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ACTINIDE COMPOSITION ANALYSIS OF LIGHT WATER REACTOR (LWR) FOR DIFFERENT REACTOR CONDITION OF BURNUP AND COOLING TIME

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ABSTRACT

Actinide production from spent nuclear fuel has being intensively monitored and controlled by the IAEA, to ensure all nuclear materials, nuclear fuel facilities including fuel reprocessing facilities are used only for civil and peaceful purposes. Spent nuclear fuel (SNF) from nuclear facilities such as from commercial reactors becomes one of the important issues in term of reducing environmental impact and fuel sustainability as well as nuclear non-proliferation point of view when those SNF materials can be recycled and utilized as "new" fuel loaded into the reactors. Several discharged fuel burnup and decay time effects which are used to evaluate those effect to actinide production of element compositions based on spent fuel of light water reactor (LWR) type have been evaluated in this present study. Those fuel burnup value are varied from 33 GWd/t up to 60 GWd/t and several decay times or cooling times process from 1 years to 30 years cooling time are also evaluated. The results show that actinide composition of each element and isotopic element have their own trend during reactor operation as burnup parameter and during cooling time process after the reactors are stop. Actinide element production is increasing with increasing burnup level except for uranium production which is decreasing as well as heavy metal in total production is also decreasing. Decreasing uranium production is estimated from reduction number of U-235 for fission reaction and it converts to fission product and capturing neutron by U-238 will convert to neptunium, plutonium up to curium. In case of longer cooling time, each actinide element is increasing except for plutonium and curium which show a decreasing trend for longer cooling time. Cooling time process is based on half-lives of material which can be estimated that decreasing plutonium and curium during cooling time process caused by some isotopes of those element have shorter half-lives.

Keywords: actinide, burnup, cooling time, plutonium, LWR.

INTRODUCTION

A program for recycling or re-use options of spent nuclear fuel (SNF) of nuclear reactors are one of the important issue, not only because of reducing factor of its effect to the environment, but also from fuel sustainability factor and nuclear non-proliferation point of view, especially for some accumulated spent nuclear fuels (SNF) of light water reactor (LWR). Actinide production from spent nuclear fuel has being intensively monitored and controlled by the IAEA, to ensure all nuclear materials, nuclear fuel facilities including fuel reprocessing facilities are used only for civil and peaceful purposes. Those actinides are by product material from nuclear reaction in the reactor core. Some researchers are defining that actinide as nuclear waste and others define as recycled nuclear fuel. Practical and commercial application nowadays has already used uranium and plutonium as new fuel after recycling and reprocessing processes. Neptunium, americium and curium as minor actinde are possible to be used and recycled as new fuel in some nuclear reactor. Spent nuclear fuel (SNF) from nuclear facilities such as from commercial reactors becomes one of the important issues in term of reducing environmental impact and fuel sustainability as well as nuclear nonproliferation point of view when those SNF materials can be recycled and utilized as "new" fuel loaded into the reactors. Adopted SNF recycling program of the reactors especially based on SNF LWR is required to recycle uranium and plutonium as initial fresh fuel, instead of once through fuel option.

The program of recycled spent nuclear fuels is adopted not only for reduction volume of SNF, but also to reduce the supply amount of enriched uranium as fresh fuel as well as more conversion ratio of nuclear fuel capability [1]. Recycling program of SNF not only for uranium fuel cycle case, but also an option of recycling SNF of thorium fuel for thermal reactor and supply fuel of fast breeder reactors[2-3]. A typical recycling option which is based on uranium and plutonium as fresh fuel of MOX fuel type as well commercialized recycling fuel option and some feasible option for recycling MAs are also can be adopted including some transuranium fuel type. Some programs for recycling trans-uranium for extending reactor operation as long life core reactor program [4-5], as well as a program to increase high burning MA capability, to improve some transmutation

TRU capability [6-7] and a program for recycling MA in LWR type as well as subcritical assembly [8-10] have been conducted many years. A reduction program based on some potential of nuclear proliferation issue should be evaluated and strengthened including the program to increase the level of proliferation resistance. One of the potential transmutation option is by adopting doping or loading minor actinide (MA) in the reactors to produce more protected plutonium proliferation by high composition of even mass of plutonium from converted Materials for increasing level of proliferation resistance of nuclear materials [11-14]. Not only thermal reactor utilization, some fast reactor technology and some other generation four of reactor types shows some advantages. Those benefit such as high fuel breeding and reducing spent fuels by burning process during reactor operations. It is commonly known that fast reactor type has easier capability to obtain criticality condition and higher fuel breeding. In addition, loading minor actinide (MAs) into the reactors also can be considered to improve higher level of nuclear non-proliferation, especially to produce more protected plutonium production [11-14].

