Neutronic analysis of subcritical assembly for $^{99}$Mo isotope production based on thorium nitrate fuel

F Yassar$^1$, A W Harto$^1$ and Syarip$^2$

$^1$Department of Nuclear Engineering and Engineering Physics, Jl. Grafika No. 2, Kampus UGM, Yogyakarta 55284
$^2$Center for Accelerator Science & Technology, National Nuclear Energy Agency (BATAN) Jl. Babarsari, Yogyakarta 55281

*syarip@batan.go.id

Abstract. The paper discusses a neutronic analysis of the SAMOP reactor based on thorium nitrate as fuel and target materials. The reactor core model consists of an annular cylindrical tube with a tube in the center, filled with thorium nitrate as fuel. The analysis is done for the variation of fuel enrichment, the first case use thorium nitrate with 16% $^{233}$U enrichment both on the annulus and the center of the core, and the second use 10% $^{233}$U enrichment in the annulus and 14% enrichment in the center. The methods used for this analysis is a modelling calculation using MCNPX computer code. The analysis results show that the neutron multiplication factor ($k_{eff}$) is $0.97706 \pm 0.00250$ with conversion ratio (CR) of 0.1175 and $k_{eff} = 0.98113 \pm 0.00216$ with CR = 0.1704, for the first and second case respectively. The need of $^{233}$U for the first case and the second case are 1.448 kg and 4.383 kg, respectively. The results show that the sub-criticality requirement ($k_{eff} < 1$) for both cases was achieved, but from the fuel utilization, the first case is more efficient. Based on the CR values the SAMOP cannot be categorized as a breeder reactor.

1. Introduction

The Subcritical Assembly for Molybdenum-99 Production (SAMOP) is a subcritical reactor device to produces $^{99}$Mo. $^{99}$Mo is a parent radioisotope used to produced $^{99m}$Tc which is used as medical diagnostics and cancer therapy. Generally, $^{99}$Mo production to $^{238}$U target (over 90%) and nuclear reactors with high neutron flux. The use of highly enriched uranium is at risk of being misused for nuclear weapons. In order to prevent such abuse, the international community is coordinating a Reduced Enrichment for Research and Test Reactor (RERTR) program, which aims to replace highly enriched uranium targets into low-enriched uranium targets or with non-uranium targets [1].

The purpose of this research are to find the amount the neutron multiplication factor ($k_{eff}$) and the Conversion Ratio (CR). $k_{eff}$ is the multiplication factor that takes leakage into account which is defined as the ratio of the neutrons produced by fission in one generation to the number of neutrons lost through absorption and leakage in the preceding generation. CR is defined as the ratio of fissile material created to fissile material consumed either by fission or absorption. The requirements of $k_{eff}$ and CR for SAMOP are $k_{ef} = 0.97-0.99$ and CR > 1. To achieve these two conditions, variations of SAMOP-related parameters include of SAMOP geometry, fuel composition, and number of UZrH fuels in the ring is varied for optimization.

The previous researches in relation to SAMOP neutronic analysis was done [2-4]. In the fulfillment of its safety aspect, the previous SAMOP design was adjusted for parameters related to the criticality
that is height and the diameter of the core is varied but the ratio should not be the same (H / D ≠ 1) [4]. A related literature review of thorium fuel information is mainly taken from IAEA-TECDOC-1450 [5]. Thorium is 3 to 4 times more abundant than uranium and is widely distributed in nature as an easily exploitable resource in many countries. Unlike natural uranium, which contains ~0.7% fissile \(^{235}\text{U}\) isotope, natural thorium does not contain any fissile material and is made up of the fertile \(^{232}\text{Th}\) isotope only. Hence, thorium and thorium-based fuel as metal, oxide or carbide, has been utilized in combination with fissile \(^{235}\text{U}\) or \(^{239}\text{Pu}\) in nuclear research and power reactors for conversion to fissile \(^{233}\text{U}\), thereby enlarging the fissile material resources [5].

The world’s demand for uranium-based fuels per year is expected to grow annually. It is estimated that the uranium demand will increase to 82,000 tonnes per year by 2025. By 2025 it is also estimated that secondary sources such as military stocks and uranium re-enrichment will decline and are expected to meet only 4-6% of the deficiencies. At that time, the introduction of the thorium fuel cycle will play a complementary role and ensure the availability of easy fuel for nuclear fission energy [5, 6]. The thorium deposit in Indonesia was investigated by Ngadenin [7]. Before undertaking large-scale utilization of thorium for nuclear power fuel, special treatment is required, i.e. to change fertile \(^{232}\text{Th}\) to the fissile \(^{233}\text{U}\). There are two ways that change thorium to \(^{233}\text{U}\), i.e. first, is an open fuel cycle, which is based on \(^{232}\text{Th}\) irradiation that produces fissile \(^{233}\text{U}\), without involving chemical separation of \(^{233}\text{U}\), and the second is a closed fuel cycle. The closed fuel cycle is based on chemical processing of irradiated thorium and recycled \(^{233}\text{U}\) fuel [5].

