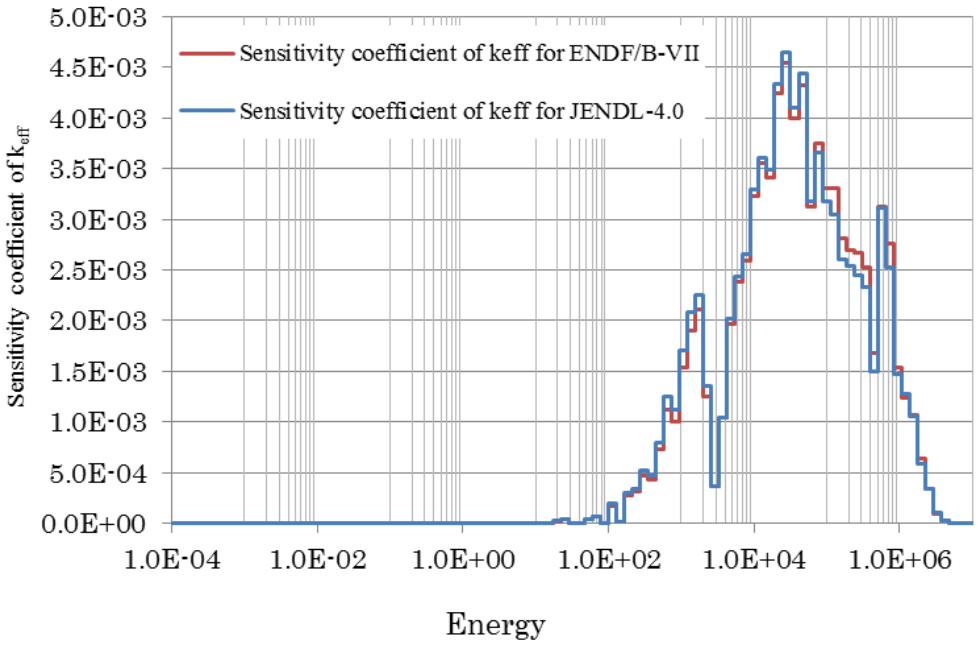


Fig. 5.8. Sensitivity coefficient of k_{eff} for ^{232}Th capture cross section of JENDL-4.0 and ENDF/B-VII libraries

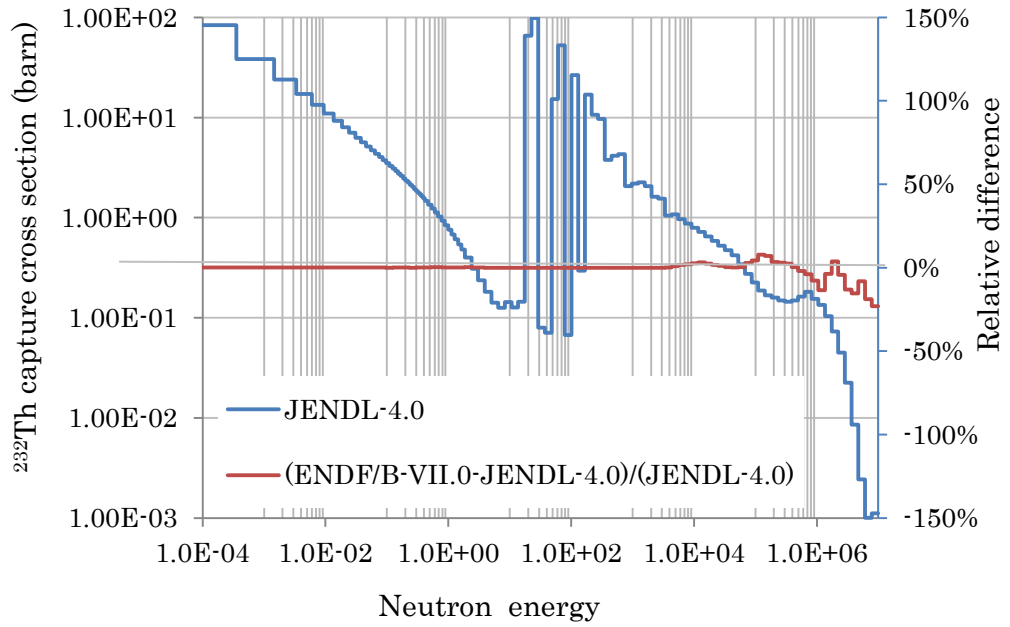


Fig. 5.9 ^{232}Th Capture Cross Sections of JENDL-4.0, and relative difference among JENDL-4.0 and ENDF/B-VII libraries

