Analysis of Mo-99 production as function of CAMOLYP reactor power

D Bartolomeus¹*, S Syarip²

¹ Departments of Nuclear Engineering and Engineering Physics of Universitas Gajah Mada Jalan Grafika 2 Yogyakarta 55281, Indonesia
² Center for Accelerator Science and Technology, National Nuclear Energy Agency, Jalan Babarsari, Yogyakarta, Indonesia

*thephitonthel@gmail.com

Abstract. CAMOLYP or critical assembly for molly production is a nuclear reactor based on mixed uranium thorium fuels, which is currently being developed in BATAN Yogyakarta. This study aims to know how much the amount of ⁹⁹Mo isotope production can be achieved on 6 days reactor operation as function of different power levels. The method is a calculation of reactor physics simulation by using MCNPX computer code. The calculation is done for the reactor core configuration of 23 thorium dioxide fuel rods in the outer ring with the total mass of 73.12 kg and 1.706 kg of ²³⁵U in the annulus. The analysis result shows that the CAMOLYP’s ⁹⁹Mo production can be formulated as linear function of the reactor power level in watt. From the analysis result, it can be concluded that the CAMOLYP reactor can produce 39.91 curie ⁹⁹Mo at 1000 watts and 6 days operations. This analysis result indicates that the CAMOLYP reactor as a low power reactor has succeeded in producing ⁹⁹Mo isotope which is sufficient for domestic needs.

1. Introduction

Radioisotope molybdenum-99 (⁹⁹Mo) is one of the most commonly used nuclear medicine in the world. It is used as a parent nuclide to produce technetium-99m (⁹⁹mTc) for diagnostic procedures. About 85% of medical diagnostic procedures are using ⁹⁹mTc as a radioactive tracer, or about 20 million procedures each year. Radioisotope ⁹⁹mTc emits detectable gamma rays with energy of 140 keV with 8.8 pm wavelength. It is about the same wavelength emitted by commonly used X-ray devices. The half-life of ⁹⁹mTc is relatively low, about 6.0058 hours, which means it will decay into less than 10% under 24 hours. This relatively low half-life allows it to be used rapidly for scanning procedures, but keep the total dose of the patients relatively low [1,2].

A shortage of ⁹⁹mTc began to emerge in the late 2008s because two aging nuclear reactors, National Research Universal (NRU) and Petten Nuclear Reactor (HFR), were shut down for maintenance. Those two reactors provide about 66% of the world’s supply of ⁹⁹Mo [3,4]. In May 2009, a small leak of heavy water has been detected in the NRU reactor, causing it to be out of service until the completion of repairment in August 2010. And then, in August 2008, a formation of gas bubble jets at HFR was detected and the reactor was stopped for a safety investigation. And for safety reasons, 2 Canadian reactors constructed in 1990s were closed [5,6].
Because of the relatively low half-life, an unexpected shutdown of reactors are detrimental because each reactor has their specified regional market. For example, a shipment of $^{99}$Mo from Australia to Canada will make it unfavorable since the $^{99}$Mo will decays a lot along its shipment time.

Based on those facts, research toward reactor for radioisotope production are continuing. Recently, BATAN has developed SAMOP reactor for $^{99}$Mo production [7,8,9,10]. The study of using thorium-based fuel for SAMOP reactor has also been studied [11,12,13]. BATAN continued its research by developing a more sophisticated reactor called CAMOLYP, which is designed to be operated in a critical condition and utilizing thorium to conserve its uranium fuel. The neutronic calculation for the first design proposed by BATAN shows that it could not achieved criticality so a study has been done for the optimization of number of fuels, reflector width, and uranium density [14]. Later in this year, two CAMOLYP’s designs were proposed by BATAN, and the neutronic calculation shows that its design could achieve criticality condition [15].

One of the most important aspect of isotope production reactor such as CAMOLYP is the radioisotopes amount that can be produced. Therefore, a study needs to be done in order calculate the amount of $^{99}$Mo isotope production of CAMOLYP.

2. Method

This method relies deeply into Monte Carlo N-Particle computer program for calculating the isotope production of one of the two design proposed by BATAN. Monte Carlo Method is a type of computation that relies on random sampling to achieve a numerical result. The concept that underlies this method is to use randomness to solve a deterministic problem. By using the law of large number, integral that is described by a value of random variable can be approximated by taking the value of its sample mean of its independent sample. For example, to determine the diffusion of neutron in materials, the Monte Carlo Method is used to calculate samples of particle that is close to the actual value. This method was later developed into Monte Carlo N-Particle to calculate nuclear particles.

