Neutronic Analysis of SAMOP Reactor Experimental Facility Using SCALE Code System

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Abstract. The Subcritical Assembly for ⁹⁹Mo Production (SAMOP) reactor is considered a suitable candidate to produce ⁹⁹mTc isotope on a small scale. The design of SAMOP reactor using uranyl nitrate (UN) as fuel has been studied at National Nuclear Energy Agency. The neutronic analysis of the SAMOP Experimental Facility will be reported in this paper. The SAMOP reactor core design consists of an annular tube containing UN surrounded by the ring of UN tubes, and a fuel tube absorber follower is inserted in the center of the core. The work covers the criticality calculation to secure the sub-criticality level and neutron flux distribution analysis. The computational method is using SCALE computer code. The analysis result shows that for the required neutron multiplication factor of 0.98 to 0.99 (sub-criticality level) can be achieved by a combination of fuel (UN) height in the annular tube of 38 cm surrounded by 5 – 8 of UN tubes in the ring. For the UN height in the annular tube of 39 cm, it should be surrounded by 3 – 6 of UN tubes in the ring.

Keywords: ⁹⁹Mo, SAMOP, uranyl nitrate, SCALE code, reactor, criticality calculation.

1. Introduction

Technetium-99m (⁹⁹mTc) is the most commonly used isotope for medical purposes but since ⁹⁹mTc has a short half-life of 6.0058 hours, a stock pile cannot be produced. Therefore the parent isotope of ⁹⁹mTc, which is ⁹⁹Mo, is delivered to the medical centers so when ⁹⁹mTc isotope is needed, it can be extracted from ⁹⁹Mo. This extraction process called ⁹⁹mTc generator [1,2].

The current conventional method to produce ⁹⁹Mo is by irradiating low enriched uranium (LEU) or high enriched uranium (HEU) targets in a high neutron flux environment such as in a nuclear reactor for a couple of days. After irradiation or fission process, the targets are brought to the reprocessing facility to extract ⁹⁹Mo and then prepared to be shipped to the medical centers. In the fission method, the fission yield of ⁹⁹Mo is about 6.1%, so a larger quantity of ⁹⁹Mo produced could be reached with this process. However the fission method required considerable capital investment and produced large quantities of radioactive waste [2].

The other method is by irradiating ⁹⁸Mo with neutrons to convert natural ⁹⁸Mo with neutron capture reaction and became ⁹⁹Mo, but this process produced much lower ⁹⁹Mo than fission method using HEU or LEU targets. It’s because the cross section of ⁹⁸Mo (n,γ) ⁹⁹Mo reaction is small (σ ≈ 0.14 barns),
resulting a very small quantity of $^{98}$Mo is converted to $^{99}$Mo. In order to produce high quality $^{99m}$Tc generators from capture reaction method, advanced generator technologies are required [2,14,15].

The demand for $^{99}$Mo as medical isotope will continue to increase in the future. Therefore, a lot of research about optimizing the production of $^{99}$Mo and medical isotopes had been done. Ball R M describe that an Aqueous Homogeneous Reactor (AHR) fueled with uranyl nitrate could be used to produce medical isotopes [3]. Since then, the initiated program to assess the feasibility of utilizing AHRs for the production of medical isotopes in Russian, United States, Japan, France, Canada, China, Indonesia and other countries, have been done [4,5,6]. In Indonesia, The National Nuclear Energy Agency investigates a Subcritical Assembly for $^{99}$Mo Production (SAMOP) reactor based on uranyl nitrate solution [7,8].

An idea to design a facility dedicated to producing $^{99}$Mo radioisotope for medical application, named as a Sub-critical Assembly for $^{99}$Mo Production (SAMOP) had been launched few years ago [9,10]. The idea came up in conjunction with the rising problems related to the radioisotope production such as the international conversion program that would affect the high enriched uranium supply used in the current’s practice of the radioisotope production. Another problem related with the current’s method used in the production process of the medical radioisotopes is the inefficiency of the $^{235}$U utilization; i.e. around 1 %, and the rest containing high $^{235}$U concentration must be wasted and sent back to the original exporting country.

The proposed design of the SAMOP system was intended to solve the above two problems i.e. it was designed to use of low enriched uranyl nitrate (UN) solution, serving for both the reactor fuel and $^{99}$Mo producing source as well. Based on this concept, the irradiated uranium solution can be reused again after completing the extraction process and reconditioning process, then transferred back to the core tank for the next irradiation process and hence, improving the efficiency of the $^{235}$U utilization.

This paper presents a criticality calculation to secure the sub-criticality level the SAMOP experimental facility, by using SCALE computer code. The SCALE computer code system is a widely used modeling and simulation suite for nuclear safety analysis and design that is developed, maintained, tested, and managed by the Reactor and Nuclear Systems Division (RNSD) of the Oak Ridge National Laboratory (ORNL). SCALE provides a comprehensive, verified and validated, user-friendly tool set for criticality safety, reactor physics, radiation shielding, radioactive source term characterization, and sensitivity and uncertainty analysis [11,12]. The analysis was also intended to determine the allowable number of UN solution tubes or TRIGA fuels inserted into the ring or peripheral holes to keep the core subcritical at any condition. From the previous calculations, the preferable nominal uranium density is 300 g U/L [10].

