Source Term Analysis of SAMOP Reactor Experimental Facility

Wahid L\textsuperscript{1,3}, M I Farezza W\textsuperscript{1,4}, Syarip\textsuperscript{2,5}

\textsuperscript{1}Department of Nuclear Engineering and Engineering Physics, University of Gadjah Mada, Yogyakarta
\textsuperscript{2}Centre for Accelerator Science & Technology, Jl. Babarsari, POB 6101 ykbb, Yogyakarta

Email: \textsuperscript{1}luthfi_doank@yahoo.com, \textsuperscript{4}ryakuzza@gmail.com, \textsuperscript{5}syarip@batan.go.id

Abstract. The Subcritical Assembly for Molybdenum-99 Production (SAMOP) system by using low enriched uranyl nitrate solution as reactor fuel is being developed and designed in the Centre for Accelerator Science & Technology, Yogyakarta. The source term analysis of the SAMOP experimental facility is done in conjunction with the radiation shielding design. The method used is by using MCNPX software for fuel burn-up calculations. The burnup calculation is done both in the reactor operation without \(^{99}\text{Mo}\) extraction and with 100\% \(^{99}\text{Mo}\) extraction per batch (6 day + 1 day reactor off) along 1 month operation (5 batch). The calculation was done for two conditions i.e. without and with \(^{99}\text{Mo}\) extraction. The calculation result shows that the radioactivity inside the SAMOP reactor core reach 523 Ci for the condition without \(^{99}\text{Mo}\) extraction. The effective dose at a distance of 50 cm from the outer surface of the SAMOP reactor coolant tank is 1600 mSv/h. When the SAMOP reactor is operated then followed by \(^{99}\text{Mo}\) extraction, the overall radioactivity of the reactor core is slightly lower than the above value. The calculation results also shows that the highest dose from volatile nuclide is caused by \(^{132}\text{Te}\) isotope i.e. 92.12 mSv/h.

Keywords: Subcritical, reactor, source term, \(^{99}\text{Mo}\), extraction, MCNPX, radioactivity.

1. Introduction

The \(^{99}\text{Mo}\) radioisotope is indispensable as a \(^{99m}\text{Tc}\) generator, of which \(^{99m}\text{Tc}\) radioisotope is the most widely used radioisotope for diagnostics in the nuclear medicine [1,2,3]. However, the increased need for the \(^{99m}\text{Tc}\) isotope can’t be fulfilled because the \(^{99}\text{Mo}\) commonly used as a \(^{99m}\text{Tc}\) generator can only be produced in a nuclear reactor. The half-life of \(^{99}\text{Mo}\) which only 66 hours is also a factor making it difficult to store and mobilize its spread. This problem can be solved by shortening the \(^{99}\text{Mo}\) production process as well as shortening the mobilization of radioisotopes spreading distance.

In general, the \(^{99}\text{Mo}\) production is done by splitting the \(^{235}\text{U}\) nuclide, which \(^{235}\text{U}\) is irradiated with thermal neutrons inside a nuclear reactor so that a critical reactor is required. The licensing issues for critical reactors are complex because they have to fulfill strict requirements. This problem requires a very huge funds to realize, excluding the cost of waste management products. To solve this problem BATAN or the National Nuclear Energy Agency designed a reactor that could produce \(^{99}\text{Mo}\) but did not categorized as critical reactor and did not use high-enriched uranium. This reactor is simple and safer. The reactor applies subcritical reactor method with an external neutron source or an isotropic neutron source, known as SAMOP or subcritical assembly for \(^{99}\text{Mo}\) production [4].

This research is based on previous studies, related to SAMOP design and safety analysis [5,6]. 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 [7]. SAMOP
system is similar with an Aqueous Homogenous Reactor (AHR) but its operate a subcritical condition. AHR is a type of reactor in which uranium is dissolved in water. The fuel used is a mixture of coolants such as water which also act as a moderator and uranium salt, often referred to as homogeneous reactors. The high negative temperature reactivity coefficient makes the AHR more stable than conventional reactors. Another positive aspect of AHR is its small size and low total power [8,9]. However AHR cannot be used to generate electrical power because the fuel should not boil, otherwise the fuel concentration will increase due to evaporation of the water and the system is no longer critical. Using AHR, other isotopes can be extracted from solutions other than 99Mo. Until 2016, there are only 5 Aqueous Homogenous Reactors operating in accordance with IAEA databases [9,10].

