Analysis of Uranyl Nitrate Hexahydrate Composition for Optimum Neutron Multiplication Factor of SAMOP

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Abstract. Subcritical Assembly for Molybdenum Production (SAMOP) is a concept of nuclear reactor where the neutron multiplication factor ($k_{\rm eff}$) less than one ($k_{\rm eff} \sim 0.99$). Therefore, SAMOP reactor need an external neutron source for its operation. Uranyl Nitrate Hexahydrate, UO₂(NO₃)₂ 6H₂O or UNH is used as fuel as well as target material for ⁹⁹Mo production. The ⁹⁹Mo isotope is used to generate ^{99m}Tc which is the most widely used radioisotopes for diagnostic in nuclear medical fields. The composition of Uranyl Nitrate Hexahydrate (UNH) should be analyzed by calculation methods to achieve the required subcritical level in the SAMOP core i.e. k-eff ~ 0.99. Methodology for this analysis is an elemental composition calculation of UNH, with enrichment of uranium of 19.75% and 2.81 gr/cm³ of density. The analysis result shows that the elemental composition of ²³⁵U, ²³⁸U, N, O, and H, are 4.9715%, 20.2009%, 5.3448%, 37.4138%, and 32.069% respectively, and therefore, the composition of U and N-O-H are 25.1724%, and 74.8276% respectively. The composition is then used as input for the neutronic analysis using the WIMS-D/4 computer code for $k_{\rm eff}$ calculation of SAMOP. The result shows that the optimum $k_{\rm eff}$ of 0.99 is achieved with the above elemental compositions.

Keywords: SAMOP, multiplication factor, uranyl nitrate, composition, neutronic.

1. Introduction

Subcritical Assembly for Molybdenum Production (SAMOP) is a concept of nuclear reactor where the neutron multiplication factor (k_{eff}) less than one. Neutron multiplication factor, k_{eff} is a ratio of the neutron production rate with the neutron absorption and neutron leakage rates in the nuclear reactor system. The SAMOP reactor need an external neutron source for it continue operation. Uranyl Nitrate Hexahydrate, $UO_2(NO_3)_2$ 6H₂O or UNH is used as fuel as well as target material for Mo⁹⁹ production [1,2]. The Mo⁹⁹ isotope is used to generate ^{99m}Tc which is the most widely radioisotopes used for diagnostic in nuclear medical fields [3,4].

The development of the SAMOP reactor system is initiated by the need of Mo production without critical reactor, that is by developing a concept of subcritical reactor system [5]. This concept needs a careful analysis in the content composition of the reactor fuel and target (UNH) as well as the material and geometric of the reactor core for achieving the value of $k_{\text{eff}} \sim 0.99$. Modelling for analysis

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of UNH composition for optimum $k_{\rm eff}$ needs a preparation input of UNH composition, material and geometric of the reactor core. The parameter is one of important factors that would contribute to the accuracy of the neutronic calculations result, as such used in reactor core design, core power distributions etc.

The composition of UNH should be analysed to achieve the required subcritical level in the SAMOP core i.e. $k_{eff} \sim 0.99$ [5,6]. The elements composition of UNH is then used as input in the reactor calculation by using WIMS-D/4 computer code. The other similar parameters which is already analysed is number density, that is number of atoms or nuclides per cm³ (atoms/cm³), or in unit of atoms per 10^{24} barn cm (atom / barn cm) [7]. This analysis is done as a benchmarking in neutronic calculation where several neutronic analyses have been previously conducted [8,9,10,11].

2. Material and methods

2.1. Description of SAMOP reactor

The SAMOP reactor core consists of annular cylindrical tube containing uranyl nitrate UO₂ (NO₃)₂6H₂O or UNH as fuels and target, surrounded by ring of UNH tubes and or TRIGA fuel elements. The TRIGA fuel elements is loaded in the ring together with UNH tubes to increase neutron multiplication factor. The enrichment of all UNH is similar with TRIGA fuels i.e. 19.75% ²³⁵U. The SAMOP reactor core is surrounded by neutron reflector made of graphite. The SAMOP reactor core and reflector design concept is shown in Figure 1.

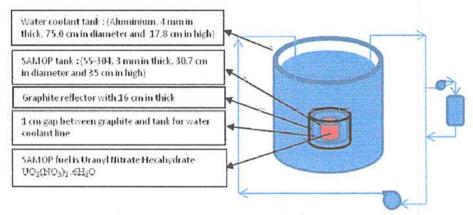


Figure 1. SAMOP reactor core design concept

2.2. Methodology

Analytical calculation methods is used for elemental composition determination of UNH, with using 19.75% enrichment of ²³⁵U and 2.81 gr/cc of uranium density. The elemental composition of U, N, O, and H in UNH is then used for the analysis of subcritical level of SAMOP reactor by using WIMS=D/4 computer code. The WIMS-D/4 code is a freely available thermal reactor physics lattice cell code [12,13] that is widely used in many laboratories for thermal research reactor and power reactor calculations. WIMSD-D/4 version is thermal reactor physics lattice cell code developed by Winfrith-United Kingdom

The UNH is fuels and also as target for ⁹⁹Mo isotope production in SAMOP reactor. Whiles, SS-304, graphite, water and aluminium are as materials contain in SAMOP reactor core and reflector. The materials for WIMSD code input are as follows: UNH as fuel, SS-304 for tank, graphite for moderator, H₂O for coolant, Al for tank, and light water r for coolant. The annulus geometric is used for each geometric input. The modeling of SAMOP concept is described in CELL model input of WIMSD for cylindrical geometric, see Figure 2.