2. Theoretical Background

The National Nuclear Energy Agency (BATAN) has designed a reactor that could produce \(^{99}\text{Mo}\) but did not categorized it use as critical reactor and did not use highly-enriched uranium, known as SAMOP or Subcritical Assembly for \(^{99}\text{Mo}\) Production [1, 3]. The system is simple and safer if it compared to critical reactor because it is operated at a subcritical condition, but it need a continuous external neutron source along it operation.

2.1. Nuclear Criticality

Criticality is the ratio of the rate of fission reactions in one generation to the rate of catch and the rate of neutrons leaked in the previous generation. In general, this reaction can be described in equation (1).

\[
^{235}_{92}\text{U} + \frac{1}{3}\text{n} \rightarrow X1 + X2 + (2 - 3) \frac{1}{3}\text{n} + E
\]  

(1)

\(X1\) and \(X2\) are fission fragments followed by 2 or 3 neutrons (n) and \(E\) is energy generated in a fission. In a nuclear reactor, for a stable fission reaction the fission production rate must be kept constant. Figure 1 is illustrating the process of fission chain reaction [8].

![Figure 1. Chain fission scheme.](image)

Neutrons will be generated from fission reaction, then the neutron will be absorbed or emitted out of the reactor. Comparison of the number of neutron populations in the new generation with the neutron population in the previous generation is called neutron multiplication factor (\(k_{\text{eff}}\)). The reactor criticality can be stated by the \(k_{\text{eff}}\), when the number of neutron populations in one generation equals with the number of neutron populations in the previous generation or \(k_{\text{eff}} = 1\), the reactor can be said to be critical or stable. In this critical condition the fission chain reaction will depend on time. When the
number of populations in one generation is smaller than the neutron population in the previous generation \((k_{\text{eff}} < 1)\), the reactor condition is in subcritical condition. Under the subcritical conditions, the number of neutron populations will decrease as the fission reaction runs until the fission reaction is completely stopped. When the number of neutron populations in one generation is greater than the number of previous neutron populations \((k_{\text{eff}} > 1)\), the reactor is said to be in supercritical condition.

2.2. Conversion Ratio

A quantity that characterizes this conversion of fertile into fissile material is known as conversion ratio (CR) or conversion factor. The CR is defined as the ratio of fissile material created to fissile material consumed either by fission or absorption [9]. If the ratio is greater than one, it is often referred to as the breeding ratio, the reactor is creating more fissile material than it is consuming. In this case, the CR is the ratio between the thorium capture reaction rate and the uranium uptake reaction rate in the center and the annulus such as described in equation (2).

\[
CR = \frac{\text{neutron absorption reaction rate in fertile material}}{\text{neutron fission reaction rate in fissile material}}
\]

The reactor may be said as a breeder if \(CR > 1\) i.e. the neutron capture rate in the thorium is greater than the uranium absorption neutron reaction rate. The calculations performed to find the magnitude of CR can be expressed in equation (3).

\[
CR = \frac{\phi_A \sigma_{c,\text{Th}} N_{\text{Th}}^{232} V_A + \phi_{\text{Triga}} \sigma_{c,\text{U}}^{238} N_{\text{U}}^{238} V_{\text{Triga}}}{\phi_C \sigma_{f,\text{U}}^{233} N_{\text{U}}^{233} V_C + \phi_{\text{Triga}} \sigma_{f,\text{U}}^{235} N_{\text{U}}^{235} V_{\text{Triga}}}
\]

where: \(V_A\), \(V_C\), and \(V_{\text{Triga}}\) are volume (\(\text{cm}^3\)) of annulus, center tube, and Triga fuel respectively, \(\phi_A\), \(\phi_C\), and \(\phi_{\text{Triga}}\) are neutron flux (\(\text{n/cm}^2\)s) at the annulus, center, and Triga fuel, \(\sigma_{c,\text{Th}}\) and \(\sigma_{f,\text{U}}\) are neutron capture and absorption cross section (barn) of thorium and uranium respectively. \(N_{\text{Th}}\) and \(N_{\text{U}}\) are atomic density of thorium and uranium (atom/\(\text{cm}^3\)). In equation (3), atomic density \((N)\) is defined as the number of nuclides or atoms per unit volume. In this calculation the density of this atom can be approximated by modifying Avogadro’s law which can be seen in equations (4) [10, 11].

\[
N = \frac{\rho}{M} A_V \left[\text{atom} \right] = \frac{\rho}{M} A_V \left[\text{atom/cm}^3 \right] = \frac{\rho_{\text{eff}}}{M_e} A_V \left[\text{atom/cm}^3 \right]
\]

where: \(\rho_{\text{eff}}\), \(M_e\), and \(A_e\) are effective density of material (\(\text{g/cm}^3\)), mass number, and Avogadro number.