The purpose of this research is to analyze the source term of SAMOP reactor experimental facility by using simulation method. The simulation modeling is done using MCNPX software [11]. The analysis results will be used for designing the radiation shielding of SAMOP experimental facility. In this source term calculation, the burn-up calculations is done. The burnup calculation is done both in the reactor operation without $^{99}$Mo extraction and with 100% $^{99}$Mo extraction per batch (6 day + 1 day reactor power off) within 1 month operation or 5 batch.

2. Modelling 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 $10^8 \text{n/cm}^2\text{s}$ [7]. The SAMOP core consists of annular cylindrical tube containing uranyl nitrate [UO$_2$(NO$_3$)$_2$] as fuels and target, surrounded by ring of UO$_2$(NO$_3$)$_2$ fuel rods. The TRIGA fuel elements can be loaded in the ring together with UO$_2$(NO$_3$)$_2$ tubes to increase neutron multiplication factor. Uranyl nitrate that is used in this research has enriched to 19.75% $^{235}$U. Figure 1 and Figure 2 shows the dimensions of SAMOP experimental facility, and the dimensions of core and reflector system.

![Figure 1. SAMOP experimental facility](image1)

![Figure 2. Dimensions of core and reflector system](image2)

3. Methods

3.1. Isotope production calculation

Isotope production calculation, or source term calculation is a calculation of isotope production within reactor operation that can be formed in the reactor through nuclear fission reaction, fusion (in case of fusion reactor), and nuclear transmutation reaction. Isotope that produce within reactor could be stable
or may be unstable that may lead to nuclear decay and emitting radiation, such as alpha, beta or gamma ray radiation. These three types of radiation might be harmful for environment, reactor operator, and people. The calculation of isotope production becomes important to do in order to calculate the effective dose of radiation received by the worker from the operation of the reactor. From this calculation, we will find the number of isotopes present in the reactor core and the resulting radioactive activity.

Isotope production calculation can be done using burnup calculation that provided by neutronic or reactor code system, such as MCNPX, Serpent, SCALE, etc. In this study, MCNPX is used as a code to solve burnup calculations and produces information of number of isotopes produced inside the reactor core. Both of the previously described SAMOP reactor core configurations are then modeled using MCNPX and then burnup codes are run for two conditions, reactor cores operation without extraction with 6-day operating intervals, and reactor cores operation with 100% 99Mo extraction in all fuel rods per batch. One batch of SAMOP operation is 6 days operation in full power 1kW and one day reactor power off to extract 99Mo.

3.2. Effective dose calculation
The effective dose calculation resulting from the radioisotope present in the reactor core is done by multiplying the radioisotope activity with the corresponding specific gamma-ray dose constants. The specific gamma-ray dose constants value of the radioisotopes is derived from the ORNL technical document for dosimetry and radiological assessment [12].

This effective dosage calculation does not include the dose received due to the gamma ray radiation exposure of the neutron capture (n, gamma) when the reactor operates. In other words, the dose calculations reported in this article are limited to doses of radionuclides decaying inside the reactor core. Further consideration is desirable when selecting which radionuclides contribute significantly to the dose [13]. With medium sized power reactors, it usually suffices to consider the following set of radionuclides: whole body noble gases (particularly 85Kr, 135Xe and 133Xe); thyroid (particularly 131I, 131I); lung/internal: volatile nuclides (e.g. 131I, 132Te, 106Ru, 134Cs, 137Cs), and 89Sr for scenarios of high core temperatures (>1000°C).