Monte Carlo N-Particle Transport Code, or MCNP, is a software to simulate a nuclear process, developed by Los Alamos National Laboratory (LANL) since 1957 [16,17]. It is distributed by the United States of America under the organization of Radiation Safety Information Computational Center in Oak Ridge, and internationally distributed by Nuclear Energy Agency in Paris, France. While MCNP is usually used to simulate nuclear processes, it has the capability to simulate particle interaction of neutron, photon, and electron.

This paper will use MCNPX, a version of MCNP program that is capable to simulate more than 34 particle interactions and more than 2000 heavy ions in a vast range of energy spectrum. This code can determine whether a reactor system is critical, and calculate their isotope production in a given power and time.

This CAMOLYP design uses uranyl nitrate (UN) at the inner tube and at the annular core, and thorium dioxide ($\text{ThO}_2$) at the outer ring. Both specification of the uranyl nitrate and thorium dioxide ($\text{ThO}_2$) is shown at Table 1 [14,15]. The calculation is done for the reactor core configuration of 23 thorium dioxide fuel rods in the outer ring with the total mass of 73.12 kg and 1.706 kg of $^{235}$U in the annulus.

The CAMOLYP’s design is visualised by Visual Editor program, as shown in figure 1. The atomic density of each atom is calculated using the same formula for calculating the uranyl-nitrate density of SAMOP [18]. A burn up code is then inserted into MCNPX program to calculate the isotope production in a given power level function: 1 W, 10 W, 50 W, 100 W, 500 W, 1000 W. It is performed in a one 6-day operation for each variation of power level. The hypothesis is that the production will increase linearly as the power level increases. Although the program will generate enough information of every isotope production, the paper will only focus to determine the $^{99}$Mo production at every power levels. The production of every isotopes will be analyzed only at 1000 W power level, as the CAMOLYP operates mainly on 1000 W.
### Table 1. Design specification of CAMOYLP

| Parameter                          | Material/ Value                  |
|------------------------------------|----------------------------------|
| Fuel in inner tube/annular core    | Uranyl Nitrate (UN)              |
| Diameter of inner tube/annular core| 5 cm / 31.4 cm                   |
| Height of inner tube/annular core  | 38 cm / 38 cm                    |
| Maximum number of fuel rods        | 23                               |
| Fuel in the ring                   | Th-U Nitrate                     |
| Reflector material                 | Graphite                         |
| Moderator material                 | Demineralized water              |
| Density of U                       | 300 g U/L                        |
| Enrichment of $^{235}$U in Uranyl Nitrate | 19.75%                         |
| Atomic density of $^{235}$U in Uranyl Nitrate | $1.50227 \times 10^{23}$ atom/L  |
| Atomic density of $^{238}$U in Uranyl Nitrate | $6.10416 \times 10^{23}$ atom/L  |
| Atomic density of N in Uranyl Nitrate | $1.52129 \times 10^{24}$ atom/L  |
| Atomic density of H in Uranyl Nitrate | $5.48299 \times 10^{25}$ atom/L  |
| Atomic density of O in Uranyl Nitrate | $3.50010 \times 10^{25}$ atom/L  |
| Atomic density of Th in Thorium Dioxide | $2.28106 \times 10^{25}$ atom/L  |
| Atomic density of O Thorium Dioxide   | $4.56212 \times 10^{25}$ atom/L  |

![Figure 1: CAMOLYP's geometry design](image)

3. Result and Analysis

A burn up calculation of different power level function has been done. The result is shown as figure 2. The result shows that the previous hypothesis is correct: the production will increase linearly as the power level increases. This is because as the power level increase, the neutron flux will increase. Thus, the fission reaction occurs more often. As a result, the $^{99}$Mo production will increase as a consequence of the fission of $^{235}$U fissile material.

By using linear regression method, the $^{99}$Mo isotope production of CAMOLYP can be formulated as: $Y = 0.0399X + 0.0049$, where $Y$ is the $^{99}$Mo production in 6-days Ci and $X$ is the power level of CAMOLYP reactor in watt. This can be used to predict the production at higher power level, such as 2000 W. But one should note the negative coefficient feedback of CAMOLYP reactor, because as the power level increase, the temperature will increase. Thus, it can lead to a decrease of $k_{eff}$, making the
reactor in a subcritical condition. The average $^{99}$Mo isotope production of CAMOLYP operated at 1000 W power level (considered as low power reactor) is 39.9 Ci per batch. This production rate is sufficient for domestic needs where recently the $^{99}$Mo is about 4 Ci per week [19].

![Figure 2](image-url)

**Figure 2**: The $^{99}$Mo isotope production on different power level of CAMOLYP reactor

As CAMOLYP normally operates in 1000 W, it’s important to identify the other isotopes production in that power level. The analysis of isotopes production has been done represented in activity (Ci) and mass ($\mu$g), and the results are shown in figure 3 and figure 4 respectively.