In the present study, several discharged fuel burnup level and decay time effects are used to evaluate those effect to actinide production of element compositions based on spent fuel of light water reactor (LWR) type have been evaluated. Those fuel burnup values are varied from 33 GWd/t up to 60 GWd/t. Actinide compositions after discharged from the reactors for several decay times from 0 years to 30 years cooling time are also evaluated. Some actinides are based on the composition of uranium, plutonium as well as minor actinide which consist of neptunium, americium and curium. Burnup level or irradiation process will be estimated from fission process in the reactors as a main contributor and cooling time process will be basically comes from the decay actinide process.

METHODS

Spent fuel of LWR is evaluated as a main purposes which is one of the important issue for recycling fuel scheme from LWR fuel cycle to be used into other reactors including thermal reactor such as Ligh water reactor (:WR) as well as fast reactor such as fast breeder reactors (FBR). In order to obtain specific spent fuel of LWR is required to be produced from LWR operation as discharged fuel. Spent fuel composition of LWR type is obtained from simulation by using ORIGEN code [15] for different fuel burnup levels and cooling time process of PWR. The simulation of ORIGEN is performed to evaluate fuel behavior and fuel composition which is estimated to be sensitive to the different burnup and decay fuel time processes. Fuel behaviors of LWR from initial fresh fuel up to end operation and cooling time process after discharged fuel from the reactors are based on fuel burnup level of 33 GWd/t up to 60 GWd/t burnup levels, and decay or cooling time processes from 0 year up to 30 years cooling time. The evaluation of some actinides are based on the SNF composition of LWR such as uranium and transuranium compositions. Transuranium is based on the composition of plutonium and minor actinide which consist of neptunium, americium and curium. Burnup level or irradiation process will be estimated from fission process in the reactors as a main contributor which is based on the fissile material of U-235 through capturing thermal neutron and make some fission reaction to maintain the reactor in operation. Ccooling time process is also evaluated which is basically comes from the decay actinide process of its half-live. Conversion process of actinide composition from one actinide to another actinide is estimated to have a typical composition because of the different reactor condition such as burnup level and cooling time process.

RESULTS AND DISCUSSIONS

Some important results are shown and discussed in this section such as the trend of heavy nuclide production, heavy nuclide composition for different burnup and cooling time as well as minor actinide analysis. Heavy nuclide or actinide will be shown based on the compsotion of uranium, plutonium. Neptunium, amaricium and curium for several burnup level and several cooling time process. Comparative analysis has been done for evaluating plutonium and minor actinide (MA) production based on different burnup and cooling time.

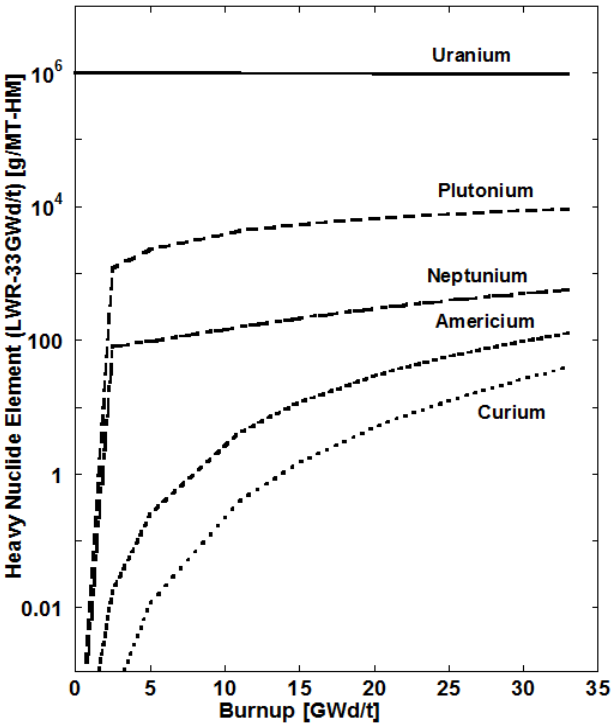

Figure-1. Heavy nuclide element composition as a function of Burnup based on 33 GWd/t LWR.