3.3. MCNPX
MCNPX™ is a general purposes Monte Carlo radiation transport code designed to track many particle types over broad ranges of energies. It is the next generation in the series of Monte Carlo transport codes that began at Los Alamos National Laboratory nearly sixty years ago. MCNPX 2.6.0 is the latest Radiation Safety Information Computational Center (RSICC) release of the code, following the 2005 release of MCNPX 2.5.0 [PEL05]. MCNPX 2.5.0 was a superset of MCNP4C [BRI00] and MCNPX 2.4.0 [WAT02b]. MCNPX 2.6.0 includes many new capabilities, particularly in the areas of transmutation, burnup [FEN06a, FEN06b, FEN08], and delayed particle production. Many new tally source and variance-reduction options have been developed. Physics improvements include a new version of the Cascade-Exciton Model (CEM), the addition of the Los Alamos Quark-Gluon String Model (LAQGSM) option, and a substantial upgrade to muon physics. The code is compatible with MCNP5, and references to MCNP™ in this manual refer to the MCNP5 version [11].

Reactor core modeling using MCNP can be done using vised application provided by the MCNP, or by creating its own input code using a text editor application. The MCNP input code structure consists of cell cards, surface cards and data cards. The cell card contains information about part of the reactor core that is bounded by surface defined on the surface card. Material information such as the type of material used, material density, cell volume (optional), and the type of particles tracked within those cells also defined here. The data card contains information about the details of the material information used in the previous cell card and what type of calculation that user wants MCNP to solve, such as the calculation of reactor core burnup. Flowchart of the calculation is shown in Figure 3.
In the case of calculating core burnup of the SAMOP reactor when operating in $^{99}$Mo extraction mode, at each step of the burnup, the material data composition that used within the reactor core was updated due to the extraction process of $^{99}$Mo and the change of material composition from previous burnup calculation [14].

4. Result and discussion

4.1. SAMOP operate without $^{99}$Mo extraction

The first analysis was to look the quantity of radionuclide on SAMOP reactor core that operate without $^{99}$Mo extraction for 3 months using 6-day intervals. The time period of 3 months were selected to see the long-term effects of radioactive material growth during SAMOP operation. The overall radioactivity of the SAMOP reactor core during the first 3 months can be seen in the Figure 4. The effective gamma-radiation dose generated from the SAMOP reactor core at a distance of 50 cm from the outer side of the tank (1 meter from the radial section) can be seen in the Figure 5.

The total activity of radioactive substances in the reactor core until day 30th reaches 516.681 Ci with effective gamma-ray dose of 1557.523 mSv/h. The largest radioactivity is come from of $^{239}$Np which is 89.32 Ci, followed by $^{131}$Xe 54.68 Ci and then $^{99}$Mo that can reach 51.29 Ci. The radioactivity

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**Figure 3.** Flowchart of the source term calculation

**Figure 4.** Overall core activity without $^{99}$Mo extraction

**Figure 5.** Overall effective gamma dose 50 cm from outer coolant tank without $^{99}$Mo extraction
of radioactive substances in the reactor core increases exponentially, and the effective dose of gamma radiation increases significantly after 24 days of operation. This happens because the growth of $^{140}$La isotope with half-life of 1.7 d is shown by MCNP after 24 days of operation due nuclides with atom fractions below $10^{-10}$ for a material are zeroed by MCNP. $^{140}$La itself derived from the fission reaction, plus the neutron capture by $^{139}$La and the beta decay of $^{140}$Ba. Furthermore, the specific gamma-ray dose constant for $^{140}$La is greatest among the other radionuclides, 3.424 $10^{-4}$ mSv/MBq or 12.66880 mSv/Ci. It can be seen in Figure 4, the overall effective dose of the reactor core due to the source term (radioactive material) at a distance of 50 cm from the outer surface of the coolant tank can be as high as 1600 mSv/h after 1 month, 1860 mSv/h in the 2nd month, and 1980 mSv/h in the third month.

The amount of radionuclides that important to be taken into account because its activity over 1 month operation of SAMOP can be seen in Table 1. It can be seen from the specific gamma-ray dose information that the largest dose among the nuclides is caused by $^{140}$La with 471.91 mSv/h (1 m from point source). The highest dose from volatile nuclide is caused by $^{132}$Te (half-life 3.3 d) i.e. 100.66 mSv/h.

