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Research Article

Design and Development of Subcritical Reactor by Using Aqueous Fuel for ^{99}Mo Production

Syarip*, Tegas Sutondo, Edi Trijono Budisantoso, and Endang Susiantini

Research Centre for Accelerator Science and Technology (CAST),
National Nuclear Energy Agency (BATAN), Jalan Babarsari, Yogyakarta 55281, Indonesia

Abstract: A non-critical reactor system for ^{99}Mo production has been designed and will be developed at the Centre for Accelerator Science and Technology (CAST), National Nuclear Energy Agency (BATAN). The system is called subcritical aqueous reactor "assembly" for ^{99}Mo production (SAMOP) which is fueled with uranyl nitrate and driven by external neutron source from a neutron generator. The design characteristics of criticality analysis, safety aspect, and calculation of ^{99}Mo specific activities, choice and estimation of technology for ^{99}Mo separation from the irradiated uranyl nitrate are presented in this paper. The analysis result showed that the SAMOP system using low enriched uranyl nitrate $UO_2(NO_3)_2$ of 300 g U L^{-1} has an effective neutron multiplication factor of 0.98 to 0.99 with an average neutron flux of 10^{10} n cm^{-2} s. The total volumes of $UO_2(NO_3)_2$ and uranium content in the core are 23 L and 3.8 kg, respectively. The SAMOP system using low enriched uranyl nitrate with total volume of 23 L and 3.8 kg uranium content may produce 111 GBq/batch (3 000 mCi/batch) of ^{99}Mo.

Keywords: Criticality analysis, ^{99}Mo, production, subcritical, aqueous, uranyl nitrate

1. INTRODUCTION

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, with some 40,000,000 procedures to 45,000,000 procedures per year (16700,000 in USA in 2012, 550,000 in Australia), 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 is scheduled for shutdown in 2016, which means that another ^{99}Mo shortage is imminent unless a potential domestic ^{99}Mo producer fills the void [5]. Recent ^{99}Mo production capacity of PT INUKI (Indonesian Nuclear Industry) is about 70 Ci per week while the national need is about 100 Ci per week, required by nuclear medical division in 13 hospitals (personal communication with Production Director of PT-INUKI). Whiles, the ^{99}Mo production capability of Indonesian reactor (RSG-GAS) can reach 300 Ci per batch (week) [6]. The ^{99}Mo producing system without a nuclear reactor and without using highly-enriched uranium is being implemented at our research center. Its production

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*Email: Syarip: syarip@batan.go.id

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 [5].

The objective of this work was to study the possibility of producing ^{99m}Tc in useful activities by using non-critical reactor based on an accelerator driven system. A non-critical reactor or subcritical aqueous homogeneous reactor 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), National Nuclear Energy Agency (BATAN). The aim of the research included the determination of the criticality level, calculation of ^{99}Mo specific activities, determination of optimal irradiation conditions and choice and estimation of technology for ^{99m}Tc separation from the irradiated uranyl nitrate. It is expected that the SAMOP system will reduce much less waste than current ^{99}Mo production methods. 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].

2. MATERIALS AND METHODS

The method used for this design analysis is modeling and calculation by using a MCNPX computer code for criticality analysis, and ORIGEN-2 for calculation of ^{99}Mo specific activities, and analytical calculation for thermal hydraulics analysis. Reference study is intended to choose and estimate the technology for ^{99}Mo separation from the irradiated uranyl nitrate. MCNPX is a general-purpose particle transport Monte Carlo code developed by the Los Alamos National Laboratory designed to track many particle types over broad ranges of energies [8]. 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 [9].

2.1. Brief Description of SAMOP

The schematic diagram of Subcritical Assembly for Molybdenum Production (SAMOP) is depicted in Fig. 1. The SAMOP system consists of a 26 L-core tank surrounded by graphite reflectors, filled with 23 L of uranyl nitrate solution. The entire system is placed in a tank filled with water for cooling purposes. The critical uranium concentration depends on factors as the thickness of the reflector and the placement of the reactor in the cooling tank, but has a minimum value of 300 g 19.75 %

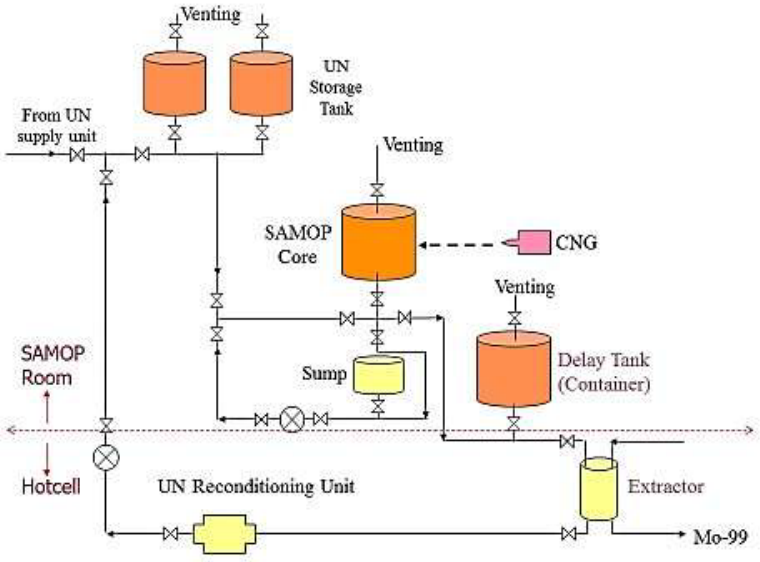


Fig. 1. Schematic diagram of SAMOP.

