Analysis of fission product gas pressure and radioactivity in SAMOP reactor experimental facility

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Abstract. The fission-product gas pressure and radioactivity analysis of a subcritical assembly for ⁹⁹Mo production (SAMOP) experimental facility have been done. SAMOP reactor is fueled with low enriched uranyl nitrate solution UO₂(NO₃)₂ of 300 g U/L, the reactor core is in the form an annular tube surrounded by a ring of fuel tubes. The SAMOP system is designed to be operated at 100 to 120 hours periodic operation per batch. The analysis method is done by using ORIGEN2 computer code, at the condition of the maximum fuel temperature of 54 °C, 120 hours reactor operating time, and neutron flux varied from 10¹⁰ to 10¹² n/cm²s. The result show that the inventory of the gaseous fission products is dominantly consisted of the xenon, radium, helium, and tritium isotopes with the total activity of 1.3775x10¹³ Bq (372.29 Ci) and total gas volume of 0.0986 cm³ or giving a total gas pressure of 5.78 kPa. The isotopes of xenon and krypton are the major contributors to the total gas pressure. The analysis result shows that the gas pressure of the fission products is very small, therefore the pressure monitoring in the SAMOP reactor core is not necessary.

1. Introduction
The subcritical reactor assembly for ⁹⁹Mo isotope production (SAMOP) experimental facility is being developed in our research center [1,2]. The SAMOP experimental facility as a test facility which use external neutron source from the beam-port of Kartini TRIGA reactor, which has been identified suitable for this purpose [3,4]. SAMOP system is similar with an Aqueous Homogenous Reactor (AHR) but it operates in 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 [5,6]. 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 [7].

The fission products of SAMOP have been analyzed as source term and radiological safety aspect of SAMOP reactor have also been investigated [8,9,10]. The aim of this research is to calculate and to analyze the pressure and radioactivity fission products gasses of SAMOP reactor experimental facility.
The calculation is done by using ORIGEN2 computer code [11]. The analysis results will be used for designing the gas pressure monitoring of SAMOP experimental facility.

2. Description of SAMOP reactor experimental facility

The SAMOP experimental facility is a subcritical nuclear reactor 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 $10^8$ n/cm$^2$ s. The SAMOP core consists of annular cylindrical tube containing uranyl nitrate $[\text{UO}_2(\text{NO}_3)_2]$ or UN as fuels and target, surrounded by ring of $\text{UO}_2(\text{NO}_3)_2$ tubes. The TRIGA fuel elements is loaded in the ring together with $\text{UO}_2(\text{NO}_3)_2$ tubes to increase neutron multiplication factor. The enrichment of all UN used in SAMOP is 19.75% $^{235}\text{U}$.

The SAMOP reactor core and reflector is a cylinder with 40.4 cm in diameter and 43 cm in height. The core and reflector is located in the cooling tank with diameter and height of 120 cm and 400 cm respectively. The SAMOP experimental facility is provided by an instrumentation and control system in such that if there is a criticality indication, the boral control rod neutron absorber will dropped automatically inserted to the SAMOP reactor core. The SAMOP core, reflector, boral rod neutron absorber as well as the location of SAMOP in the Kartini reactor, is described in Figure 1.

![SAMOP core and reflector](image)

**Figure 1.** The SAMOP core/ reflector and its location in the Kartini reactor.

3. Material and methods

The method used are calculations of type and amount of gaseous isotopes fission product resulted in SAMOP reactor. The calculation is done by using ORIGEN2 computer code. ORIGEN2 is a computer code designed to calculate the composition and characteristics of nuclear materials as a function of decay time and the changes the materials undergo during various fuel cycle operations [11].

According to the SAMOP reactor core design geometry, for this calculation the core is divided into two areas or regions i.e. Batch-1 and Batch-2. Batch-1 is the fuel and target / $\text{UO}_2(\text{NO}_3)_2$ in a cylindrical tube located in the central core, and Batch-2 is $\text{UO}_2(\text{NO}_3)_2$ in an annular tube. The elemental composition of batch-1 and Batch-2 is described in Table-1.

To estimate gas conditions at various possible temperatures in the SAMOP reactor, equation (1) can be used to predict the gas volume as function of reactor fuel temperature.
\[
\frac{P_1 \times V_1}{T_1} = \frac{P_2 \times V_2}{T_2}
\]

where \(P_1 = P_2 = 1\) atm, \(T_1 = 273\) K and \(T_2 = 333\) K, \(V_1\) and \(V_2\) will be calculated, the relationship between pressure and temperature in the equation (1) can be applied if there is no change in the amount of gas molecule during the process.

Table 1. SAMOP fuel batch specification.

| No | Parameters                  | Batch-1                  | Batch-2                  |
|----|-----------------------------|--------------------------|--------------------------|
| 1  | Volume                      | 26819.56 cm\(^3\)        | 386.60 cm\(^3\)          |
| 2  | Weight of \(^{235}\)U       | 1609.174 g               | 23.196 g                 |
| 3  | Weight of \(^{239}\)U       | 6436.694 g               | 92.784 g                 |
| 4  | Weight of Oxygen             | 26050.100 g              | 375.508 g                |
| 5  | Weight of Nitrogen           | 948.965 g                | 13.679 g                 |
| 6  | Weight of Hydrogen           | 2713.997 g               | 39.122 g                 |
| 7  | Specific mass of solution    | 1.408 g/cm\(^3\)         | 1.408 gr/cm\(^3\)        |
| 8  | Total weight                | 37758.93 g               | 544.289 g                |

4. Results and discussion

The calculation results of the gas fission products inventory and its radioactivity in Batch 1 and Batch 2 by using ORIGEN 2 computer code [11] are shown in Table 2 and Table 3 respectively. The calculation was done as function of neutron flux level i.e. irradiation time of 120 hours and neutron flux is varied from \(10^{10}\) n/cm\(^2\)s to \(10^{12}\) n/cm\(^2\)s. Each inventory is expressed in units of weight (gram), radioactivity in curie (Ci) and volume in cc. As can be seen from Table 2 and Table 3, the gas fission product nuclides consisted of Helium, Radon, Hydrogen, Krypton and Xenon, this result is in accordance with the similar aqueous reactor [7]. The gas volume is obtained from the noble gas law which states that 1 mole of noble gas ~ 22.4 litres of STP (temperature of 273 K and pressure of 1 atmosphere).