2. SAMOP experimental facility
The schematic diagram of overall concept of SAMOP system is depicted in Figure 1.
The SAMOP core fuelled by uranyl nitrate solution \([\text{UO}_2 (\text{NO}_3)_2]\). As shown in Figure 1, the UN fuel solution is continuously pumped around. In the delay tank short-lived isotopes will decay. In the extraction column the \(^{99}\text{Mo}\) will be recovered. The recondition facility ensures a constant concentration of uranium and the solution tank used as a the buffer tank for the fuel.

The SAMOP experimental facility as a test facility for SAMOP design which use an external neutron source from the radial beam-port of Kartini reactor. The external neutron source has been identified as thermal neutron in order of \(10^8 \text{n/cm}^2 \text{s}\) [13]. Figure 2 shows the schematic layout of SAMOP experimental facility, and Figure 3 shows the core system. The SAMOP core consists of annular cylindrical tube containing uranyl nitrate \([\text{UO}_2 (\text{NO}_3)_2]\) as fuels and target, surrounded by ring of \(\text{UO}_2 (\text{NO}_3)_2\) tubes or TRIGA fuel elements. The TRIGA fuel elements can be loaded in the ring together with \(\text{UO}_2 (\text{NO}_3)_2\) tubes to increase neutron multiplication factor. The dimension of the SAMOP core, reflector, boral rod neutron absorber, and coolant tank is described in Table 1. The cooling system of SAMOP is using \(\text{H}_2\text{O}\) as its coolant and also as an axial radiation shielding. 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. The system shall ensure the sub-critical state at any condition with adequate margin.

*Figure 2. Layout of SAMOP experimental facility [13]*

*Figure 3. SAMOP reactor core*

| Component | Dimension |
|------------|-----------|
| Annular core: |         |
| - diameter | 32.4 cm   |
| - height   | 43 cm     |
| Ring of UN tubes: | 1.70 |
| - diameter of ring | 40.4 cm |
| - tube (fuel) diameter | 3.2 cm |
| Coolant tank: |         |
| - diameter | 120 cm    |
| - height   | 400 cm    |
| Neutron absorber tube: |         |
| - diameter | 3.2 cm    |
| - height   | 38 cm     |

Table 1. The dimension of SAMOP reactor
3. Methodology
The neutronic calculation in this work is done using SCALE (Standardized Computer Analyses for Licensing Evaluation) code system which developed and maintained by Oak Ridge National Laboratory (ORNL) under contract with the U.S. Nuclear Regulatory Commission (NRC) [11]. Within SCALE code system, module KENO IV was used to analyse the effective neutron multiplication factor (k$_{eff}$) of SAMOP reactor. KENO IV calculates k$_{eff}$ of a 3-D system using the Monte Carlo method. In KENO IV module, the neutron source, the material and geometry of SAMOP reactor is modeled.

The material composition of SAMOP reactor is given in the Table 2. Uranyl nitrate with uranium concentration of 300 gU/L and U$^{235}$ enrichment of 20% is used in this work. The specification of uranyl nitrate is given in Table 3.

| Table 2. Material composition of SAMOP reactor | Table 3. Material specification of uranyl nitrate |
|-----------------------------------------------|-----------------------------------------------|
| Material          | Density (g/cm$^3$)     | Nuclide | Atomic Density (atom/barn cm$^3$) |
| Uranyl nitrate    | 2.81                    | U-235   | 1.537245 x 10$^4$                   |
| SS304             | 7.94                    | U-238   | 6.07131 x 10$^4$                   |
| Dry air           | 1.205 x 10$^{-3}$      | H-1     | 5.48779 x 10$^2$                   |
| Graphite          | 1.70                    | N-14    | 1.521711 x 10$^3$                  |
| H$_2$O            | 9.982 x 10$^{-1}$      | O-16    | 2.743895 x 10$^2$                  |

SAMOP reactor core design consists of an annular tube containing uranyl nitrate fuel surrounded by the ring of uranyl nitrate tubes and a fuel tube absorber follower is inserted in the center of the core. The effective multiplication factor (k$_{eff}$) that required for this SAMOP design is between 0.98 – 0.99. In order to reach the required value of k$_{eff}$ for SAMOP reactor, uranyl nitrate tubes and fuel tube absorber follower are inserted one by one into the reactor. The fuel tube absorber follower is inserted first into the center of annulus then followed by 12 uranyl nitrate tubes into the ring. There are three variation of uranyl nitrate fuel’s height in both annulus and tubes are applied in this work. Those variation are 38 cm, 39 cm and 40 cm. This condition are applied to analyse the effect of fuel’s height in either annulus or fuel tube absorber follower to the value of k$_{eff}$.

The safety aspect also investigated in this work. In order to secure the value of k$_{eff}$ did not exceed 1.00, SAMOP reactor has a fuel tube absorber follower in the center of its the core. There are 2 variety of absorber material that investigated in this work. Those absorber material are B$_4$C (boron carbide) and Boral. To analyse the effect of each absorber material, the fuel tube absorber follower is inserted slowly from 0% to 100% of fuel tube absorber follower is inserted into the core.