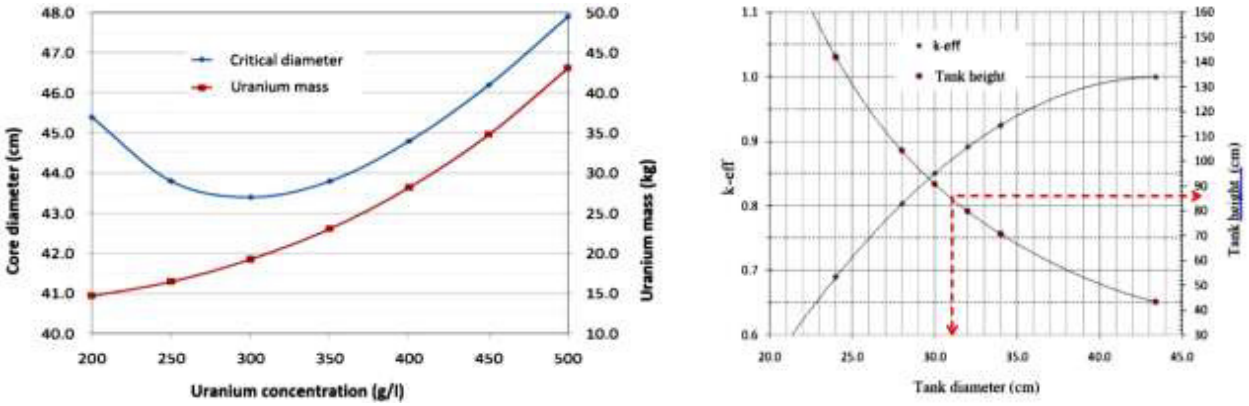


Fig. 2. Criticality analysis result: critical diameter as function of uranium concentration and neutron multiplication (k_{ef}) as function of tank height.

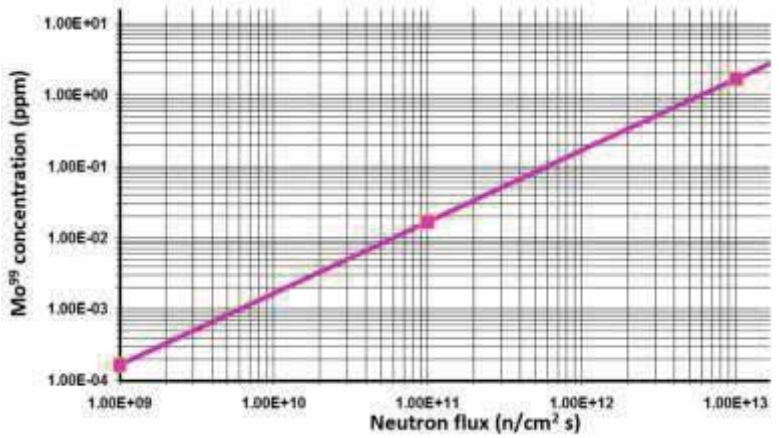


Fig. 3. ^{99}Mo production as a function of neutron flux.

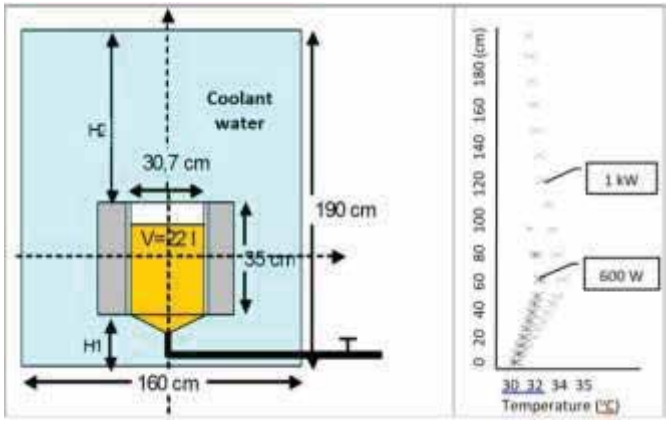
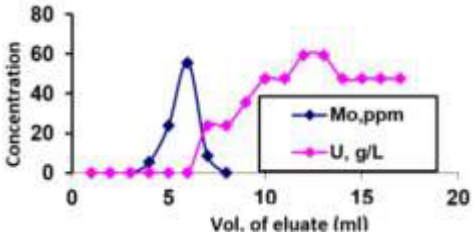


Fig. 4. Axial temperature distribution of SAMOP water coolant tank.

enriched uranium per liter. After irradiation, the fuel is first stored for some time in the delay tank. Subsequently, the molybdenum is extracted in the extraction column. In the reconditioning facility, the remainder of the solution is reconditioned and prepared to be reinserted in the reactor.