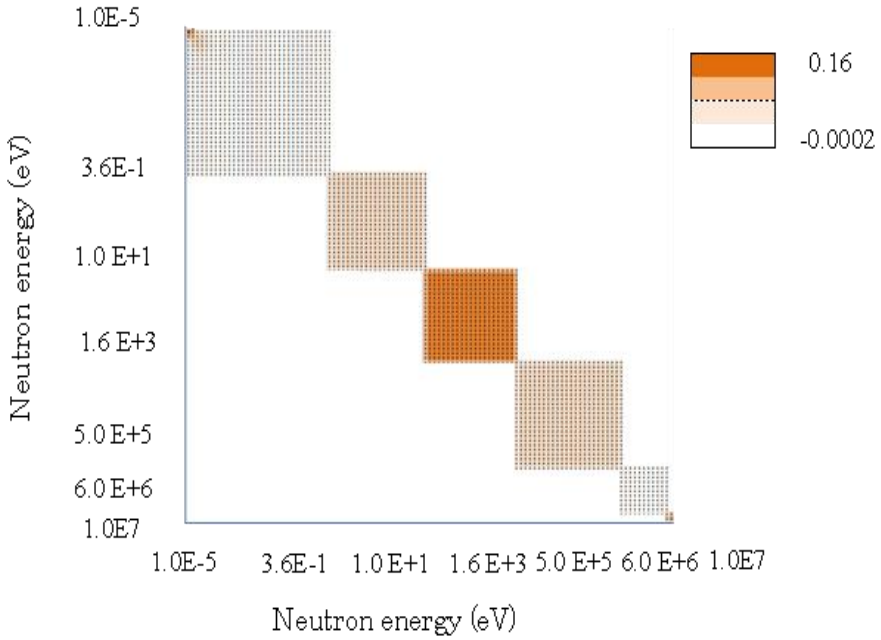


Fig. 5.10a. ^{232}Th Covariance Matrix of (n,γ) Reaction of JENDL-4.0

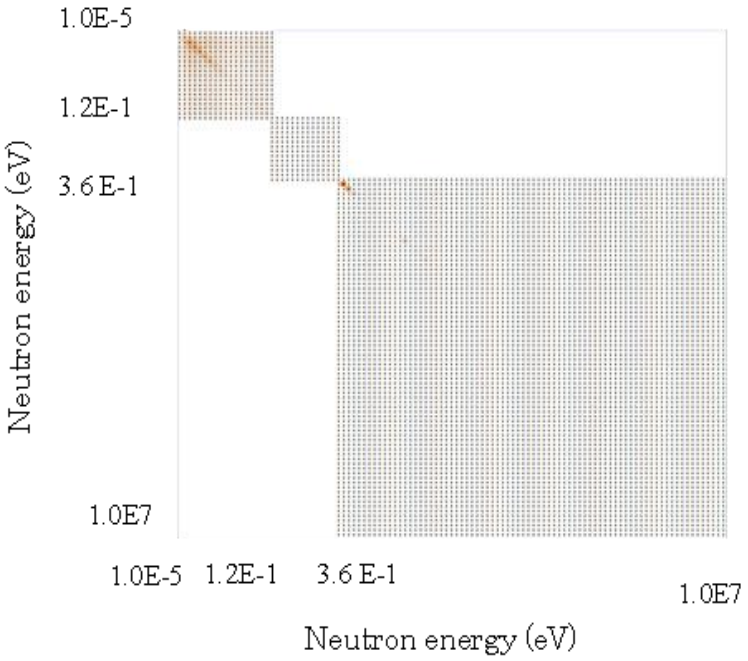


Fig. 5.10b. ^{232}Th Covariance Matrix of (n, γ) Reaction of ENDF/B-VII

5.5 Conclusions

In order to investigate the accuracy of the neutronics calculation method associated with ^{232}Th cross section library, analyses of B7/8”P12ETEETEE and B10/8”P9ETEETEE critical core experiments have been performed and compared with the calculated results obtained by MVP calculation. From the comparison between the measurement and calculation, it has been found that the calculation overestimates effective multiplication factor for critical cores about (0.90±0.01-0.99±0.01)%. The underestimation of ^{232}Th capture cross section of JENDL-4.0 library is concluded to mainly contribute on this discrepancy (0.78%). By comparing the measured Th sample worth with the calculated one, ^{232}Th capture underestimation of JENDL-4.0 library is reassured. The calculations of sensitivity coefficient of Th reactivity worth were conducted and its result shows the more contribution of ^{232}Th cross section uncertainty at the thermal range to the calculation error. It suggests that ^{232}Th capture cross section is needed to be adjusted at thermal energy range to obtain more reliable calculated data.

Regarding the influence of thorium cross section on the reactivity calculation of the thorium utilized fast system such as the seed and blanket ADS, the uncertainty of the k_{eff} cause by ^{232}Th capture cross section of JENDL-4.0 is about 1.3%. The uncertainty of JENDL-4.0 is needed to be enhanced in order to provide more reliable results on reactivity calculation for the fast system. The impact of uncertainty of ^{232}Th capture cross section of ENDF/B-VII is small (0.1%). Therefore, it will cause no significant impact of the thorium cross section library on the thorium utilized ADS design calculation.

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Chapter 6: **Conclusions**

The present thesis describes the results of the investigation on the seed and blanket ADS conceptual design for efficient TRU transmutation and power generation utilizing thorium-reprocessed fuel and its application in the case of nuclear power installation in Vietnam.

