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Abstract. SAMOP (Subcritical Assembly for Molybdenum-99 Production) has the potential to use thorium as fuel to produce ^{99}Mo after modifying the design, but the production performance has not been discovered yet. A study needs to be done to obtain the correlation between ^{99}Mo production with the mixed fuel composition of uranium and with SAMOP power on the modified SAMOP design. The study aims to obtain the production of ^{99}Mo based thorium nitrate fuel on SAMOP's modified designs. Monte Carlo N-Particle eXtended (MCNPX) is required to simulate the operation of the assembly by varying the composition of the uranium-thorium nitrate mixed fuel, geometry and power fraction on the SAMOP modified designs. The burnup command on the MCNPX is used to confirm the ^{99}Mo production result. The assembly is simulated to operate for 6 days with subcritical neutron multiplication factor (k_{eff} = 0.97-0.99). The neutron multiplication factor of the modified design (k_{eff}) is 0.97, the activity obtained from ^{99}Mo is 18.58 Ci at 1 kW power operation.

1. Introduction

Radioisotope ^{99}Mo is one of the most essential radioisotopes in the nuclear medical field. It is required as a parent radionuclide in a radioisotope generator system that produces ^{99m}Tc for diagnostic purposes. Radionuclides ^{99}Mo can be produced in several ways, i.e.: through a ^{235}U fission reaction fired with neutrons produced on the deuteron and proton accelerator reaction (D, n) and (p, n), through the activation of neutrons from ^{98}Mo (n, γ) ^{99}Mo, through the process of photography, by utilizing ^{100}Mo isotope, which is ^{100}Mo (γ, n) ^{99}Mo. [1]

The world's demand for ^{99}Mo is produced by some of the world's major reactors such as the National Research Universal (NRU) in Canada, High Flux Reactor (HFR) in the Netherlands, Belgian Reactor-2 (BR-2) in Belgium, South African Fundamental Atomic Research Installation-1 (Safari-1) in South Africa and most recently the Australian Open Water Pool Light Reactor (OPAL) in Australia [2]. However, the global crisis of ^{99}Mo has happened recently because of some reactors have been ceased. Two major reactors, HFR and SAFARI-1, were shut down in 2013-2014, this led to a major ^{99}Mo supply disruption. OSIRIS, a light-water-cooled research reactor located in France, was shut down in December 2015. The shutdown of BR-2 happened in March 2015 due to refurbishment, including the replacement of the Beryllium reflector. The operating license of NRU should be ended in 2016, but the Canadian government has announced that NRU will continue to operate until March 2018 in order to support the global demand crisis, but NRU will not be used for routine production of

^{99}Mo. This extended operating license was a settlement due to the unexpected circumstances of ^{99}Mo shortage from 2016 to 2018. [3]

Based on those facts, new research toward reactors continue to develop to meet the world's needs for ^{99}Mo. Nearly ^{99}Mo of total production with reactor using HEU (High Enriched Uranium) as target. The development of other alternative fuels and targets is also continuously sought with respect to the RERTR (Reduced Enrichment for Research and Test Reactors) program linked to the use of HEU to LEU (Low Enriched Uranium) with less than 20% uranium enrichment [4]. The OPAL reactor is one of the nuclear facilities that uses LEU as nuclear fuel as well as nuclear target to produce ^{99}Mo.

The half-life of radioisotope ^{99}Mo is about 66 hours, and the half-life of its daughter, ^{99m}Tc, is about 6 hours. Therefore, it is needed an assembly that can provide the ^{99}Mo locally and regionally. In Indonesia, BATAN has proposed Subcritical Assembly for Molybdenum-99 Production (SAMOP) in order to supply the needs of ^{99}Mo especially in Indonesia. The initial concept of SAMOP is to use a fuel containing slightly ^{235}U with a neutron multiplication factor value (k_{eff}) smaller than one or subcritical ($0.97 \leq k_{eff} \leq 0.99$). With this concept, SAMOP does not require the same harsh reactor safety requirements. SAMOP is designed to use low enriched uranyl nitrate as the fuel. [5]

