Neutronic analysis of critical assembly for moly-99 production reactor based on mixed Th-U fuels

B Delphito, Syarip
Departments of Nuclear Engineering and Engineering Physics of Gajah Mada University, Jl. Grafika 2 Yogyakarta 55281, Indonesia
Center for Accelerator Science and Technology, National Nuclear Energy Agency, Jalan Babarsari, Yogyakarta, Indonesia
E-mail: thephitonthel@gmail.com

Abstract. Neutronic analysis of nuclear reactor systems for 99Mo isotope production based on mixed thorium-uranium (Th-U) nitrate fuels has been done. The 99Mo isotope is a 99mTc generator which is the most widely used radioisotope in nuclear medicine. The proposed critical reactor system or critical assembly for 99Mo production, called CAMOLYP, is currently being developed in BATAN Yogyakarta as a continuation of the development of subcritical assembly for 99Mo production called SAMOP R&D program. The aim of this research is to obtain the optimum CAMOLYP core configurations, the core is an annular cylinder provided by the outer ring. The method used is the calculation of the critical level of CAMOLYP for two reactor core configuration models, using the MCNPX computer code. The analysis result shows that there are various configurations of CAMOLYP core to achieve criticality condition. The best configuration of Design-A is by using a Th-U nitrate density of 300 g Th-U/L or 120 g U/L, surrounded by 38 TRIGA wt-12% in the outer ring (k_e=1.000334). While in Design-B, the best configuration is by using 24 fuel rods with a UN volume of 29.0978 L using the UN density of 3000 g U/L (k_e=1.0562). The amount of 235U in the first core design is 3376.74 g, while the second design is 1724.046 g. Based on the total amount of U needed to achieve criticality, the author concluded that the best design for CAMOLYP is Design-B.

1. Introduction
Thorium has been used as a fuel on a prototype scale since 1962 in the United States [1]. Thorium is considered better because of its greater availability compared to uranium so that it can meet the world's energy demand for longer [2]. It also has a greater advantage in terms of low nuclear waste and proliferation [3,4]. As a breeder reactor that produces uranium-233 that can be used as fuel for nuclear reactors, thorium can also indirectly produce radioisotope molybdenum (99Mo) which is one of the most strategic materials in medical activities in the field of nuclear medicine [5].

The concept of isotope production reactor (RPI) or critical assembly for moly production (CAMOLYP) is based on the fission process of uranium-235 and uranium-233. Uranium-235 is needed early in the cycle to reach a critical condition. As the fission happens, thorium-232 will capture neutrons and decay into fissile uranium-233. It is expected that in the next cycle, uranium-235 is not needed because the fissile fuel is sufficient through the breeder process of thorium-232 [6,7].

CAMOLYP reactor core is developed based on subcritical assembly for 99Mo production (SAMOP), where the CAMOLYP reactor design uses thorium-uranyl nitrate solution fuel which refers to the...
SAMOP geometry design. The preliminary calculation was performed and the effective neutron multiplication factor ($k_{ef}$) value shown 0.948 [8], therefore, to reach a critical condition or $k_{ef} = 1$ further research is still needed. These works are a continuation of the neutronic analysis of the CAMOLYP reactor to find the optimum core configuration and dimensions.

2. Reactor design
CAMOLYP is a modular reactor, where the reactor core and reflector design is shown in Figure 1. The CAMOLYP reactor tube section consists of an inner tube and an inner shell or cylindrical annular core. The inner tube is made of SS-316 alloy. The reactor shell is in the form of cooling water. The inner tube and cylindrical annular core contain nitrate solution (uranyl thorium nitrate or uranyl nitrate) which functions as reactor fuel and as a target for $^{99}$Mo production. The top of the inner tube is connected with a neutron absorbent rod [9, 10].

![Figure 1. The 3D model of CAMOLYP.](image-url)

The CAMOLYP reactor core is surrounded by a ring containing UZrH type reactor or TRIGA type 104 standard fuel element type, and control rods made of B4C (boron carbide). The reactor core is surrounded by a reflector in the form of graphite and the reactor cooling water in the form of demineralized water.

3. Methodology
There are two proposed designs that will be calculated using the MCNPX computer program. The first design, which is design A, is specified in Table 1. This design uses thorium-uranyl nitrate as a fuel in the tube. The ratio of uranium and thorium in the nitrate solution is also specified in Table 1. Since thorium will absorb a neutron and acts as a neutron poisoning, it will have a lower $k_{ef}$ compared to uranyl nitrate.
**Table 1.** Technical specification of design-A.

| Parameter                                      | Material/ Value          |
|-----------------------------------------------|--------------------------|
| Fuel in inner tube/inner shell                | Th-U nitrate             |
| Diameter of inner tube/annular core           | 5 cm / 38 cm             |
| Height of inner tube/annular core             | 38 cm / 38 cm            |
| Maximum number of fuel rods                   | 34                       |
| Fuel in the ring                              | TRIGA 104                |
| Reflector material                            | Graphite                 |
| Moderator material                            | Demineralized water      |
| Density of Th-U                               | 300 g Th-U/L             |
| The ratio of uranium:thorium                  | 40%:60%                  |
| Density of uranium                            | 120 g U/L                |
| Density of thorium                            | 180 g Th/L               |
| Enrichment of $^{235}$U                       | 19.75%                   |
| Atomic density of $^{235}$U                   | 6.00908E+22 atom/L       |
| Atomic density of $^{238}$U                   | 2.44166E+23 atom/L       |
| Atomic density of $^{232}$Th                  | 4.66992E+23 atom/L       |
| Atomic density of N                           | 2.47648E+24 atom/L       |
| The atomic density of H                       | 5.30488E+25 atom/L       |
| The atomic density of O                       | 3.45624E+25 atom/L       |

Design-A will be calculated using diameter variations at the increment of 5 cm by using multiple fuel rods variations: TRIGA fuel type of 8.5%-weight, and 12%-weight.