For this objective, a series of investigation and studies has been done and presented in this thesis as follows:

1. An investigation on the transmutation performance and safety features of the seed and blanket ADS using thorium-reprocessed fuel design concept.
2. An investigation on multi-cycle thorium-reprocessed fuel ADS in order to achieve the high utilization of thorium fuel for energy production and more efficient TRU transmutation.
3. The application of seed and blanket ADS to transmute the nuclear waste of future nuclear reactors in Vietnam is studied.
4. The thorium cross section data verification was done using the experimental and calculated data and the impact of thorium cross section uncertainty on seed and blanket ADS calculation was estimated.

The outline of this thesis and the results obtained in the course of this study are summarized as followed:

Chapter 1 gives an overview of energy production problems and the background of nuclear waste transmutation and thorium utilization potential for better understanding of the purpose of this thesis.

The design parameters and core configuration of the seed and blanket ADS using thorium – reprocessed fuel for efficient TRU transmutation and power production is described in Chapter 2. In this chapter, the innovative point of the designed compared to other designs was explained. With the reference configuration of the ADS, it shows a great potential of transmuting TRUs but the reactivity swing is large and is an obstacle for employing this core concept technically. Hence, the parametric survey on seed and blanket

ADS was done in order to reduce the reactivity swing as well as improve the core performance characteristics. It was found out that by introducing a larger amount of MA into the core, the more TRUs are transmuted. The smaller reactivity swing in the system can be achieved by getting more efficient thorium utilization. Besides, the increasing MA content also reduces the negative void reactivity of the system. On the other hand, increasing the core size by introducing more thorium and reprocessed fuel assemblies into the system reduces the reactivity swing as well as the void reactivity of the system. It also slightly reduces the TRU transmutation efficiency of the system. In order to match the safety feature during operation the original seed and blanket ADS configuration using the reprocessed fuel with enriched MA should be employed for efficient TRU transmutation and thorium utilization in the safe operation.

In chapter 3, the multi-cycle seed and blanket ADS core for higher thorium utilization and TRU waste transmutation is presented. Instead of wasting thorium fuel and its breeding ^{233}U, thorium fuel assemblies are remained inside the core and utilized to produce energy by employing multiple cycles. At the equilibrium state, the thorium fuel contribution to total core power is improved (35.7%) compared to the startup cycle (15%). It also has benefit in reducing the axial power peaking at BOC from 2.50 at startup cycle to 1.42 at equilibrium state. Compared to the startup cycle, the TRU transmutation rate of the equilibrium cycle reduces about 15 kg per ton of spent HM. However, the once burnt reprocessed fuel assemblies are shuffled and transmute more for a second time, thus, the TRUs are transmuted into a deeper level. At discharge, the reduction of radiotoxicity from the TRU waste is about 20% with 1% uncertainty, mainly due to the reduction of Pu and Am.

Chapter 4 discusses the application of the seed and blanket ADS in the case of nuclear power installation in Vietnam. The nuclear power employment in Vietnam in the future was described. The accumulating amount of spent nuclear fuel brings potential risk to the environment of the nuclear installation holding country. Although great amount of validation and verification are needed to be done to finalize the design of the seed and blanket ADS and much more effort is needed to utilize the seed and blanket ADS using thorium-reprocessed fuel, this design offers a potential solution to solve the TRU waste management in

Vietnam. With the TRU transmutation capability of the seed and blanket ADS, 14 units of ADS would be required to deal with the TRUs from the future reactors to be constructed in Vietnam and add about 4 GWe (40% thermal efficiency) to the grid. The seed and blanket ADS is not only the solution for the waste management problem in Vietnam but also improve the energy security in Vietnam by utilizing the thorium resource.

In Chapter 5, an additional study was performed in order to verify the thorium cross section data using the experimental and calculated data. From the comparison between the measurement and calculation, it was found that the calculation overestimates effective multiplication factor for critical cores about (0.90±0.01-0.99±0.01)%. The underestimation of ^{232}Th capture cross section of JENDL-4.0 library was concluded to cause the larger eigenvalue compared to that of the core using enriched U only in MVP calculation's results. By comparing the measured Th sample worth with the calculated one, ^{232}Th capture underestimation of JENDL-4.0 library was reassured. The calculations of sensitivity coefficient of Th reactivity worth were conducted and its result shows the more contribution of Th cross section uncertainty at the thermal range to the calculation error. It suggests that ^{232}Th capture cross section is needed to be adjusted at thermal energy range to obtain more reliable calculated data. Regarding the influence of thorium cross section on the reactivity calculation of the thorium utilized fast system such as the seed and blanket ADS, the uncertainty of the k_{eff} cause by Th capture cross section of JENDL-4.0 is about 1.3%. The impact of uncertainty of ^{232}Th capture cross section of ENDF/B-VII is small (0.1%). Therefore, ENDF/B-VII will cause no significant impact of the thorium cross section library on the thorium utilized ADS design calculation.

Throughout this study, the conceptual design of seed and blanket ADS using thorium-reprocessed fuel was proposed and its transmutation potential, thorium utilization, and other core performance were investigated. The seed and blanket ADS shows a great potential in TRU transmutation and utilizing thorium fuel to produce energy. It can be considered as the technology in the few decades for energy production without accumulating high radiotoxicity level waste.

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Achievements

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