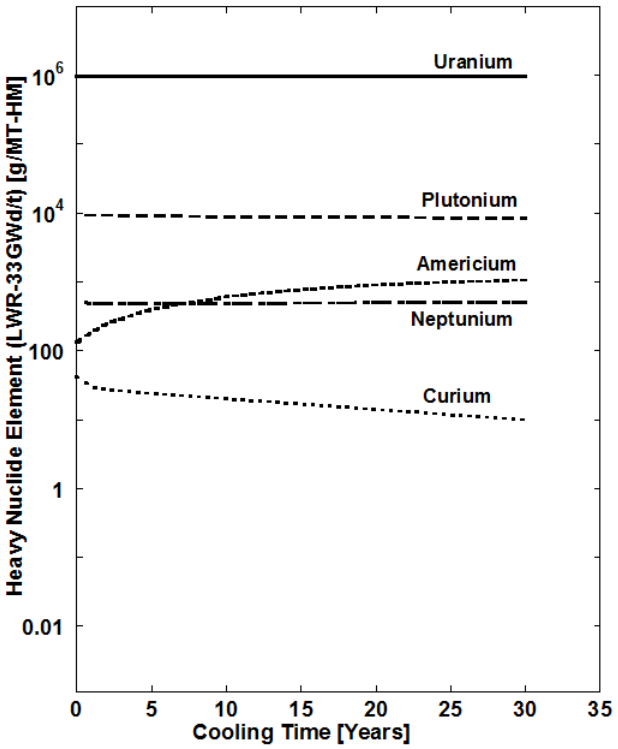

Figure-2. Heavy nuclide composition as a function of decay or cooling time based on 33 GWd/t LWR.

Figure-1 shows heavy nuclide element profile as a function of burnup based on the ligh water reactor (LWR) of 33 GWd/t burnup level. The function of burnup shows a reactor operation or fuel irradiation process which gives a significant production profile for actinide of uranium, neptunium, plutonium, americium and curium elements. As expected, uranium as fresh fuel gives a dominant composition followed by plutonium as the second dominat actinide element and neptunium, americium and curium. At the beginning of operation, only uranium exist as a fuel initial loading in the reactor and by increasing the operation time or irradiation process, actinide elemnt of trans-uranium from neptunium up to curium are produced until the reactor is shutdown or end of operation. At the end of operation, the main actinides are shown by uranium and plutonium and the less actinide or some minor actinides (MA) are shown by neptunium, americium and curium [16]. Uranium is decreasing with increasing the time, while others actinides are increasing along the operation time. Decreasing uranium actinide is estimated from the decreasing composition of U-235 caused by the fission process to maintain the criticality condition for the reactor operation as well as in the same time, some isotop U-238 are converted to neptunium and plutonium. This phenomenon shwos fission process is estimated by U-235 through neutron capture to produce some fission products and fuel convertion process of U-238 into neptunium and plutonium. This is why, uranium is monotonically decreasing along the operation time. Neptunium is produced by uranium and in short time they convert to plutonium and plutonium will convert to americium and into curium. In plutonium isotopes there are some fissile materials which have high probability to make a fission reaction during reactor operation, however, due to the production of plutonium is still higher, the total plutonium element production is remining higher during operation.