The maximum neutron flux at the SAMOP reactor core is \(\sim 10^{10}\) n/cm\(^2\)s [1,3], therefore, from Table 2 and Table 3 can be found that the total radioactivity of gas fission product in Batch-1 and Batch-2 are 367 Ci and 5.29 Ci respectively. Its mean that the total radioactivity of SAMOP gas fission product is 372.29 Ci or \(1.3775 \times 10^{13}\) Bq.

The calculation of gas fission products in the operation of solution reactors will also generates radiolytic-gas bubbles. The formation of these bubbles creates a void volume in the solution core that introduces a negative coefficient of reactivity, resulting in a power reduction [12]. Although this phenomenon is found in critical aqueous reactor, but for the subcritical reactor system like SAMOP should also be considered. Therefore, the radiolytic gas bubble formation in SAMOP reactor core should be further investigated.
### Table 2. Gas fission product inventory in Batch-1 after 120 h irradiation time.

| Neutron flux | 10^{10} n/cm²s | 10^{12} n/cm²s |
|--------------|-----------------|-----------------|
| Elements     | Weight (g)      | Activity (Ci)   | Volume (cc) | Weight (g) | Activity (Ci) | Volume (cc) |
| ^{4}\text{He} | 5.99E-10        | 0               | 3.36E-06 | 5.99E-08 | 0           | 3.36E-04 |
| ^{219}\text{Rn} | 3.93E-26        | 5.115E-16       | 4.02E-24 | 3.93E-24 | 5.12E-14   | 4.02E-22 |
| ^{3}\text{H} | 5.68E-09        | 5.49E-05        | 4.24E-05 | 5.68E-07 | 5.49E-03   | 4.24E-03 |
| ^{80}\text{Kr}, ^{81}\text{Kr}, ^{81m}\text{Kr}, ^{82}\text{Kr} | 6.03E-05 | 150.4202 | 1.59E-02 | 6.03E-03 | 15042.42   | 1.59E+00 |
| ^{83}\text{Kr}, ^{83m}\text{Kr}, ^{84}\text{Kr}, ^{85}\text{Kr}, ^{85m}\text{Kr}, ^{86}\text{Kr}, ^{87}\text{Kr}, ^{88}\text{Kr} | 4.86E-04 | 216.4956 | 8.10E-02 | 4.86E-02 | 2.16E+04   | 8.10E+00 |
| TOTAL | 5.46E-04 | 3.67E+02 | 9.69E-02 | 5.46E-02 | 3.67E+04 | 9.69E+00 |

### Table 3. Gas fission product inventory in Batch-2 after 120 h irradiation time.

| Neutron flux | 10^{10} n/cm²s | 10^{12} n/cm²s |
|--------------|-----------------|-----------------|
| Elements     | Weight (g)      | Activity (Ci)   | Volume (cc) | Weight (g) | Activity (Ci) | Volume (cc) |
| ^{4}\text{He} | 8.64E-12        | 0               | 4.84E-08 | 8.64E-10 | 0           | 4.84E-06 |
| ^{219}\text{Rn} | 5.67E-28        | 7.37E-18        | 5.80E-26 | 5.67E-26 | 7.37E-16   | 5.80E-24 |
| ^{3}\text{H} | 8.19E-11        | 7.91E-07        | 6.12E-07 | 8.19E-09 | 7.91E-05   | 6.12E-05 |
| ^{80}\text{Kr}, ^{81}\text{Kr}, ^{81m}\text{Kr}, ^{82}\text{Kr} | 8.68E-07 | 2.17002561 | 2.29E-04 | 8.68E-05 | 217.0026   | 2.29E-02 |
| ^{83}\text{Kr}, ^{83m}\text{Kr}, ^{84}\text{Kr}, ^{85}\text{Kr}, ^{85m}\text{Kr}, ^{86}\text{Kr}, ^{87}\text{Kr}, ^{88}\text{Kr} | 7.00E-06 | 3.12141466 | 1.17E-03 | 7.00E-04 | 312.1415   | 1.17E-01 |
| TOTAL | 7.87E-06 | 5.29E+00 | 1.40E-03 | 7.87E-04 | 5.29E+02 | 1.40E-01 |
Table 4. Fission product gas volume in the fuel batch at as function of neutron flux and temperature.

| Region | Gas temp. (K) | Neutron flux $10^{10}$ n/cm² s | Neutron flux $10^{12}$ n/cm² s |
|--------|---------------|-------------------------------|-------------------------------|
|        |               | Gas volume (cc) | Gas volume (cc) |
| Batch-1| 273           | 0.0969            | 9.69  |
|        | 333           | 0.118             | 11.82 |
| Batch-2| 273           | 0.0014            | 0.1400 |
|        | 333           | 0.0017            | 0.1707 |

5. Conclusion
The pressure due to the fission product gases is relatively very small, therefore the pressure monitoring in the SAMOP reactor core is not necessary. The isotopes of xenon and krypton are the major contributors to the total gas pressure. The total radioactivity generated from gas fission product is 372.29 Ci or 1.3775x10¹³ Bq.

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