Since the maximum number of fuel rods will increase as the diameter increase, the calculation will use the maximum possible number of fuel rods. The number of fuel rods (TRIGA fuel rods) in the ring as a function of reactor core diameter is shown in Table 2.

**Table 2.** The number of fuel rods in the ring.

| Annular Core Diameter | Number of Fuel Rods |
|-----------------------|---------------------|
| 38 cm                 | 34                  |
| 43 cm                 | 38                  |
| 48 cm                 | 42                  |
| 53 cm                 | 46                  |
| 58 cm                 | 50                  |
| 63 cm                 | 55                  |
| 68 cm                 | 59                  |

The second model (Design-B) uses uranyl nitrate (UN) instead of thorium-uranil nitrate (Th-UN) at the inner tube and at the annular core. Design-B also uses thorium-uranil nitrate at the outer ring instead of TRIGA 104 fuel such as specified in Table 3. The specification of the uranyl nitrate is also shown in
Table 3. Design specification of Design-B.

| Parameter                          | Material/ Value                        |
|-----------------------------------|----------------------------------------|
| Fuel in inner tube/annular core   | Uranyl Nitrate (UN)                    |
| Diameter of inner tube/annular core | 5 cm / 31.4 cm                        |
| Height of inner tube/annular core | 38 cm / 38 cm                         |
| Maximum number of fuel rods       | 24                                     |
| Fuel in the ring                  | Th-UN                                  |
| Reflector material                | Graphite                               |
| Moderator material                | Demineralized water                    |
| Density of U                      | 300 g Th-U/L                          |
| Enrichment of $^{235}$U           | 19.75%                                 |
| Atomic density of $^{235}$U       | 1.50227E+23 atom/L                    |
| Atomic density of $^{238}$U       | 6.10416E+23 atom/L                    |
| Atomic density of N               | 1.52129E+24 atom/L                    |
| The atomic density of H           | 5.48299E+25 atom/L                    |
| The atomic density of O           | 3.35001E+25 atom/L                    |

4. Results and Discussion

Calculations to achieve critical condition were done based on Design-A by using MCNPX. The result is shown in Figure 2.

Figure 2. The $k_{ef}$ as function annular core diameter on design A.
By using TRIGA 104 wt-8.5%, the criticality is achieved at around 63 cm – 68 cm. Using an interpolation technique, the criticality is achieved at 64 cm with a $k_{ef}$ value of 1.0139.

By using TRIGA 104 wt-12%, the criticality is achieved at around 43 cm – 48 cm. Using an interpolation technique, the criticality is achieved at 44 cm with a $k_{ef}$ value of 1.0003. To choose the best design, one thing to consider is the amount of uranium-235 needed to achieve criticality. The author then calculated the total amount of uranium-235 needed at each variation as shown in figure 3.

**Figure 3.** The total mass of $^{235}\text{U}$ to achieve $k_{ef}$ on Design-A.

Figure 3 shows us that the CAMOLYP requires a lesser amount of uranium-235 by using TRIGA 104 wt-12% (around 3000 g $^{235}\text{U}$), compared to TRIGA 104 wt-8.5% (around 5000 g $^{235}\text{U}$). Moreover, by using TRIGA 104 wt-12%, the diameter of CAMOLYP can be reduced to 44 cm, which requires around 1347.65 g $^{235}\text{U}$ in uranyl nitrate, while a diameter of 64 cm by using TRIGA 104 wt-8.5% requires around 2859.82 g $^{235}\text{U}$ in uranyl nitrate. The author then concluded that it is better to use TRIGA 104 wt-12% because it requires a lesser amount of uranium-235 in both UZrH and uranyl nitrate.

Calculations were done based on Design-B by using MCNPX. The result is shown in figure 4.

**Figure 4.** The total mass of $^{235}\text{U}$ to achieve $k_{ef}$ on Design-A.
Figures 4 shows that even without a single fuel rod, a criticality condition is achieved with a $k_{\text{eff}}$ value of 1.0214, with a volume of 29097.828 cm$^3$ or 29.0978 L. This value was in good accordance compared with a similar model previously done by other researchers [6,11].

Because Design-B was designed to have a maximum of 24 fuel rods at a diameter of 31.4 cm, and a lower density of Th-UN is easier to manufacture, the author believed that the best configuration for this design is to use 24 fuel rods by using 300 g Th-U/L with a $k_{\text{eff}}$ value of 1.0562. The reactivity excess of this design, which is 5.32%, will come handy, while the low density of Th-U will make it easier to manufacture.

5. Conclusions
The best CAMOLYP core configurations for design-A, which is an annular cylindrical reactor core with a minimum diameter of 44 cm, provided by the outer ring filled with TRIGA wt-12%, while the best configuration for Design-B is by using 24 fuel rods with a Th-U density of 300 g/L. The amount of $^{235}$U in the first core model is 3376.74 g, while the second model is 1724.046 g. Based on the total amount of $^{235}$U needed to achieve criticality, the author concluded that the best design for CAMOLYP is Design-B. A further study to determine the conversion ratio is needed in order to evaluate the best design for both models.

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