Obtained result which shows the heavy nuclide element profile as a function of cooling time or decay actinide process is shown in Figure-2. Figure shows a dreasing trend of heavy nuclide element compsotion along cooling time except for americum which gives an increasing composition with increasing cooling time. Naturally, decreasing composition of nuclides is estimated from half-live constant of actinides which decreasing with time. Decreasing composition means a conversion process from initial or mother of actinide converts to other actinide as a daugther actinide some neptunium decays into plutonium as well as plutoniums decay to some americium isotopes. Decreasing and increasing composition of actinides during cooling time are depending on the rate of production and consumption. If some actinides have relatively higher consumption than production will obtain some decreasing composition. In the same way for increasing production of actinide composition. Americum is increasing with time because of the production rate from converted plutonium is relatively higher than conversion process of americium to other actinides. Higher production of americium composition is mainly coming from production of amercium isotope of Am-241 from decay process of plutonium isotope of Pu-241 which relatively short half live as mentioned in this references [13-14,17]. Different with actinide or heavy nuclide comspotion during irradiation process or burnup as mentioned in Figure-1, heavy nuclide comspotion during cooling time obtains relatively higher composiition of americum for longer time in comparing with neptunium which has relatively higher compsotion during irradiation process.

To estimate the effect of burnup value or irradiation level to the heavy nuclide composition, some different level of burnup are employed such as burnup level of 33 GWd/t up to 60 GWd/t. Obtained results of burnup level effect to the composition of heavy nuclides are shown in Figure-3. The datas are obtained form the composition of actinide when the reactor is shutdown and no cooling time process. It shows higher burnup level affect to decrease and to increase some heavy nuclide which is estimated from the fission process as well as absorption and capturing processes of those heavy nuclides.

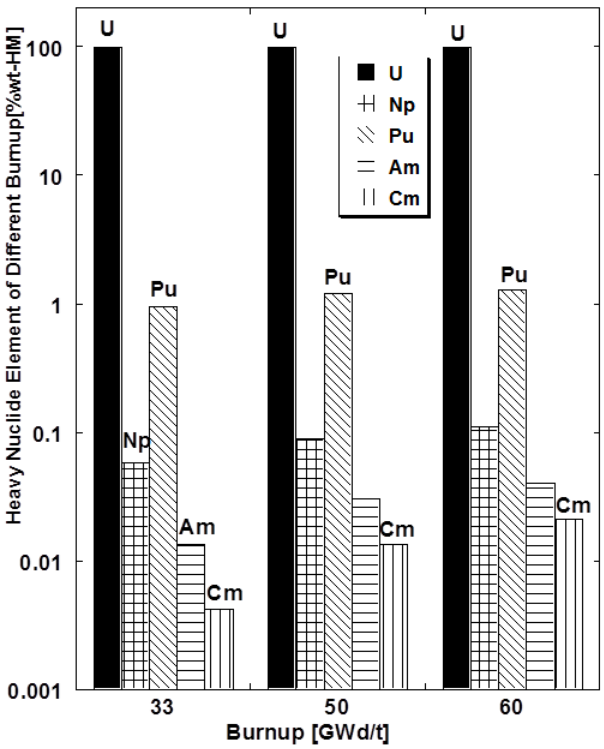

Figure-3. Heavy nuclide element composition for different burnup at no cooling time.

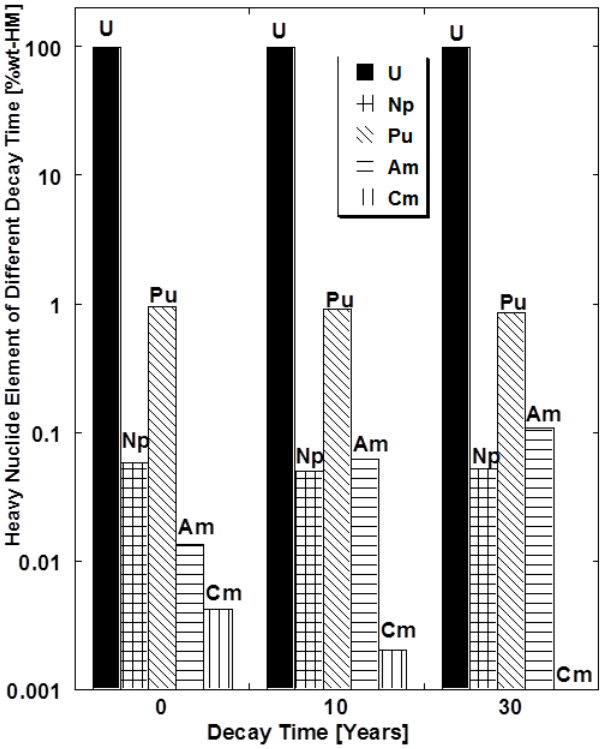

Figure-4. Heavy nuclide element composition for different cooling or decay time based on 33 GWd/t LWR.

