Mo-99 Isotope Production Calculation of SAMOP Reactor Experimental Facility

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Abstract. The calculation of $^{99}$Mo isotope production of SAMOP (Subcritical Assembly for $^{99}$Mo Production) experimental test facility using SRAC computer code has been done. The SAMOP experimental test facility is a reactor in subcritical condition fueled with low-enriched uranium in the form of uranyl nitrate solution, with uranium concentration of 300 g U/L, and driven by an external neutron source. The calculation method is using Standard Reactor Analysis Code (SRAC) which is a computer code package system applicable to neutronic analysis for various types and forms of the nuclear reactor core. The calculation results show that the neutron multiplication factor of 0.981 to 0.992 can be achieved with 30.2989 L to 31.4587 L uranyl nitrate in the certain core configuration of SAMOP test reactor. The $^{99}$Mo isotope production calculation for 6-day operation (batch) shows that the SAMOP test facility can produce approximately 437.03 mCi (16170.11 MBq) per batch, and the $^{99}$Mo production will reach a saturated condition at twelve day operation.

Keywords: $^{99}$Mo, isotope production, SAMOP, SRAC, neutron multiplication, uranyl nitrate.

1. Introduction
The need of radioisotopes for nuclear medicine in Indonesia has grown rapidly in recent years. One of the most common aspects of nuclear medicine is diagnostics is using the $^{99m}$Tc radioisotope. The increased need for the $^{99m}$Tc isotope cannot be fulfilled because $^{99}$Mo commonly used as a $^{99m}$Tc generator, can only be produced in a nuclear reactor and the half-life of $^{99}$Mo is only 66 hours making it difficult to store and mobilize its spread. Solving this problem can be done by shortening the $^{99}$Mo production process as well as shortening the mobilization of radioisotopes spreading distance.

Over 10000 hospitals worldwide use radioisotopes in medicine, and about 90 % of the procedures are for diagnosis. The most common radioisotope used in diagnosis is $^{99m}$Tc which is daughter isotope of $^{99}$Mo, accounting for 80 % of all nuclear medicine procedures worldwide [1, 2]. Commonly, most $^{99}$Mo is produced using highly-enriched uranium placed in high-power nuclear research reactors. Recently there are eight medical isotopes producing reactors that provide over 90 % of the world’s $^{99}$Mo needs. These reactors are government-owned and -subsidized, and the respective governments control the funding, priorities, and operational schedule of these reactors. The unscheduled shutdown of two of these reactors in 2009 and 2010 (Canadian and Dutch reactors)
caused worldwide shortages of $^{99}$Mo, leading to the delay or cancellation of many medical procedures [3, 4].

The medical community has been plagued by $^{99}$Mo shortages due to aging reactors, such as the NRU (National Research Universal) reactor in Canada. There are currently no US producers of $^{99}$Mo, and NRU was being shut-down in 2016, which means that another $^{99}$Mo shortage is imminent unless a potential domestic $^{99}$Mo producer fills the void [5]. The $^{99}$Mo producing system without a nuclear reactor and without using highly-enriched uranium is being implemented at National Nuclear Energy Agency (BATAN). Its production process will use a subcritical assembly driven by an external neutron source from neutron generator or a particle accelerator and target that generate neutron in the core of subcritical assembly. Compact neutron generator (CNG) is a particle accelerator, as predicted that the accelerators will probably the best-known uses are for cancer therapy, medical isotope production, and food irradiation [6]. The similar method is being developed by SHINE which plans to produce at least one-half of the U.S. need for $^{99}$Mo by 2016 [7].

In general, the production of $^{99}$Mo is by splitting the $^{235}$U atom with neutrons inside a nuclear reactor so that a critical reactor is required. Licensing issues for critical reactors are also complex because they have to fulfill the very strict technical and safety requirements, and it needs huge funds as well. To solve this problem BATAN or the National Nuclear Energy Agency designed a reactor that could produce $^{99}$Mo but did not categorized as the critical reactor and did not use high-enriched uranium. A non-critical reactor or subcritical assembly fueled with uranyl nitrate, called subcritical assembly for $^{99}$Mo production (SAMOP) has been designed and will be developed further at the Center for Accelerator Science and Technology (CAST) BATAN [8]. The SAMOP experimental facility as a test facility which use an external neutron source from the beam-port of Kartini TRIGA reactor, which has been identified suitable for this purpose [9]. It is expected that the SAMOP system will reduce much less waste than current $^{99}$Mo production methods.