![Figure 3](image-url)

**Figure 3**: Isotopes production of CAMOLYP represented in activity (Ci)

Isotopes from fission product with high activity tend to have low mass. It can be seen from figure 5 that the production of isotopes measured by weight of mass and their activities are inversely proportional. This is happened as a consequence of radioactive decay. As the isotopes decay and emits radiation, it loses its mass by decaying into another isotopes. Thus, the faster it decays and emits radiation, the faster it loses its mass. It can be further explained by a mathematical model, $\lambda \Delta t = - \frac{dN}{N}$ [20]. The negative sign in the equation indicates that the mass (N) decreases over time. But note that not
all isotopes follows this tendency, such as $^{149}$Pm and $^{140}$La. This is because those two isotopes have relatively low probability of fission yield [21].

**Figure 4.** Isotopes production of CAMOLYP represented in mass (μg)

**Figure 5.** Comparison of mass and activity of the production of isotopes

4. **Conclusions**
The analysis of $^{99}$Mo isotope production of CAMOLYP reactor using MCNPX computer code has been done. The production can be formulated as: $Y = 0.0399X + 0.0049$, where $Y$ is the $^{99}$Mo production in 6-days Ci and $X$ is the power level of CAMOLYP reactor in watt. At 1000 W reactor power level, CAMOLYP can produce 39.9 Ci of $^{99}$Mo radioisotope, which is sufficient for domestic needs.

5. **Acknowledgement**
We would like to thank to Sekretariat Program Insinas 2019 Kemenristekdikti for funding this research (06/INS-1/PPK/E4/2019)
References

[1] International Atomic Energy Agency. 2008 Homogeneous Aqueous Solution Nuclear Reactors for the Production of Mo-99 and Other Short-Lived Radioisotopes. IAEA TECDOC Report 1601. Vienna.

[2] Ruth, Thomas. Nature 457, 536.

[3] Thomas G S, Maddahi J. 2010 Journal of Nuclear Cardiology 17, 993.

[4] Thomas J, Ruth. 2014 J. Nucl. Med. Technol. 42, 245.

[5] Pillai M R, Dash A, Knapp F Jr. 2013 J Nucl Med. 54, 313.

[6] Amanda J Youker, Sergey D Chemerisov, Michael Kalensky, Peter Tkac, Delbert L and George F. 2013 A Solution-based approach for Mo-99 production: considerations for nitrate versus sulfate media (Hindawi Publishing Corporation, Science and Technology of Nuclear Installations) Vol 2013, Article ID 402570.

[7] Syarip S, Tegas S, Edi Trijono B, and Endang S. 2018 Proceedings of the Pakistan Academy of Sciences: A. Physical and Computational Sciences 55, p21.

[8] M. Iqbal Farezza W and Syarip. 2018 J. Phys.: Conf. Ser. 1090, 012013.

[9] Syarip, P I Wahyono, W Susilo and K Donny. 2019 J. Phys.: Conf. Ser. 1198, 022023.

[10] Dedy P Hermawan, Rionaldy and Syarip S. 2018 J. Phys.: Conf. Ser. 1090, 012032.

[11] Syarip, E Togatoropor. 2018 J. Phys.: Conf. Ser. 978, 012072.

[12] F Yassar, A W Harto and Syarip. 2018 J. Phys.: Conf. Ser. 1080, 012021.

[13] Syarip, Susilo Widodo, Muzakky and Sukirno. 2019 J. Phys.: Conf. Ser. 1204, 012005.

[14] Delphito B N, Syarip. 2019 “Kajian Awal Analisis Neutronik Reaktor Produksi Isotop Molly Berbasis Thorium” Paper presented at Seminar Keselamatan Nuklir Nasional (SKN).

[15] Delphito B N, Syarip. 2019 “Neutronic Analysis of Critical Assembly for Moly-99 Production Reactor Based on Mixed Th-U Fuels”. Paper presented at The International Conference of Nuclear Capacity Building, Education, Research and Applications (I-CONCERN).

[16] Cashwell E D, Everett C J. 1959 A Practical Manual on the Monte Carlo Method for Random Walk Problems (London: Pergamon Press.)

[17] Pelowitz D B. 2008 MCNPX User’s Manual Version 2.6.0 (New Mexico: LANL).

[18] Syarip and Tegas Sutondo. 2018 J. Phys.: Conf. Ser. 1090, 012036.

[19] Yudiutomo Imarjoko. 2019 Prospek Mo-99 di Indonesia. Paper presented at the SAMOP Workshop.

[20] Patel S B. 2000 Nuclear physics: an introduction (New Delhi: New Age International) pp. 62–72.

[21] "Cumulative Fission Yields". www-nds.iaea.org. IAEA.