3. Methods

The reference design used for this research is the latest SAMOP BATAN design which was designed using uranil nitrate fuel. The SAMOP BATAN design will be re-modeled using MCNPX with the fuel replaced by thorium nitrate. Thorium nitrate is mixed with \(^{233}\text{U}\) fissile material, and then the calculation and analyses is done for various geometry model configurations to find the value of \(k_{\text{eff}}\) and conversion ratio (CR).

The parameter varied to calculate the \(k_{\text{eff}}\) for the SAMOP BATAN design is \(^{233}\text{U}\) mass fraction parameters on thorium nitrate fuel. If the \(k_{\text{eff}}\) obtained from SAMOP BATAN has not reached the expected \(k_{\text{eff}}\) range, then another parameter variation should be performed. In this study the geometry variation is done by adding the number of TRIGA fuel (UZrH) and changing the size of the fuel geometry of the solution. This research will also calculate the value of CR. The flow diagram used in the study can be seen in figure 2.
Figure 2. Flowchart for SAMOP neutronic analysis.

4. Result & Discussion

Redesigning of the SAMOP BATAN design, resulting in two new designs ie SAMOP Mod-1 and SAMOP Mod-2. The design has some supporting features in the form of addition (loading) of TRIGA UZrH fuels suround the reactor core. The function of TRIGA UZrH fuels to increase neutron multiplication, the characteristic of this fuel can be found in reference [12]. Loading of UZrH fuels continued until the SAMOP core reaches the optimum subcritical criterion value. The technical specification of SAMOP BATAN can be seen in figure 3, whiles, SAMOP Mod-1 and SAMOP Mod-2 are shown in figure 4. The technical specification of the SAMOP Mod-1 and SAMOP Mod-2 are described in table 1.

| Parameter                      | SAMOP Mod-1     | SAMOP Mod-2     |
|--------------------------------|-----------------|-----------------|
| Fuel                           | Thoriurn nitrate| Thoriurn nitrate|
| Diameter of Center Tube        | 5 cm            | 19 cm           |
| Diameter of Annulus            | 31,4 cm         | 69,4 cm         |
| Height of Center Tube          | 40 cm           | 40 cm           |
| Height of Annulus              | 46 cm           | 46 cm           |
| No. of TRIGA Fuel Rod          | 12              | 12              |
| Fuel Diameter (TRIGA)          | 3,6 cm          | 3,6 cm          |
| Reflector Thickness            | 20 cm           | 20 cm           |
| Cladding Thickness             | 0,1 cm          | 0,1 cm          |
| Reflector                      | Graphite        | Graphite        |
| Coolant                        | Water (H₂O)     | Water (H₂O)     |
The calculation and analysis result of $k_{\text{eff}}$ values as function of mass fraction of $^{233}\text{U}$ in thorium nitrate for the SAMOP model reactor based on the patent and SAMOP Mod-1 is described in figure 5. The variations of $^{233}\text{U}$ fraction in the thorium nitrate solution ranging from 10%, 12%, 14%, 16%, 18% and 20%. From figure 5 can be seen that for SAMOP design based on BATAN patent, at the maximum $^{233}\text{U}$ fraction of 20%, the maximum $k_{\text{eff}}$ can be reached is $0.89787 \pm 0.00269$. Whiles at SAMOP Mod-1, at the maximum fraction of $^{233}\text{U}$ (20%), the reactor to be in critical condition ($k_{\text{eff}} > 1$). The optimum subcritical condition for SAMOP Mod-1 can be achieved i.e. $k_{\text{eff}} = 0.97706 \pm 0.00250$ at the fraction of $^{233}\text{U}$ mass of 16%, with the CR = 0.1175.