The aim of the research is the calculation and determination of $^{99}$Mo specific activities, and determination of optimal irradiation conditions, as well as the effect of some variables of parameters will affect the $^{99}$Mo production of the SAMOP reactor. The $^{239}$Pu byproducts will also be analyzed considering $^{239}$Pu is a fissile material so that the SAMOP reactor can later meet the safeguard requirements. The isotopes calculation will be approximated by the deterministic method applied in Cell Burn of the SRAC computer code.

The use of subcritical aqueous homogenous reactors driven by accelerators presents an attractive alternative for producing $^{99}$Mo. In this method, the medical isotope production system itself is used to extract $^{99}$Mo or other radioisotopes so that there is no need to irradiate common targets. In addition, it can operate at much lower power compared to a traditional reactor to produce the same amount of $^{99}$Mo by irradiating targets [10]. Study to produce $^{99}$Mo with activation method by using TRIGA reactor have also been done to fulfill the need of $^{99}$Mo as $^{99}$m Tc generator for use in nuclear medicine [11,12,13]

2. Description of SAMOP experimental facility

The SAMOP experimental facility as a test facility which use an external neutron source emerge from the radial beam-port of Kartini reactor. The external neutron source has been identified as thermal neutron in order of $10^8$ n/cm$^2$ s [9]. The SAMOP core consists of annular cylindrical tube containing uranyl nitrate [$^{235}$UO$_2$ (NO$_3$)$_2$] as fuels and target, surrounded by ring of UO$_2$ (NO$_3$)$_2$ tubes or TRIGA fuel elements. The TRIGA fuel elements can be loaded in the ring together with UO$_2$ (NO$_3$)$_2$ tubes to increase neutron multiplication factor.

The dimension of the SAMOP core, reflector, boral rod neutron absorber, and coolant tank is described in Figure 1. The SAMOP experimental facility is provided by an instrumentation and control system in such that there is a criticality indication, the boral control rod neutron absorber will dropped automatically inserted to the SAMOP reactor core.
3. Materials and methods

3.1. $^{99}$Mo Production methods

In general, the production of $^{99}$Mo is by splitting the $^{235}$U atom with neutrons inside a nuclear reactor so that a critical reactor is required. There are two primary approaches for producing the medical isotope $^{99}$Mo i.e.: fission of $^{235}$U or $^{235}$U(n,f)$^{99}$Mo, which produces $^{99}$Mo and other medically important isotopes such as $^{131}$I and $^{133}$Xe, and neutron capture by $^{98}$Mo or $^{98}$Mo(n,γ)$^{99}$Mo [14]. The fission of $^{235}$U produces a large number of fission products, including $^{99}$Mo. The global producers of $^{99}$Mo use the fission of $^{235}$U method instead of neutron capture method to produce $^{99}$Mo because of its inefficiencies. However, the neutron capture process can be used to make smaller quantities of $^{99}$Mo. Table 1 compares the two methods of $^{99}$Mo production [15].

|                  | $^{235}$U(n,f)$^{99}$Mo | $^{98}$Mo(n,γ)$^{99}$Mo |
|------------------|-------------------------|--------------------------|
| Produces high specific activity $^{99}$Mo | Produces low specific activity $^{99}$Mo |
| Requires enriched $^{235}$U target | Requires highly enriched $^{98}$Mo target |
| Complex chemical processing | Simple chemical processing |
| Requires dedicated processing facility | Requires high flux neutron source |
| Generates high-level radioactive waste | Generates minimal waste |

The isotope production rate, which is of interest here, is proportional with several conditions as illustrated in the equation (1) below:

$$ R \approx n \sigma \phi $$

(1)
where: $R$ is a rate of reaction (i.e., number of reactions per unit time and volume), which is related to the amount of the new substance that can be produced, $n$ is the number of target nuclei present (i.e., the target nuclei density in atoms per unit volume), $\phi$ is the flux of particles causing the reaction (neutrons per cm$^2$ per second), and $\sigma$ is the probability that the reaction will occur, expressed as an area. The calculation steps by using SRAC computer code is described in Figure 2. The analysis of $^{99}$Mo production is done as a function of uranyl nitrate enrichment and SAMOP reactor power, at a subcritical conditions or effective neutron multiplication factor ($k_{ef}$) small than 1 (0.9 to 0.99).

