Comparison of Irradiation Effect of ThO$_2$ and ThZrH on Neutronic Parameter of the TRIGA 2000 Reactor

Rasito$^{1,2,*}$, S. Permana$^1$, and P. Ilham Y.$^1$

$^1$Nuclear Physics Laboratory, Physics Department, Bandung Institute of Technology, Jl. Ganeca 10, Bandung, Indonesia

$^2$Center for Science and Applied Nuclear Technology, National Nuclear Energy Agency, Jl. Tamansari 71, Bandung, Indonesia

Email: rasito@batan.go.id

Abstract. The present work discusses two different models of thorium irradiation in the TRIGA 2000 reactor core to compare the neutronic characteristics of irradiation of thorium dioxide (ThO$_2$) and thorium zirconium hydride (ThZrH). The Monte Carlo N-Particle (MCNP) Transport code, based on the Monte Carlo method, is used to design three dimensional models for TRIGA reactor core at typical operating power 2 MW. These models are used to determine the effective multiplication factor (k$_{\text{eff}}$) and neutron flux distribution. The ORIGEN code is used to calculate the $^{233}$U isotope production. It is found that decreasing of k$_{\text{eff}}$ from the effect of hydride irradiation rod is a little lower than the oxide irradiation rod.

1. Introduction

The $^{232}$Th is a fertile material and its neutron irradiation yields a new fissile nuclide $^{233}$U by two successive beta decays of $^{233}$Th as depicted in Fig. 1. When $^{232}$Th absorbs a neutron, it becomes $^{233}$Th, which has a half-life of only 22 minutes. $^{233}$Th decays into $^{233}$Pa through beta decay. $^{233}$Pa has a half-life of 27 days and beta decays into $^{233}$U. The $^{233}$U isotope is a long-lived isotope that has a half-life of 160,000 years. $^{233}$U usually fissions on neutron absorption, but sometimes retains the neutron, becoming $^{234}$U.

![Figure 1. Nuclear transformations of $^{232}$Th after neutron irradiation [1]](image-url)
The fissile contents $^{233}\text{U}$ yields 2.5 neutrons per fission and therefore can be bred from $^{232}\text{Th}$ with external neutron source without requirement of enrichment technology [2]. From the beginning of a nuclear power programme, the immense potential of $^{232}\text{Th}$ for breeding fissile isotope $^{233}\text{U}$ efficiently in a thermal reactor has been recognised [3]. The TRIGA 2000 reactor at the Center for Science and Applied Nuclear Technology (PSTNT) of the National Nuclear Energy Agency (BATAN) is a pool type thermal reactor with a power of 2000 kW. The TRIGA 2000 reactor can be used to produce $^{233}\text{U}$ by irradiation of thorium.

The simulation of thorium ($\text{ThO}_2$) irradiation in the TRIGA 2000 reactor core to produces $^{233}\text{U}$ has been done in our previous study [4]. The ThO$_2$ rod irradiated in G4, E8, and A1 positions for one operating cycle at 2 MW power. The simulation result was shown that the irradiation of ThO$_2$ at the A1 position is produce largest $^{233}\text{U}$ isotope mass. For this reason in this paper a Thorium irradiation in two composition, ThO$_2$ and ThZrH, was simulated in the TRIGA 2000 reactor core. This study was conducted to answer two problems. First is effect of oxide and zirconium hidryde to percentage of $^{233}\text{U}$ can be produced. An the second is effect of oxide and zirconium hidryde to effective multiplicator factor ($k_{\text{eff}}$) of the core.

The effective multiplication factor ($k_{\text{eff}}$) is the neutronic parameter with respect to the control and safety of reactor. The $k_{\text{eff}}$ defined as the ratio of the neutrons generated to the sum of the neutrons absorbed plus those lost in any one generation of system. The change in the reactor power level is directly affected by $k_{\text{eff}}$. The power level in reactor is directly proportional to the neutron density, the reactor is subcritical when $k_{\text{eff}} < 1$, and the reactor is supercritical if $k_{\text{eff}} > 1$. Finally, $k_{\text{eff}} = 1.0$, the reactor is critical and operates at a constant power level [5].