The materials for WIMSD code input are as follows: UNH as fuel, SS-304 for tank, graphite for moderator, H_2O for coolant, Al for tank, and light water r for coolant. The annulus geometric is used for each geometric input. The modeling of SAMOP concept is described in CELL model input of WIMSD for cylindrical geometric with 8 regions, see Figure 2.

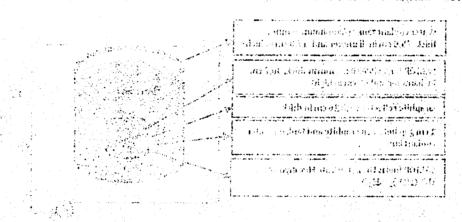
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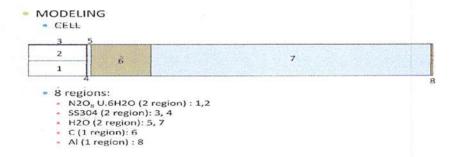


Figure 2. Eight regions of SAMOP lattice cell modeling for WIMSD input.

3. Result and discussion

The UNH with 300.0 gU/L in 25908.05 cm 3 volume of SAMOP reactor core contains variations in composition of $_{92}U^{235}$, $_{92}U^{238}$, $_{7}N^{14}$, $_{1}H^{1}$, $_{8}O^{16}$. The material composition for each region of SAMOP reactor is shown in Table 1.

Table 1. Composition of materials in the region of SAMOP reactor

Région	Material	Density	Volume (cm ³)	Composition
1 and 2	UNH	300.0 gU/L	25908.05	0.681035% of ₉₂ U ²³⁵ ; 2.76725% of ₉₂ U ²³⁸ ; 6.8966% of ₇ N ¹⁴ ;48.2415% of ₈ O ¹⁶ ; 41.3794% of ₁ H ¹
3 4	SS304	8.0 g/cm ³	222.069 520.0592	79.9 % of Fe; 18.0% for Cr; 0.8% for Ni; 1% for Si; 0.1% for Nd; 0.2% for Mn
5	H_2O	1 g/cm ³	1806.227	88.89% for H and 11.11% for O
6	C	1.6 g/cm ³	44109.97	100% of C
7	H_2O	1 g/cm ³	254939.6	88.89% for H and 11.11% for O
8	Al	2.7 g/cm ³	3364.17	99.5% of Al; 0.18% of Cu; 0.2% of Fe; 0.03% of Cu; 0.03% of Mg; 0.03% of Mn; 0.03% V

The neutronic calculation using WIMSD code of the SAMOP concept is done withcell modellings related to different composition of the uranium in UNH, to find the optimum k_{eff} . The model is shown in Figure 2 above. The different composition of UNH with 300.0 gU/ltr in 25908.05 cm³ volume is varied in composition of ${}_{92}U^{235}$, ${}_{92}U^{238}$, ${}_{7}N^{14}$, ${}_{1}H^{1}$, ${}_{8}O^{16}$.

The optimum result shows that the SAMOP reactor operation is still under subcritical condition (k_{eff} <1), see Figure 3. The analysis result shows that the optimum k_{eff} is 0.9919 with the composition of U^{235} , U^{238} , N^{14} , O^{16} , H^1 are 4.9715%, 20.2009%, 5.3448%, 37.4138%, 32.069% respectively. Therefore, the composition of U and N-O-H are 25.1724% and 74.8276% respectively. It is shown in Figure 3 that the uranium composition of 25% in UNH is the optimum percentage in order to reach a neutron multiplication factor of 0.99 or k_{eff} ~ 0.99 i.e. the optimum subcritical level for SAMOP.



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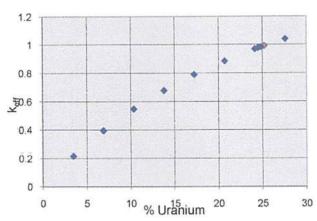


Figure 3. The keff calculation results for different % of uranium in UNH fuel

In a solution nuclear reactor design calculation, some variations of ²³⁵U percentage in the solution are commonly needed. This will affect the isotopic compositions of the main uranium isotopes i.e. ²³⁵U and ²³⁸U for the respective uranium concentrations. This phenomenon is in accordance with references [14,15] and with the previous analysis result [2,7].

4. Conclusion

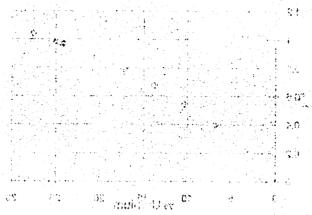
The analysis of elements compotion by calculation methods contained in a homogeneous uranyl nitrate hexahydrate solution (UNH) has been done. The composition of UNH for enrichment of uranium of 19.75% and 2.81 gr/cc of density shows that the elemental composition of 235 U, 238 U, N, O, and H, are 4.9715%, 20.2009%, 5.3448%, 37.4138%, and 32.069% respectively. Its mean that the composition of U and N-O-H are 25.1724%, and 74.8276% respectively. The analysis result shows that the optimum k_{eff} of 0.99 or neutron multiplication factor in SAMOP reactor can be achieved with that elemental compositions.

5. Acknowledgements

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