Uranium is relatively less composition for higher burnup level and other heavy nuclides such as neptunium up to curium as transuranium actinides are increasing for higher burnup level. Decreasing uranium composition is estimated from the fission process of U-235 for maintaining the reactor operation [1, 16-17] and absorption process of U-238 which converts to transuranium actinide such as neptunium and plutonium. As the second main actinide, the composition of plutonium is higher for higher burnup level because of the conversion process of U-238 through neutron capture. More than 1% of composition is obtained for higher burnup level incomparing with 33 GWd/t burnup level which obtaines less than 1 % plutonium composition. Some significant increasing compositions are shown by minor actinide such as neptunium, americium and curium. More minor actinide (MA) production is estimated from the decreasing uranium which converts to some neptuinium and plutonium and moreover through neutron capture from plutonium it converts to higher actinide such as americium and curium as shown in Figure-3. Burnup level of LWR will give some different compositions of heavy nuclides which bring some increasing composition or give some decreasing value of actinide composition. To choose the SNF LWR will be depending on the reactor condition such as burnup level that produces different composition for heavy nuclide in different burnup levels.

Effect of cooling time process or decay time after the reactor is shuttdown to heavy nuclide composition has been also evaluated for, some different level of cooling time from 0 up to 30 years. These cooling time effects to heavy nuclide composition is shown in Figure-4 based on burnup level of 33 GWd/t. Obtained data shows 0 year, 10 years and 30 years as representative cooling time level for beginning of cooling time of 0 year, intermediate cooling time of 10 years in this evaluation and 30 years cooling time for longer cooling time condition. The datas are obtained form the composition of actinide when the reactor is shutdown and no cooling time process. As mentioned before in Figure-1, heavy metal composition trend is relatively same trend for all condition as a function of cooling time which shows longer cooling time produce less of heavy metals composition except for americium composition that obtains higher than beginning of cooling time process. Neptunium composition at the beginning of cooling (0 years cooling) show higher than americium, and its composition of americium becomes higher after 10 years cooling time up to 30 years of cooling time in comparing with neptunium. Similar to the effect of burnup level that produce different compositions of heavy nuclides, cooling time effect also gives some different compositions of heavy nuclides. This condition is estimated that to choose a spesific actinide composition should be in a spesific parameter such as in what level of

burnup of LWR and in what certain level of cooling time after the reacyor is shutdown. A database of actinide or heavy compositions will be one of the important issue to optimize the actinide composition level to be used for recycling option as well as one through cycle process.

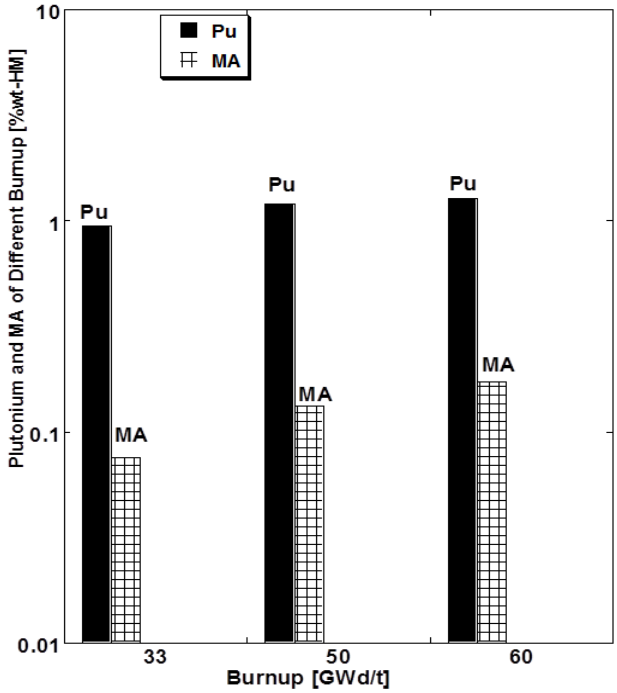

Figure-5. Plutonium and minor actinide (MA) composition for different burnup.