2. Methods

The thorium (ThO$_2$ and ThZrH) irradiation in the TRIGA 2000 reactor core was simulated with the Monte Carlo method using the MCNPX computer code. The MCNP code will calculate the flux and neutron energy in the A1 irradiation position and the $k_{\text{eff}}$ value of the reactor core. The isotopes product from irradiation were calculated using ORIGEN. In these calculations it needs several models; geometry and composition of irradiation rod, irradiation position in the core, and configuration of TRIGA 2000 core.

2.1 Thorium irradiation rod

The geometry of Thorium irradiation rod adjusts to the fuel geometry in the TRIGA reactor. The thorium irradiation rod has a length of 54.102 cm and a diameter of 3.32 cm in a 4.2 mm thick aluminum tube as shown on Fig. 2. This first simulation uses ThO$_2$ and the second simulation use ThZrH. The composition of ThO$_2$ rod and ThZrH was shown in the Table 1.

![Figure 2. Thorium irradiation rod](image-url)
Table 1. Composition of ThO$_2$ and ThZrH rods

| Element | Mass (g) | ThZrH rod | ThO$_2$ rod |
|---------|----------|-----------|-------------|
| Th      | 1.5E+02  | 1.5E+02   |             |
| Zr      | 2.0E+03  | -         |             |
| H       | 3.5E+01  | -         |             |
| O       | -        | 2.1E+01   |             |

2.2 Irradiation positions

The TRIGA core is shaped as a right hexagonal featuring 90 slots, distributed over 7 concentric rings, which can contain 82 fuel elements, 3 graphite elements, 5 control rods, and irradiation channels. The fuel consists of a uniform mixture of uranium 8% wt. (41 fuels), 12% wt. (36 fuels), and 20% wt. (5 fuels) with enriched 20% wt. in $^{235}$U, zirconium (91% wt.) and hydrogen (1% wt.).

Figure 3. Irradiation positions in the TRIGA core

ThO$_2$ or ThZrH irradiation rods was placed inside the TRIGA core in A1 positions, as shown on Fig. 3. The A1 irradiation position has largest neutron flux in the core. Variations of irradiation materials will be used to observe the effect of irradiation on the $k_{\text{eff}}$ value and $^{233}$U production.

2.3 The $k_{\text{eff}}$ values and $^{233}$U production

The $^{233}$U activity produced in neutron irradiations depends on cross sections, neutron spectrum, flux distribution, concentration of precursors of each radionuclide, and irradiation time. After irradiation, activities decrease with time and disintegration constants. Activation reactions can be simulated using MCNPX. It is a code based on the Monte Carlo (MC) method and the number of reactions calculated can be converted into activity [6]. Assuming that the fluxes and energy particles remain constant during the time step and locally in selected volume, the concentrations of $^{233}$U or other nuclides at the end of irradiation step can be obtained. This assume was used the ORIGEN2 [7] depletion code to obtain the nuclide concentrations at the end of irradiation [8].
There are two inputs of MCNPX; ThO$_2$ and ThZrH irradiated at A1. Each MCNPX input was run on a computer to get the $k_{\text{eff}}$ value and neutron flux in each irradiation position. In MCNPX input was used KCODE and F4 tally [9]. Has been used the KCODE with 100000 initial neutrons, 250 effective cycles and 50 ineffective cycles for the parameter calculations. The neutron flux value was used as an ORIGEN input to calculate the number of isotopes produced, both in irradiated rods and in fuel. From the calculation of ORIGEN, the percentage of $^{233}$U isotope will be obtained and also the composition of the new material in the fuel and irradiation rod. The new composition in the fuel rod and irradiation will be used to modify a new MCNPX input to calculate the new $k_{\text{eff}}$. Thus this was done until the value of $k_{\text{eff}} > 1$, which means that the reactor core is not critical. The calculation algorithm used was as shown in Fig. 4.

3. Results and Discussion

The MCNPX input has been made for irradiation position at A1. In order to make the calculation easier, the initial condition of the reactor core is contained fresh fuels. MCNPX output was obtained after the four inputs are run using a computer with an Intel core i5-2.3 GHz processor, 4GB RAM, and Windows 8.1 operating system. To run each input using the computer it takes an average of 145 minutes for the initial conditions, and 1000 minutes after the terrace reaches subcritical. In this simulation the interaction of neutrons with materials was taken from ENDF60 cross sections.