The development of design and production analysis is needed to complete the data towards SAMOP's compatibility. The flexibility of SAMOP using different fuel needs to be discovered, for that reason, the research to use thorium as fuel is conducted. Apart from the use of LEU, the use of thorium as nuclear fuel is also widely developed. Thorium (^{232}Th) is a fertile material that can be used to produce fissile material of ^{233}U isotope inside the reactor. The use of thorium as a fuel has several advantages over the use of uranium, other than its abundance of 3 to 4 times more than uranium abundance, the thorium-^{233}U cycle also produces fewer minor actinides than uranium-plutonium cycles. Then, thorium is also superior in safety aspect because it does not require enrichment process, and it produces less waste than uranium. [6]

Studies to see the potential of SAMOP using thorium nitrate mixed fuel have been done, however, the neutron multiplication value (k_{eff}) obtained is much smaller than the expected range. Therefore, a modified design is considered to meet the range of subcritical neutron multiplication factor value (k_{eff}). SAMOP is able to operate with thorium nitrate after modifying the patent design, but, study of the ^{99}Mo productivity characteristics from the modified design has not yet been analyzed [7]. Thus, this study is important to be done to determine the productivity characteristics of ^{99}Mo if using the mixture of thorium nitrate as its fuel and target.

2. Monte Carlo N-Particle eXtended (MCNPX)

The Monte Carlo method is a method of calculation by applying probability and statistical laws to the natural sciences. This method uses various random number distributions in which each distribution is a representation of a process that occurs sequentially. The process, for example, is the diffusion of neutrons in various materials, Monte Carlo method is used to calculate samples that is close to the actual value of the neutron diffusion. The Monte Carlo method was later converted into a computational science system used to predict random events in nature.

MCNPX (Monte Carlo N-Particle Code eXtended) is developed by Los Alamos National Laboratory (Los Alamos National Laboratory). MCNPX contains radiation transfer codes. It is designed to track many types of particles over a wide energy range. MCNPX is used for simulating nuclear processes such as fission and other simulations involving neutrons, photons and electrons. Specifically, MCNPX can be used for simulation of radiation protection and dosimetry, radiation shield, radiography, medical physics, nuclear safety, detector design and analysis, target accelerator design, fission and fusion reactor design, decontamination and decommissioning. The MCNPX software is basically a newer version of its own MCNP software. An important additional feature of MCNPX is the burnup calculation used in this study to obtain fission product results after the reactor is operated. [8]

The basic structure of MCNPX code consists of cell card, surface card, and data card. Data entered in the form of cell card contains information in the form of cell number, material density and cell

position based on certain reference point. The surface card contains the constituent form of the design, such as circles, rectangles and balls. The data card contains the desired command to be completed, such as the burnup code and also the type of reactor constituent material, including the composition fraction, the mass number and the atomic number of the material [8]. The MCNPX software comes with Visual Editor software used to see the design of reactor geometry that has been created in either two or three dimensions.

3. Method
A study towards the design of the assembly according to SAMOP patent number 34511 is conducted, then the related parameters that affect the design especially the neutronic characteristic is determined. The geometry design of SAMOP is created using program MCNPX. The patent SAMOP design is the reference to determining the thickness and height of the cores, cladding, and reflector. [5]

The fuel composition using thorium nitrate (Th $(NO_3)_4$) is calculated. SAMOP uses two main fuels, the TRIGA fuel, which is a component of the Kartini reactor [9], and the mixture of uranium-thorium nitrate. The mixture of uranium-thorium nitrate lies at the center and annulus fuel tube of the assembly. After incorporating the design dimension and fuel composition, the MCNPX program is run to determine the value of the reactor neutron multiplication factor. The desired value of the neutron multiplication factor is in the range 0.97 to 0.99 to be regarded as a subcritical reactor. Variations to obtain the desired criticality value can be done by changing the design of the reactor or changing the reactor fuel composition.