| Nuclide | Specific $\gamma$ dose (mSv/h/Ci) | Radioactivity (Ci) | Max. effective dose (mSv/h) |
|---------|-----------------------------------|-------------------|-----------------------------|
| Sr-90   | 0.0196                            | 0.0392            | day 6                       |
| Ru-106  | 0.0022                            | 0.1722            | day 12                      |
| Te-132  | 2.7931                            | 24.20             | day 18                      |
| I-131   | 2.8268                            | 8.658             | day 24                      |
| Xe-133  | 1.0297                            | 25.37             | day 30                      |
| Cs-137  | 3.7629                            | 33.06             | day 6                       |
| La-140  | 12.6688                           | 73.87             | day 12                      |
| Np-239  | 5.1282                            | 86.85             | day 18                      |

|          | Radioactivity (Ci) | Max. effective dose (mSv/h) |
|----------|--------------------|-----------------------------|
| Sr-90    | 0.0783             | day 24                      |
| Ru-106   | 0.0978             | day 30                      |
| Te-132   | 0.1722             | day 6                       |
| I-131    | 22.77              | day 12                      |
| Xe-133   | 54.68              | day 18                      |
| Cs-137   | 0.3702             | day 30                      |
| La-140   | 471.91             | day 6                       |
| Np-239   | 458.05             | day 12                      |

### Table 1. Amount of source term (radionuclide) of SAMOP core without $^{99}$Mo extraction

4.2. **SAMOP operate with $^{99}$Mo extraction**

The next analysis was to look at the quantity of radionuclide on SAMOP reactor core with $^{99}$Mo extraction mode for 5 batch or approximately 1 month operation. The overall activity of the SAMOP reactor core during the first month can be seen in the Figure 6. The effective gamma-radiation dose generated from the SAMOP reactor core at a distance of 50 cm from the outer side of the tank (1 meter from the radial section) can be seen in the Figure 7. The amount of radionuclides that important to be taken into account for the radiation safety is the activity over 5 batch operations (approx. 34 day) of SAMOP, can be seen in Table 2. It can be seen that the maximum specific gamma-ray ($\gamma$) dose i.e. after day 30, the largest dose among the nuclides is caused by $^{239}$Np with 438.15 mSv/h (1 m from point source). The highest dose from volatile nuclide is caused by $^{132}$Te i.e. 88.91 mSv/h.

![Figure 6](image6.png)  **Overall core activity with $^{99}$Mo extraction**

![Figure 7](image7.png)  **Overall effective gamma dose 50 cm from outer coolant tank with $^{99}$Mo extraction**
Then to show the maximum dose contained in the fuel rod, the nuclide information in the fuel rod in the center of the reactor is shown in Table 3. The fuel rod is selected because this fuel rod has the highest activity among the other fuel rods on the radial section of the reactor core. It can be seen from Table 3 that the highest activity and effective gamma-ray dose at a distance of 1 m from the fuel rod is still owned by a Type 1 configuration. But when all the radioactive materials inside the reactor are added up, the condition reverses.

| Table 2. Amount of source term (radionuclide) of SAMOP core with 100% 99Mo extraction |
| Nuclide  | Specific γ dose (mSv/h/Ci) | Radioactivity (Ci) | Max. effective dose (mSv/h) |
|----------|----------------------------|--------------------|-----------------------------|
| Sr-90    | 0.0196                     | 0.0392             | 0.0588                      | 0.0784                      | 0.0979                      |
| Te-132   | 2.7931                     | 24.20              | 29.71                       | 30.92                       | 31.66                       | 31.83                       | 48.91                       |
| I-131    | 2.8268                     | 8.66               | 13.41                       | 16                          | 17.42                       | 18.19                       | 51.42                       |
| Xe-133   | 1.0297                     | 25.37              | 35.43                       | 39.42                       | 40.99                       | 41.63                       | 42.87                       |
| Cs-137   | 3.7629                     | 0.020              | 0.039                       | 0.059                       | 0.079                       | 0.099                       | 0.3707                      |
| La-140   | 12.6688                    | 0.4589             | 32.69                       | 414.14                      |
| Np-239   | 5.1282                     | 73.87              | 83.77                       | 85.22                       | 85.33                       | 85.44                       | 438.15                      |