**Figure 4.** Algorithm of simulation

**Figure 5.** Neutron spectra in the irradiation position
Table 2. Neutron flux at the A1 position

| Neutron | Flux (cm$^{-2}$s$^{-1}$) |
|---------|-------------------------|
|         | ThZrH rod | ThO$_2$ rod |
| Termal  | 4.65E+13  | 3.20E+13    |
| Epitermal | 3.44E+13  | 3.53E+13    |
| Fast    | 3.46E+13  | 3.99E+13    |
|         | 1.15E+14  | 1.07E+14    |

The MCNPX simulation produces neutron flux and $k_{\text{eff}}$ values for normal conditions and irradiation conditions with 2 different material. Various irradiation material on the reactor core generally have the same shape of the neutron spectrum with only a different total flux value. In Fig. 5 it has shown the neutron spectrum at position A1 with the total neutron flux at ThO$_2$ is lower than ThZrH, $1.07 \times 10^{14}$ n/cm$^2$/s for ThO$_2$ and $1.15 \times 10^{14}$ n/cm$^2$/s for ThZrH. The termal neutron flux at ThO$_2$ is lower than ThZrH, $3.2 \times 10^{13}$ n/cm$^2$/s for ThO$_2$ and $4.65 \times 10^{13}$ n/cm$^2$/s for ThZrH. But the fast neutron flux at ThO$_2$ is higher than ThZrH, $3.99 \times 10^{13}$ n/cm$^2$/s for ThO$_2$ and $3.46 \times 10^{13}$ n/cm$^2$/s for ThZrH.

In normal conditions the $k_{\text{eff}}$ value of TRIGA core is 1.10231. The ThO$_2$ irradiation rod in A1 decreases the $k_{\text{eff}}$ value to 1.10097, as well as the irradiation of ThZrH rod with a $k_{\text{eff}}$ value of 1.09996. This $k_{\text{eff}}$ value shows that irradiation of ThO$_2$ rod will make the fuel life longer than the irradiation of ThZrH rod. However, when viewed from the aspect of the neutron flux value, the rate of reaction of $^{232}\text{Th}(n,\gamma)^{233}\text{U}$ will be greater at ThZrH.

Neutron flux value on the different thorium irradiation that produced by MCNPX output was used as an ORIGEN input to calculate the production of isotopes in the irradiation rods and in the fuels. To be able to calculate the number of isotopes produced, the ORIGEN input also includes the operating time of the reactor. A significant decrease in $k_{\text{eff}}$ value generally occurs when the reactor was operated at 0.5 - 1 day. The ORIGEN output lists the isotopes produced in the fuel and irradiated rods. New compositions in the fuel and irradiation rods have been used to modify material data in the MCNPX input. The new MCNPX input with the new composition materials in the fuels and the irradiation rods is then run on the computer to get a new $k_{\text{eff}}$ value.

The $k_{\text{eff}}$ values as function of irradiation times

The calculation of the $k_{\text{eff}}$ value of the composition of new materials produced by ORIGEN was carried out up to a certain operating time to obtain a pattern of decreasing $k_{\text{eff}}$ values and isotope production rates of $^{233}\text{U}$. The pattern of decreasing $k_{\text{eff}}$ value was also used to predict the reactor life.
were shown in Fig. 6. The reactor will reach subcritical conditions when the $k_{\text{eff}}$ value is $< 1$. The isotope production in ThZrH irradiation rod is shown in Fig. 7.

The $k_{\text{eff}}$ value of the reactor core as a function of the reactor operating time for ThO$_2$ and ThZrH is decrease with the same pattern as shown in Fig. 6. For normal conditions, the TRIGA 2000 reactor operating time is around 510 days. The addition of the ThO$_2$ irradiation rod in A1 decreases the $k_{\text{eff}}$ value by 0.01 and is almost constant for the operating time function. The difference in the $k_{\text{eff}}$ value makes the reactor operating age to be 236 days for ThZrH rod irradiation, 7 days shorter than ThO$_2$ rod.