The burn code on MCNPX is then inserted to get the result of fission product especially to get the radioisotope ^{99}Mo result. Burn-up is performed in one 6-day operation cycle for each variation of the fuel composition. The variation of SAMOP power is also performed. The results on each variation of the nuclear mixed fuel and the power variation are analyzed and then compared. The minimum production of ^{99}Mo is expected to be worth 300 mCi of each operating cycle. The Kartini reactor in BATAN, Yogyakarta, currently operates at a power of 1 kW. Thus, the ^{99}Mo production value obtained and can be applied in the near term is at a power of 1 kW. The flowchart of the research is described in Figure 1.
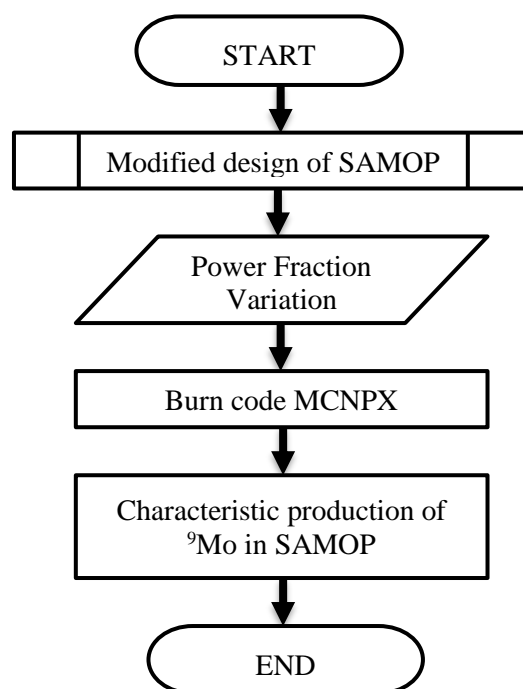

Figure 1. Flowchart of the Research

4. SAMOP Modified Design

SAMOP modified design has the following geometric dimension as shown in the Table 1.

Table 1. The Geometry of SAMOP Modified Designs

Parameter	Mod-1
Fuel	Thorium Nitrate
Center Fuel (Tube) Diameter	5 cm
Annulus Fuel (Tube) Diameter	31.4 cm
Center Fuel (Tube) Height	46 cm
Annulus Fuel (Tube) Height	40 cm
Number of Fuel Rods	12
Cladding Thickness	3.6 cm
Reflector Thickness	20 cm
Cladding	0.1 cm
Reflector	Graphite
Coolant	Water
Framework	SS-316

The designs later inserted in the MCNPX cards. Both designs can be viewed using Visual Editor as shown in Figure 2. It can be seen that the color determines each materials used or contained in the design. The light blue color on the edge of the design is the framework of the assembly (SS316), while dark blue is the water coolant, yellow is the reflector (graphite), orange is the TRIGA fuel, red in the center and annulus reactor is the fuel in the form of mixed uranium-thorium nitrate, and light red above the fuel is the air.
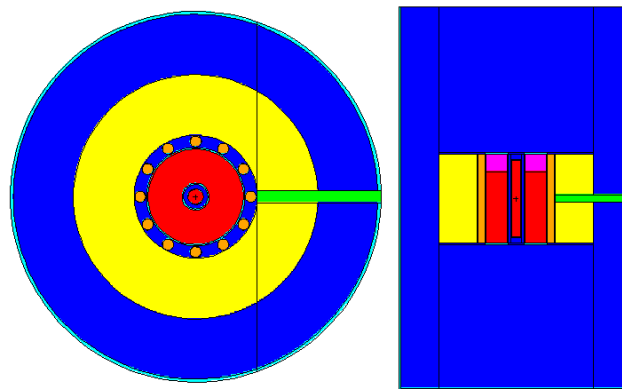

Figure 2. SAMOP Modified Design

5. Result and Analysis

The modified design is analyzed by varying the composition of uranium in the mixed uranium-thorium nitrate fuel. The uranium-thorium nitrate mixed fuel consists of 95% water and 5% uranium-thorium nitrate mixed fuel. The ratio between uranium and thorium in that 5% mixed fuel is varied, if the research uses 20% of ^{233}U in the mixed fuel, it means the fraction of uranyl nitrate fraction (UO_2 (NO_3)$_2$) is 1% of the total fuel solution and the remaining 4% is thorium nitrate (Th (NO_3)$_4$).

The density of the uranium-thorium nitrate mixed fuel is calculated using the same formula to calculate the density of uranyl nitrate from the SAMOP patent [10]. The output result of the MCNPX will show information such as mass, atom density, fraction density and the activity of all the actinides and non-actinides on the first day and the sixth day operation in each burned materials. It also provide

information such as the neutron multiplication factor (k_{eff}) of the assembly. Certain information needed to analyze the performance of SAMOP is chosen to be compared as shown in Table 2. The ^{99}Mo of production is obtained with a power operation of 1 kW with operating time for 6 days.