The choice of uranyl nitrate for SAMOP fuel is due to the fact that ^{99}Mo is more easily extracted from a uranyl nitrate solution than from uranyl



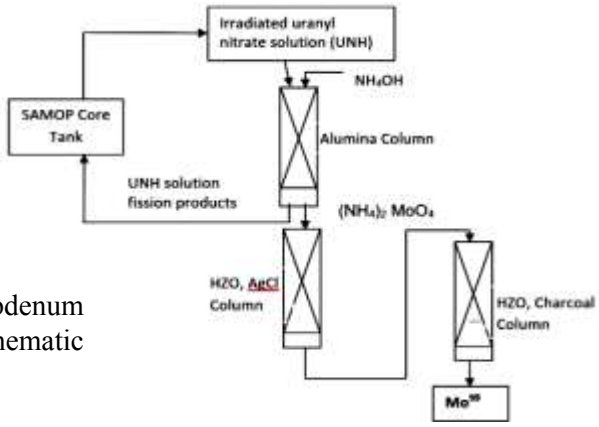


Fig. 5. Experimental result of molybdenum separation from U-nitrate solution, and schematic diagram of ^{99}Mo separation process.

sulfate. This is because competition for adsorption sites between molybdenum and sulfate is stronger than that between molybdenum and nitrate [5, 12]. Similarly, uranyl nitrate was used as a fuel in the HYPO and SUPO reactors [12].

3. RESULTS AND DISCUSSION

The criticality analysis results using MCNPX are depicted in Fig. 2. It is shown that the critical dimension is a function uranium concentration. The lowest critical diameter was found for 300 g L^{-1} uranium concentration, it means this number is the optimum concentration. Therefore, this value is used for the determination of tank diameter of SAMOP core. The SAMOP reactor core tank diameter is fixed 30 cm, then the height is optimized. The correlation between the neutron multiplication (kef) and tank height is depicted in Fig. 2. It is shown that the higher the tank height, and the higher kef the diameter will be, and this result is in accordance with the similar work done by other researchers [13 14, 15].

The calculation result of ^{99}Mo production as a function of neutron flux using ORIGEN-2 is described in Fig. 3. Based on this calculation the ^{99}Mo production can be predicted between 111 to 185 GBq (3 000 mCi to 5 000 mCi) per batch for neutron flux of 109 n cm^{-2} s^{-1} to 1010 n cm^{-2} s^{-1}.

Fig. 4 shows the axial temperature distribution of SAMOP water coolant tank calculated for average total power generation of 600 W and 1 kW power generated in SAMOP core. The maximum coolant temperature in steady state condition is 33 °C which is almost the same with ambient temperature.

Table 1. Technical design specification of SAMOP.

Parameter	**Material/Value/Dimension**
Fuel solution	U-nitrate $UO_2(NO_3)_2$
Uranium enrichment	19.75 w/o U235 enrichment
Uranium concentration	300 g U/L
Core volume	23 L
U235 total	3.8 kg
Core height	35 cm
Core diameter	30.7 cm
Reflector thickness	20 cm, graphite
Radiation shielding	60 cm, heavy concrete
Neutron multiplication factor	0.98 – 0.99
Average neutron flux	1010 n Cm^{-2} s^{-1}
Max. fuel temperature	55 oC
Thermal power	600 W
Mo99 Production	111 GBq/batch per week (3000 mCi/batch)

Whiles, the result of preliminary experiment of molybdenum separation from U-nitrate solution containing 300 mg L^{-1} of molybdenum in 10 mL of 100 g L^{-1} uranium is described in Fig. 5. It is shown that Mo appears at fourth eluate while uranium at 7th eluate, this shows that separation already existed with separation factor of 7.5.

The ^{99}Mo purification process will be done by using HZO resin from $(NH_4)_2MoO_4$ solution at pH 2 to 7. This process will be carried out together with the modified Cintichem process. The ^{99}Mo separation and purification processes are depicted in Fig. 6.

In summary, the SAMOP system using low enriched uranyl nitrate of 300 gram U L–1 with total volume of 23 L and uranium content of 3.8 kg may produce 111 GBq/batch (3,000 mCi/batch) of Mo^{99}. For the preliminary experiment in 2017, it will be used neutron source from the radial beam port of Kartini research reactor instead of using CNG (compact neutron generator). The design analysis result is summarized in Table 1.

4. CONCLUSIONS

The SAMOP system using low enriched uranyl nitrate of 300 g U L^{-1} with total volume of 23 L and uranium content of 3.8 kg has been being designed to produce 111 GBq/batch per week or 3000 mCi/batch of ^{99}Mo. Smaller dimension of SAMOP core, less than 30 cm in diameter is safer to avoid criticality. The future work, a preliminary experimental SAMOP facility by using neutron beam from Kartini research reactor, is being prepared.

5. ACKNOWLEDGMENTS

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