| Table 3. Nuclide density an effective dose in the fuel rod in the center of the SAMOP reactor |
| No | Nuclide | mass (g) | Activity (Ci) | Eff. dose (mSv/h) | No | Nuclide | mass (g) | Activity (Ci) | Eff. dose (mSv/h) |
|----|---------|----------|---------------|-------------------|----|---------|----------|---------------|-------------------|
| 1  | U-235   | 2.29E+01 | 4.95E-05      | 1.68E-04          | 16 | Te-132  | 1.59E-06 | 4.84E-01      | 1.53E+00          |
| 2  | U-236   | 8.68E-05 | 5.62E-09      | 4.14E-09          | 17 | I-131   | 2.29E-06 | 2.84E-01      | 8.01E-01          |
| 3  | U-238   | 9.31E+01 | 3.13E-05      | 2.04E-05          | 18 | Xe-133  | 3.20E-06 | 5.99E-01      | 6.16E-01          |
| 4  | Np-239  | 6.14E-06 | 1.42E+00      | 7.30E+00          | 19 | Cs-135  | 1.43E-05 | 1.64E-08      | 0.00E+00          |
| 5  | Pu-239  | 5.06E-05 | 3.14E-00      | 9.46E-06          | 20 | Cs-137  | 1.62E-05 | 1.41E-03      | 5.31E-03          |
| 6  | Rh-87   | 4.24E-06 | 3.64E-13      | 0.00E+00          | 21 | Ba-140  | 7.65E-06 | 5.60E-01      | 9.14E-01          |
| 7  | Sr-89   | 6.43E-06 | 1.87E-01      | 1.52E-04          | 22 | La-140  | 9.59E-07 | 5.34E-01      | 6.76E+00          |
| 8  | Sr-90   | 9.94E-06 | 1.40E-03      | 0.00E+00          | 23 | Ce-141  | 1.08E-05 | 3.08E-01      | 2.25E-01          |
| 9  | Y-91    | 7.54E-06 | 1.85E-01      | 3.70E-03          | 24 | Ce-142  | 1.56E-05 | 7.88E-19      | 0.00E+00          |
| 10 | Zr-93   | 1.01E-05 | 2.55E-08      | 0.00E+00          | 25 | Ce-143  | 1.06E-06 | 7.07E-01      | 1.80E+00          |
| 11 | Zr-95   | 9.88E-06 | 2.12E-01      | 9.85E-01          | 26 | Ce-144  | 1.46E-05 | 4.64E-02      | 1.08E-02          |
| 12 | Nb-95   | 6.25E-07 | 2.46E-02      | 1.18E-01          | 27 | Pr-143  | 7.47E-06 | 5.03E-01      | 0.00E+00          |
| 13 | Mo-99   | 1.20E-06 | 5.78E-01      | 6.51E-01          | 28 | Nd-145  | 1.03E-05 | 4.22E-19      | 0.00E+00          |
| 14 | Tc-99   | 5.15E-06 | 8.82E-08      | 4.05E-13          | 29 | Nd-147  | 2.63E-06 | 2.13E-01      | 2.95E-01          |
| 15 | Ru-103  | 4.52E-06 | 1.46E-01      | 4.84E-01          | 30 | Pm-147  | 2.05E-06 | 1.90E-03      | 0.00E+00          |

| 31 | TOTAL   | 510.21   | 7.00          | 22.33             |

5. Conclusion
The source term of SAMOP reactor experimental facility has been analyzed, the analysis result shows that the overall radioactivity of the reactor core will increase exponentially during reactor operation period. The radioactivity of the source term is dominated by three radioisotopes i.e. 239Np, 133Xe, and 99Mo. The calculation result shows that the radioactivity inside the SAMOP reactor core reach 523 Ci for the condition without 99Mo extraction, and the effective dose a distance of 50 cm from the outer surface of the SAMOP reactor coolant tank is 1600 mSv/h. When the SAMOP reactor
is operated and followed by $^{99}$Mo extraction, then the analysis result shows that the overall radioactivity of the reactor core is slightly lower than the above value. It can be also concluded that the highest dose i.e. 92.12 mSv/h is come from volatile nuclide $^{132}$Te.

**Acknowledgement**

The authors would like to thank the National Nuclear Energy Agency of Indonesia for the MCNPX software. Other than that, the author is also thank to the Director of Center for Accelerator Science and Technology Yogyakarta to Head of Reactor Division and all staffs for their support, and to Secretariat of INSINAS RISTEKDIKTI for budget support to this project (Project Code: InSinas RT-2016-0151).

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