Table 2. K_{eff} Value and the ^{99}Mo Based on the ^{233}U Fraction Variations

^{233}U fraction	K_{eff}	Std. Deviation	^{99}Mo in the U-Th Fuel		^{99}Mo in the TRIGA UZrH	
			Mass (gr)	Activity (Ci)	Mass (gr)	Activity (Ci)
20%	1.02795	0.00290	4.077E-05	19.59	3.244E-05	15.59
18%	1.01366	0.00255	3.976E-05	19.11	1.911E+01	16.40
16%	0.97542	0.00250	3.867E-05	18.58	3.541E-05	17.02
14%	0.94605	0.00246	3.714E-05	17.85	3.670E-05	17.64
12%	0.91305	0.00275	3.561E-05	17.11	3.915E-05	18.81
10%	0.87221	0.00243	3.318E-05	15.94	4.207E-05	20.22

It can be seen from the Table 2 that 16% uranium fraction meets the qualification of neutron multiplication factors in the subcritical range, which is 0.975. The assembly operates at a 1 kW power operation, and it obtains about 18.58 Ci of the ^{99}Mo activity.

SAMOP operates at the subcritical state, therefore, the composition with 16% of ^{233}U is used as the control variable to obtain the ^{99}Mo activity by varying the SAMOP power. The activity of ^{99}Mo multiplies along the multiplication of the power. It can be seen in the Table 3 the activity resulted in different value of power. SAMOP Power is multiplied ten times from 1 kW to 100 kW.

Table 3. Activity of ^{99}Mo based on the Power Variations

Power (kW)	^{99}Mo Activity (Ci)
1	18.58
10	184.4
100	1832

Besides ^{99}Mo, there are more fission products (non-actinides) resulted in the assembly. Below is shown radioisotopes using 16% fraction of ^{233}U as control variable and at 1 kW SAMOP power.
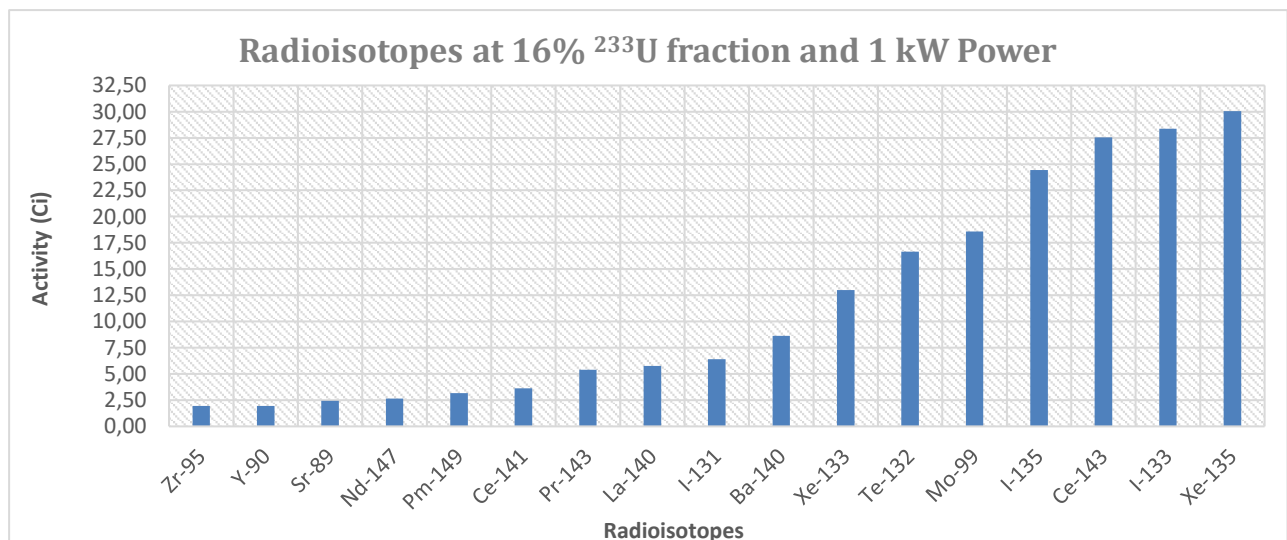

Figure 3. Radioisotopes Resulted in the Assembly

6. Conclusion

MCNPX can be used to simulate a nuclear assembly to obtain the neutronic and productivity characteristic. The characteristic that is obtained by analyzing the result can be used to complete the data regarding to the performance of SAMOP, particularly using thorium nitrate as fuel mixture. It is also provide the consideration for refinement and development of modified SAMOP design. The total activity of radioisotope ^{99}Mo both in center and annulus fuel tubes on the modified design is 18.58 Ci with a 1 kW power operation, neutron multiplication factor = 0.975 (subcritical).

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