In SAMOP Mod-2, a variation of fuel composition was performed. At the center, filled with a solution of thorium nitrate mixed with $^{233}\text{U}$ nitrate by 14% and in the annulus section filled by thorium nitrate solution with the addition of a mass fraction of $^{235}\text{U}$ of 10%. The calculation and analysis results on SAMOP Mod-2 shown that the $k_{\text{eff}} = 0.98113 \pm 0.00216$, but this value is too close to the critical value. The $k_{\text{eff}}$ of SAMOP Mod-2 is greater compared to SAMOP Mod-1 and SAMOP based on the patent. This is because the enlarging of the fuel column diameter for the aqueous solution thus increasing the chance for fission reactions to occur. The increase in $k_{\text{eff}}$ is also followed by an increase in CR of 0.1704. The increasing of CR is caused by the fact that $^{233}\text{U}$ mass fraction required to reach the $k_{\text{eff}}$ is smaller, so that the fertile material on the fuel of SAMOP Mod-2 much more than the SAMOP Mod-1. The more fertile material in the annulus and center increases the chance of neutron fission reaction from $^{235}\text{U}$ fissile material in UZrH fuels and $^{233}\text{U}$ in thorium nitrate solution so that resulting in CR of greater than SAMOP BATAN (patented design) and SAMOP Mod-1. The $k_{\text{eff}}$ and
CR values for SAMOP BATAN, SAMOP Mod-1 and SAMOP Mod-2 is shown in table 2, where this result is in accordance with similar calculation reported by Syarip [13].

![Graph showing k$_{eff}$ values as function of mass fraction of $^{233}$U in thorium nitrate for the model of SAMOP BATAN (patented) and SAMOP Mod-1.](image)

**Table 2.** Neutron multiplication ($k_{eff}$) and CR of SAMOP BATAN, SAMOP Mod-1 and SAMOP Mod-2.

| Model       | $k_{eff}$                  | CR    |
|-------------|----------------------------|-------|
| SAMOP BATAN | 0.89787 ± 0.00269          | 0.0871|
| SAMOP Mod-1 | 0.97706 ± 0.00250          | 0.1175|
| SAMOP Mod-2 | 0.98113 ± 0.00216          | 0.1704|

5. Conclusion
The SAMOP reactor model based on BATAN patent can not be fueled using thorium nitrate because of resulting in smaller neutron multiplication factor ($k_{eff}$). The $k_{eff}$ obtained for SAMOP re-design i.e. SAMOP Mod-1 is 0.97706 ± 0.00287 with the CR of 0.1175. The redesign result of SAMOP Mod-2 give a better $k_{eff}$CR values i.e. $k_{eff}$ = 0.98113 ± 0.00216 and CR = 0.1704 but it is not recommended to be implemented because it is too close to the critical value. The re-design of SAMOP still need to be developed further in order to achieve a breeder reactor with operating under subcritical condition.

Acknowledgement
This work was part of design assessment activities, related to the neutronic aspect of the SAMOP (Subcritical Assembly for Mo$^{99}$ Production) system, INSINAS Project No.: RT-2016-0151 titled “Production technology development and application of $^{99}$Te for medical diagnoses”, funded by Ministry of Research, Technology& Higher Education (Ristekdikti). The author would like to thank to the director of PSTA-BATAN Dr. Susilo Widodo and his staffs for supporting the implementation of the project.

References
[1] BATAN 2005 Perangkat Reaktor Subkritik untuk Memproduksi $^{99}$Mo (Jakarta: PATEN. Paten No. P-00200500760. DJKI)
[2] Hermawan D P, Rionaldy and Syarip S 2017 Neutronic Analysis of SAMOP Reactor Experimental Facility Using SCALE Code System (Presented at the International Conference on Computation in Science and Engineering, ITB)
[3] Syarip S, Sutondo T, Trijono EBS and Susiantini E 2018 Proceedings of the Pakistan Academy of Sciences: A. Physical and Computational Sciences 55 21
[4] Sutondo T, Syarip S and Santoso S 2009 Safety design limits of main components of the
proposed SAMOP system (Bandung: Proceedings of the 3rd Asian Physics Symposium (APS 2009))

[5] INTERNATIONAL ATOMIC ENERGY AGENCY 2005 Thorium fuel cycle—potential benefits and challenges (Google Scholar: Tech. Rep. IAEA-TECDOC-1450)

[6] Baldova D et al. 2011 J Radioanal Nucl Chem 288 37

[7] Ngadenin, Syaeful H, Widana K S dan Nurdin M 2014 Potensi Thorium dan Uranium di Kabupaten Bangka Barat (BATAN: Magazine of Eksplorium)

[8] Duderstadt J J, Hamilton L J 1976 Nuclear Reactor Analysis (New York: John Wiley & Sons, Inc.)

[9] Permana S, Takaki N and Sekimoto H 2011 Annals of Nuclear Energy 38 337

[10] Zsolt V, Nicholl A, Wallenius M and Mayer K 2012 Analytica Chimica Acta 718 25

[11] Kokubu Y S et al. 2012 International Journal of Mass Spectrometry 310 52

[12] Sutondo T and Syarip 2014 Ganendra Majalah Iptek Nuklir 17 83

[13] Syarip et al. 2018 J. Phys.: Conf. Ser. 978 012072