![Figure 2. The $^{99}$Mo calculation steps by using SRAC computer code.](image)

### 3.2. SRAC computer code

The SRAC (Standard Reactor Analysis Code) is a set of FORTRAN computer codes created by Japan Atomic Energy Research Institute (JAERI) to analyze and solve the neutronic calculations of various models of thermal reactors [16]. This code can solve the microscopic and macroscopic cross-sections effectively to complete burnup calculations. In core reactor operations, the magnitudes of fission such as neutron flux, nuclide concentration, and temperature will vary as spatial functions. Burnup calculations must be performed to see the correlation of these parameters with time. In the SRAC system, the reactor core burn-up calculations are divided into two stages. First, calculate cell-level burn-up using SRAC to obtain a homogeneous macroscopic cross-section on each burn-up step. The latitude is then tabulated with discrete values of fuel temperature, coolant, and degree of burn-up to model the actual fuel assembly in the reactor core. The next stage is to interpolate the existing data to be distributed in the reactor core for further processing.

Cell Burn Up is an additional code SRAC system that serves to complete the calculation of reactor core burn-up with the theory of neutron diffusion. In the calculation of burn-up cell, the burnup changes of the atomic number density of the nuclides in the burn-up chain reaction model are obtained from solving the depletion equation with the reaction rate of fission, capture, and reaction (n, 2n) [16]. Here is an optional treatment that can be used in cell burn-up calculations: option to calculate the cooling time when burn-up, option to calculate the conversion of the instant or integration ratio defined by the user, burn-up calculations with constant flux levels, and burn-up calculations with fixed density numbers for specific nuclides. Several models of burn-up chains are available to choose for use in calculations [16].

In the cell burnup calculations, there are two types of time-step units adopted in the SRAC code. First is the burnup step unit with relatively common time intervals, and the second is the sub-step unit in each burnup step. When the burn-up step interval is described in the input, the sub-step interval is specified in the code. At each burn-up step, the flux calculations are performed by the
selected code. As a result, the distribution of one group collapsed flux and collapsed microscopic cross-sectional for burnable nuclides is obtained. The depletion equation for the interval from the burnup step to \( n (t_{b1}, t_{b2}) \) can be written by equation (2), on the assumption that the relative distribution of microscopic reaction rates for capture, fission, and reaction \((n, 2n)\) does not change in time interval step burn-up.

\[
\frac{dN_i^M(t)}{dt} = \sum_{j \neq 1} f_{j \rightarrow 1} \lambda_j N_i^M(t) + \sum_{k \neq 1} \left\{ g_{k \rightarrow 1} \sigma_{c,j}^M + \gamma_{k \rightarrow 1} \sigma_{f,j}^M \right\} \phi^M \text{Fact}(t) N_k^M(t) - \\
\left[ \lambda_i + (a_{a,i}^M + a_{n2i,i}^M) \phi^M \text{Fact}(t) \right] N_i^M(t)
\]

(2)

\( i, j, k \) : the depleted nuclides, \( M \) : depletion zone, associated with M-region in SRAC code, \( N \) : atomic number density, \( \lambda \), \( f \) : decay constant and branching ratio, \( \gamma \), \( g \), \( h \) : the yield fraction of each transmutation, \( \phi \) : the relative flux obtained from the eigenvalue calculation, and \( \text{Fact}(t) \) : the normalization factor for converting the relative flux to absolute flux.

4. Result and discussion

The first analysis was to look at how the influence of uranium enrichment variation of the SAMOP reactor to the sub-criticality value. From the results of the first analysis will get the relationship between the criticality levels with the uranium enrichment, then selected the value of enrichment that the criticality level is closest to 1 (subcritical). Once the enrichment value is known, Cell Burn is done to determine the amount of \(^{99}\text{Mo}\) produced on the SAMOP reactor. The result of cell burn program code will show the relationship between enrichment with \(^{99}\text{Mo}\) result. The running result of this program code can also be obtained plutonium production value which is expected still within the safe limits. The second analysis is to vary the power on the SAMOP reactor with variations of 1 kW, 10 kW, 50 kW and 100 kW. The results of this simulation will be found reactor power relationship with \(^{99}\text{Mo}\) results generated each increase of 1 kW of power. The third analysis was to run a SAMOP reactor for 24 days to get \(^{99}\text{Mo}\) of growth and the optimal operating cycle. The effective multiplication factor \((k_{ef})\) of the SAMOP reactor was calculated using the SRAC-CITATION code for variation of uranium enrichment, the result is obtained as shown in Figure 3.