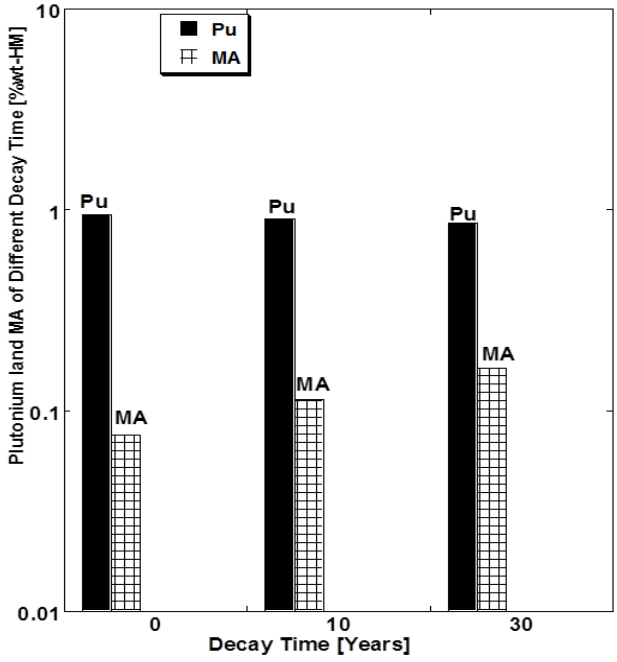

Figure-6. Plutonium and minor actinide (MA) composition for different decay time.

Comparative analysis of minor actinide (MA) and plutonium productions based on different burnup level and cooling time are also evaluated to estimate transuranium production which is shown in Figures-5 dan 6. Figures show production of plutonium and MA for some burnup levels and cooling time conditions. Minor actinide as well as plutonium obtains higher composition for higher burnup level. In case of longer cooling time, composition of MA is achived higher, while composition of plutonium is less for longer cooling time as shown in Figure 6. Higher composition of MA is achieved for higher burnup and longer cooling time is estimated from the composition trend of neptunium, americium and curium for different burnup and cooling time as mentioned before in Figures-3 and 4. As a basic composition of MA is based on the summation of neptunium, americium and curium. Both effects of burnup and cooling time show the composition of minor actinide is increasing with increasing burnup and cooling time. This condition gives the information that transuranium composition including plutonium and minor actinide will be produced more or higher for higher burnup and cooling time, except for plutonium which shows less production for longer cooling time.

Based on this analysis, actinide composition of LWR operation will be depending on the spesific parameter of burnup level as well as cooling time condition which gives some specific actinide composition. Uranium composition is estimated becomes less composition for higher burnup and longer cooling time, while plutonium composition will be produced more for longer burnup level and its composition becomes less for longer cooling time. Decreasing uranium production is estimated from reduction number of U-235 for fission reaction and it converts to fission product and capturing neutron by U-238 will convert to neptunium, plutonium up to curium. Minor actinide (MA) composition is estimated becomes more production for higher burnup level and longer cooling time condition. Some decreasing compositions are shown by all composition of actinides except for americium which shows a significant trend of increasing composition for longer cooling time because of the decay component of plutonium especially based on decay process of Pu-241 and it convert to Am-241 as a main actinide for americium element [13-14, 17]. Cooling time process is based on half-lives of material which can be estimated that decreasing plutonium and curium during 30 years caused by some isotopes of those element have shorter half-lives.

CONCLUSIONS

The effects of several discharged fuel burnups and decay time have been evaluated to the actinide production of element compositions based on spent fuel of light water reactor (LWR). Parameter of burnup level is varied from 33 GWd/t up to 60 GWd/t. Actinide compositions after discharged from the reactors for several decay times from 1 years to 30 years cooling time are also

evaluated. To optimize and analyze the burnup and decay time effects to the reactor behavior as well as fuel composition including actinide composition, An ORIGEN computation code for typical LWR type is adopted. The results show that actinide compositions of each element have their own trend during reactor operation as burnup parameter and during cooling time process after the reactor are shutdwon. Based on this analysis, actinide composition of LWR operation will be depending on the spesific parameter of burnup level as well as cooling time condition which gives some specific actinide composition. Uranium composition is estimated becomes less composition for higher burnup and longer cooling time, while plutonium composition will be produced more for longer burnup level and its composition becomes less for longer cooling time. Minor actinide (MA) composition is estimated becomes more production for higher burnup level and longer cooling time condition. Some decreasing compositions are shown by all composition of actinides except for americium which shows a significant trend of increasing composition for longer cooling time because of the decay component of plutonium especially based on decay process of Pu-241 and it convert to Am-241 as a main actinide for americium element as well as mentioned in this references [13-14,17].