![Figure 7. Isotopes production of ThO$_2$ irradiation rod at the A1 position](image1)

The amount of mass of the isotope produced depends on the rate of production and the rate of decay. Although the production rate of $^{233}$Th is high but because the decay time is short 22 minutes, the mass of $^{233}$Th functions is relatively small. Isotope $^{233}$Pa has a very large production rate derived from the decay products of $^{233}$Th, but because $^{233}$Pa has a half-life of 27 days, the decay rate is also large. Isotope $^{233}$U has a large production rate derived from $^{233}$Pa decay products, while the decay rate is very small because it has a very large half-life of 160,000 years. This phenomenon is the result of the irradiation process carried out continuously. Whereas if the irradiation is done so that only a decay occurs, the increase in the number of isotopes of $^{233}$U will occur significantly, while $^{233}$Th and $^{233}$Pa will decrease sharply.

![Figure 8. The $^{233}$U production in ThO$_2$ and ThZrH rods](image2)
As shown in Fig. 8, isotope production of $^{233}\text{U}$ for ThZrH is larger than ThO$_2$. The mass of $^{233}\text{U}$ was the result of irradiation of ThZrH where during the operating life of the reactor 300 days the level of $^{233}\text{U}$ is 0.6 g and for ThO$_2$ is 0.45 g.

4. Conclusion

The simulation of thorium irradiations at A1 position has been successfully carried out on the TRIGA 2000 reactor core for two different materials, ThO$_2$ and ThZrH using MCNPX and ORIGEN. The $k_{eff}$ value for a normal core with a whole new fuel is 1.10231, even for irradiation of ThO$_2$ rod the value decrease to 1.10097 and for irradiation of ThO$_2$ rod the value decrease to 1.09996. In one operating cycle, 300 days produced $^{233}\text{U}$ in ThZrH rod is 0.6 g and from ThO$_2$ rod is 0.45 g.

Acknowledgments

The authors gratefully acknowledge to the financial support given to this work from Menristekdikti, and from joint cooperation between National Nuclear Energy Agency (BATAN) and Bandung Institute of Technology (ITB).

References

[1] D. Baldova, R. S koda, J. Kuc’era, L. Viererbl, J. Uhlí’r, Feasibility study of 233Pa and 233U determination in neutron irradiated thorium for future applications in thorium–uranium nuclear fuel cycle, J Radioanal Nucl Chem, 288:37–42, 2011
[2] Guanheng Zhang, Advanced Burner Reactor with Breed-and-Burn Thorium Blankets for Improved Economics and Resource Utilization, dissertation, University of California, Berkeley, 2015
[3] Zohreh Gholamzadeh, Seyed Amir Hossein Feghhi, Leila Soltani, Marzieh Rezaazadeh, Claudio Tenreiro, Mahdi Joharifard, Burn-up calculation of different thorium-based fuel matrices in a thermal research reactor using MCNPX 2.6 code, NUKLEONIKA, 59(4):129-136, 2014
[4] Rasito, S. Permana, and P. Ilham Y., Simulation of Thorium Irradiation In the TRIGA 2000 Reactor Core For $^{233}\text{U}$ Production, ICES 2018, Bandung, Sept 2018
[5] Mohammed Mghar and Abdelouahed Chetaine, Effect of Graphite Reflector on the Neutronic Parameters of the TRIGA Mark II Research Reactor, Advanced Studies in Theoretical Physics, Vol. 9, no. 14, 659 – 666, 2015
[6] José RÓDENAS, Sergio GALLARDO, Elisa CECCOLINI, Wolfgang HANSEN, Validation of the Monte Carlo Model Developed to Estimate the Neutron Activation of Stainless Steel in a Nuclear Reactor, Joint International Conference on Supercomputing in Nuclear Applications and Monte Carlo 2010 (SNA + MC2010), Japan, October 17-21, 2010
[7] A. G. Croff, “A user’s manual for the ORIGEN2 computer code,” Tech. Rep. ORNL/TM-7175, Oak Ridge National Laboratory, 1980.
[8] A. Stankovskiy and G. Van den Eynde, AdvancedMethod for Calculations of Core Burn-Up, Activation of StructuralMaterials, and Spallation Products Accumulation in Accelerator-Driven Systems, Science and Technology of Nuclear Installations, Volume, 2012
[9] Pelowitz, D. B., MCNPX 2.6.0 user manual, Los Alamos: Los Alamos National Laboratory (LA--CP-07-1473), 2008