![Figure 3](image-url)

**Figure 3.** The \( k_{ef} \) of the SAMOP reactor experimental facility with variation of fuel enrichment

By using 19.5 % of \(^{235}\text{U}\) fuel enrichment, the criticality or neutron multiplication factor was also done for two configurations of SAMOP reactor core i.e. for the core with annular tube surrounded by 8 tubes of \( \text{UO}_2 (\text{NO}_3)_2 \) and 12 tubes in the ring respectively. The criticality calculation was done to investigate the change of sub-criticality levels per day of reactor operation, the result is depicted in Figure 4. It is shown that within ten days operation (for \(^{99}\text{Mo}\) production), the sub-criticality level of both SAMOP core configurations is still stable. The calculation results show that the neutron multiplication factor of 0.981 to 0.992 can be achieved with 30.2989 L to 31.4587 L uranyl nitrate in that core configurations. The criticality analysis result was in accordance with refer to similar work [17], and similar work previously done for SAMOP criticality analysis [18,19].

The \(^{99}\text{Mo}\) production calculation for 6 days operation (one batch) was done by Cell Burn as function of \(^{235}\text{U}\) enrichment, and the result is described in Figure 5. It can be seen that the \(^{99}\text{Mo}\) production value is in the range of 437.03 mCi, and increasing as uranium enrichment increases. This
increase is due to the fact that $^{99}$Mo is a fission product of $^{235}$U, so that if the value $^{235}$U in fuel increases, the $^{99}$Mo will also increase. This can be seen from the $^{99}$Mo formation scheme in the SRAC manual as seen in Figure 10. The average $^{99}$Mo production calculation result is in line with the $^{99}$Mo production forecast target for the first 300-480 mCi model for each production cycle of 6 days.

![Figure 4](image1.png) **Figure 4.** The sub-criticality levels per day of SAMOP experimental reactor operation

![Figure 5](image2.png) **Figure 5.** $^{99}$Mo production for 6 days operation (per batch) as function of $^{235}$U enrichment

The output from the Cell Burn is also obtained the $^{239}$Pu isotope produced by SAMOP reactor which is very small such as described in Figure 6. It is shown in Figure 6 that $^{239}$Pu production in a batch around 0.0207 – 0.031 mg, and the curve shows a decreasing trend in plutonium yields for the increase in $^{235}$U enrichment. This is occurred because the increasing in enrichment means decreasing the amount of $^{238}$U so that $^{239}$Pu which is the result of transmutation of $^{238}$U will also decrease.

![Figure 6](image3.png) **Figure 6.** $^{239}$Pu isotope produced by SAMOP reactor as function of $^{235}$U enrichment

![Figure 7](image4.png) **Figure 7.** Production of $^{99}$Mo as function of SAMOP operation time

The calculation of $^{99}$Mo production is also conducted for operating time until 24 days, the result shows that the $^{99}$Mo yield reaches saturation value on the 12th day such as depicted in Figure 7. It can be seen that the growth of $^{99}$Mo on the first day to day 6 amounted to 437.03 mCi or 16170.11 MBq. While from day 6 to day 12 only amounted to 102.23 mCi so it can be concluded that 6-day operating cycle time is more effective. Therefore, the operation of the SAMOP reactor is determined for 6 days in consideration that 6-day operating time was considered more economical, this is in line with the commonly $^{99}$Mo production [14,15].

5. Conclusion

The $^{99}$Mo average production of SAMOP experimental facility is 437.03 mCi or 16170.11 MBq per batch (6 days operation). The amount of $^{99}$Mo produced continues to increase until the 12th day of SAMOP operation, where the saturation of $^{99}$Mo production is achieved. The SAMOP system operate at subcritical condition with the neutron multiplication factor of 0.981 to 0.992 with 30.2989 L to 31.4587 L uranyl nitrate in the reactor core.
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