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REFERENCES

[1] Sidik P, Takaki N, Sekimoto H. 2006. Impact of Different Moderator Ratios with Light and Heavy Water Cooled Reactors in Equilibrium States, Ann. Nucl. Energy, 33, 561-572.

[2] Sidik P., Takaki N. and Sekimoto H. 2011. Breeding and void reactivity analysis on heavy metal closed-cycle water cooled thorium reactor, Ann. Nucl. En. 38, 337-347.

[3] Ismail, Peng Hong LIEM, P. Sidik, Naoyuki TAKAKI, Hiroshi SEKIMOTO. 2009. Symbiotic systems consisting of large-FBR and small water-cooled thorium reactors (WTR), Ann. Nucl. Energy, 33, 1076-1085.

[4] Sidik P., Zaki S. 2009. Core Performance and Plutonium Production of Small Long Life Fast Reactor using Doping Actinides, Proc. of ICAPP, May 10-14, 2009, Tokyo-Japan.

[5] Sidik. P., Zaki S., and Suzuki M. 2009. Trans-uranium doping utilization for increasing protected plutonium proliferation of small long life reactor, Proc. of Global, September 6-11, 2009, Paris, France.

[6] Sidik P., Takaki N., and Sekimoto H. 2011. Breeding and void reactivity analysis on heavy metal closed-cycle water cooled thorium reactor, Ann. Nucl. En. 38, 337-347.

[7] Ismail, Peng H. L., Sidik P., TAKAKI N, SEKIMOTO H. 2009. Symbiotic systems consisting of large-FBR and small water-cooled thorium reactors (WTR), Ann. Nucl. Energy, 33, 1076-1085.

[8] Choi H. And Downar T. J. 1999. A Liquid-Metal Reactor for Burning Minor Actinides of Spent Light Water Reactor Fuel-I: Neutronics Design Study, Nucl. Eng. Des. 133, 1-22.

[9] Edward A. H. and Weston M. S. 2003. Comparative Fuel Cycle Analysis of Critical and Subcritical Fast Reactor Transmutation Systems, Nucl. Technol., 144, 83.

[10] Taiwo T. A., *et al*. 2006. "Assessment of a Heterogeneous PWR Assembly for Plutonium and Minor Actinide Recycle", Nucl. Technol., 155, 34.

[11] Saito M. 2002. Multi-Component Self-Consistent Nuclear Energy System for Sustainable Growth, Prog. in Nucl. Energy, 40[3-4], 365.

[12] MEILIZA Y., SAITO M. and SAGARA H. 2008. Protected Plutonium Breeding by Transmutation of Minor Actinides in Fast Breeder Reactor, J. Nucl. Sci. Technol. Vol. 45[3], 230.

[13] Sidik P., Suzuki M., Saito M. 2011. Effect of TRU Fuel Loading on Core Performance and Plutonium Production of FBR, Nucl. Eng. Des, 241, 101-117.

[14] Sidik P., Suzuki M., Saito M. 2011. Basic Analysis on Isotopic Barrier of Material Attractiveness Based on Plutonium Composition of FBR, J. Nucl. Sci. Technol., 48[5], 724-733.

[15] Ludwig S. B. and Croff A. G. 2002. "ORIGEN 2.2 Isotope Generations and Depletion Code Matrix

Exponential Method", Oak Ridge National Laboratory.

[16] Sidik P., Abdul W, Takaki N and Sekimoto H. 2009. All Heavy Metals Closed-Cycle Analysis on Water-Cooled Reactors of Uranium and Thorium Fuel Cycle Systems, Proc. Of ICAPP, May 10-14, 2009, Tokyo-Japan.

[17] Sidik P., Suzuki M., Saito M., Novitrian, Abdul W., Zaki S., Study on material attractiveness aspect of spent nuclear fuel of LWR and FBR cycles based on isotopic plutonium production, Energy Conversion and Management. 72 (2013) 19-26.