4. Result and Discussion
The value of effective multiplication factor (k$_{eff}$) represented the criticality condition of the SAMOP reactor is calculated with SCALE code system using KENO IV module for different fuel’s height in either annulus and tubes. The first analysis was to investigate the influence of fuel’s height variation in annulus and tubes around the ring to k$_{eff}$ value. The results of this analysis will show the relationship between the criticality levels with the fuel’s height and we can determined how many fuel tubes needed to satisfy the required criticality of SAMOP reactor at specific fuel’s height. The geometrical core design model of SAMOP reactor with KENO IV can be seen in Figure 4 and Figure 5. The SCALE running results of the ring tubes fuel’s height variations at annulus fuel’s height 38 cm is shown in Figure 6.

The first analysis results shows that the variation of the ring tubes fuel’s height didn’t have significant influence toward the k$_{eff}$ value of the SAMOP reactor. In all of the fuel’s height variations in the ring tubes shows that to reach the value of required k$_{eff}$ for SAMOP reactor, the number of fuel tubes needed are between 4 to 10 tubes in the ring and one tube in the center of the core. In total, 5 – 8 uranyl nitrate tubes are needed to reach the required k$_{eff}$ value of SAMOP reactor.
Further SCALE running results for the annulus fuel’s height variations with the ring tubes fuel’s height at 38 cm is shown in Figure 7. The results shows that the variation of the annulus fuel’s height have significant influence toward the $k_{eff}$ value of SAMOP reactor. As the annulus fuel’s height increase, the $k_{eff}$ value also increasing significantly. Thus, the number of fuel tubes inserted into the core could be reduced. With the annulus fuel’s height 38 cm, fuel tubes that needed to reach the required $k_{eff}$ value for SAMOP reactor is between 5 – 8 tubes. At the height of 39 cm, it need between 3 – 6 tubes and at the height of 40 cm, it need 2 – 5 tubes to reach the required $k_{eff}$ of SAMOP reactor experimental facility. The results show that the increase of the annulus fuel’s height will significantly increasing the $k_{eff}$ of SAMOP reactor and also reduced fuel tubes that needed to reach the required $k_{eff}$ of SAMOP reactor experimental facility. The analysis result of criticality values ($k_{eff}$) is in accordance with the similar system conducted by other researchers [3,4]

The results will be useful for conducting UN fuel loading procedures, known as criticality or sub-criticality experiment, as well for deciding the optimum operation and production scenario of SAMOP reactor experimental facility in the future.

**Figure 4.** Axial cross section of SAMOP reactor design in KENO IV.

**Figure 5.** Radial cross section of SAMOP reactor design in KENO IV.

**Figure 6.** The $k_{eff}$ of the SAMOP reactor with variation of the ring tubes fuel’s height and annulus fuel’s height at 38 cm.

**Figure 7.** The $k_{eff}$ of the SAMOP reactor with variation of the annulus fuel’s height and the ring tubes fuel’s height at 38 cm.
The next analysis is to investigate boron carbide (B$_4$C) and boral as neutron absorber materials in SAMOP reactor experimental facility. The control rod as neutron absorber which is followed by UN tube or known as control rod fuel follower is located in the central of SAMOP reactor core (see Figure 4). The analysis was done to compare the effectivity between both materials to control the fission reaction. The comparison of absorber rod worth between B$_4$C and boral as the reactor control material is shown in Figure 8. This analysis result is also in a good agreement with the results for the similar system previously conducted by other researchers [7,8,9].

The result show that B$_4$C has lower reactivity than boral. When B$_4$C absorber rod is fully inserted into the core, it reactivity worth is around -4.4 %Δk/k, whereas boral’s rod reactivity worth is around -3.8 %Δk/k. It means that B$_4$C rod absorb more neutron than boral rod when it is inserted into the core, thus the value of $k_{\text{eff}}$ will be reduced more. This result will be useful for deciding which material is suitable for establishing the safety system of SAMOP reactor experimental facility in the future.

![Figure 8. The comparison of B$_4$C and boral neutron control rod worth.](image)

5. Conclusion
The required sub-criticality level i.e. neutron multiplication factor ($k_{\text{eff}}$) of 0.98 to 0.99 of SAMOP system can be achieved by a combination of uranyl nitrate (UN) fuel height in the annular core of 38 cm surrounded by 5 – 8 of UN tubes in the ring. For the UN height in the annular tube of 39 cm, it should be surrounded by 3 – 6 of UN tubes in the ring, and 2 – 5 UN tubes are needed when the height of UN in annular tube is 40 cm. The variations in the ring UN tubes fuel’s height of SAMOP reactor didn’t have significant effects toward the $k_{\text{eff}}$ value but the variations in the annulus fuel’s height did have a significant effect. For the neutron absorber rod, the B$_4$C material has a lower reactivity worth than boral. The reactivity worth is -4.4 %Δk/k when B$_4$C rod is fully inserted and -3.8 %Δk/k when boral rod is fully inserted into the core of SAMOP reactor.

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