Estimation of weekly $^{99}$Mo production by AHR 200 kW

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Abstract. The estimation of weekly $^{99}$Mo production by AHR 200 kW fueled with Low Enriched Uranium Uranyl Nitrate solution has been simulated by using MCNPX computer code. We have employed the AHR design of Babcock & Wilcox Medical Isotope Production System with $^9$Be Reflector and Stainless steel vessel. We found that when the concentration of uranium in the fresh fuel was 108 gr U/L of UO$_2$(NO$_3$)$_2$ fuel solution, the multiplication factor was 1.0517. The $^{99}$Mo concentration reached saturated at tenth day operation. The AHR can produce approximately $1.96 \times 10^3$ 6-day-Ci weekly.

1. Introduction
Isotope $^{99m}$Tc is produced from $^{99}$Mo undergoing beta decay having half-life of about 66 hours with approximately 88% of probability. Isotope $^{99m}$Tc is the most widely used radionuclide worldwide for nuclear medicine tomographic imaging technique using single photon emission computed tomography (SPECT). It’s used approximately 85% of nuclear medicine or about 12,000 six-day curies per week. The global demand for $^{99m}$Tc grows at an average annual rate 3%-8% in the new market, especially in Asia [1,2]. According to Pasqualini’s report on 2011 in “33rd International Meeting on Reduced Enrichment for Research and Test Reactor”, the major production of $^{99}$Mo around the world is from ten heterogeneous nuclear reactors. However, majority of these reactors have been working for more than 40 years. These reactors are listed in Table 1 [3].

Actually there are several method to produce $^{99}$Mo. According to Wolterbeek et al. [4], at least there are eight possible production routes. However, only the neutron-fission route is the most effective [4]. Comparing to heterogen reactor, Aquaeous Homogeneous Reactor (AHR) offers a better method with several advantages, namely low cost, small critical mass, inherent passive safety, simply fuel handling and also simple processing and purification characteristic [5]. In this paper we reported a simulation of estimation of weekly $^{99}$Mo production by AHR. The reactor was assumed operated at 200 kW and the solution was 19.75% uranil nitrate enriched.

2. Experimental Method
In this research we used MCNPX computer code to simulate AHR design and find out the interval time of saturation of $^{99}$Mo production. MCNPX is a general purpose Monte Carlo radiation transport code that design to track many particle types in the large range energies. This version is the next generation of Monte Carlo series that began in Los Alamos National Laboratory since 60 years ago.
General equation base on Monte Carlo calculation in this software have been constructed as shown in equation (1) [7].

\[
\frac{dX_i}{dt} = \sum_{j=1}^{N} l_{ij} \lambda_j X_j + \Phi \sum_{k=1}^{N} f_{ik} \sigma_k X_k - (\lambda_i + \Phi \sigma_i + r_i) X_i + F_i, \quad i = 1, ..., N
\]  

where:
- \( X_i \) = atom density of nuclide \( i \);
- \( N \) = number of nuclides;
- \( l_{ij} \) = fraction of radioactive disintegration by nuclide \( j \) which leads to formation of nuclide \( i \);
- \( \lambda_j \) = radioactive decay constant;
- \( \Phi \) = position and energy averaged neutron flux;
- \( f_{ik} \) = fraction of neutron absorption by nuclide \( k \) which leads to formation of nuclide \( i \);
- \( \sigma_k \) = spectrum averaged neutron absorption cross section of nuclide \( k \);
- \( r_i \) = continuous removal rate of nuclide \( i \) from the system;
- \( F_i \) = continuous feed rate of nuclide \( i \).

\[
\Phi = \frac{6.242 \times 10^{18} \cdot P}{\sum_{i=1}^{N} X_i \cdot \sigma_i^f \cdot R_i}
\]  

where
- \( \Phi \) = instantaneous neutron flux (n.cm\(^{-2}\).s\(^{-1}\))
- \( P \) = power (MW)
- \( X_i^f \) = amount of fissile nuclide \( i \) in fuel (g. atom)
- \( \sigma_i^f \) = microscopic fission cross section for nuclide \( i \) (barn)
- \( R_i \) = recoverable energy per fission for nuclide \( i \) (MeV/fission)

The input of MCNPX consists of a reactor core geometry specification, a source definition, and some tallies. The reactor core parameters are shown in Table 2. The fuel solution consists of 19.75% enrichment uranyl nitrate dissolved in water. The concentrations of the solution are varied from 75 up to 120 gr-U/L. The criticality of the AHR were calculated for each concentration solutions.

### Table 1. Major current \(^{99}\)Mo production reactor was working more than 40 years [3]

| Reactor | Country         | Annual Operating days | Production 6-day curies/week | Weekly % of world demand | Fuel/Target | Commissioning year |
|---------|-----------------|-----------------------|-----------------------------|--------------------------|-------------|--------------------|
| BR-2    | Belgium         | 140                   | 5200                        | 25-65                    | HEU/HEU     | 1961               |
| HFR     | Netherlands     | 300                   | 4680                        | 35-70                    | LEU/HEU     | 1961               |
| LVR-15  | Czech Republic  | Just Started          | >600                        | No data                  | HEU/HEU     | 1957               |
| MANGIA  | Poland          | Just Started          | 700-1500                    | No data                  | HEU/HEU     | 1974               |
| NRU     | Canada          | 300                   | 4680                        | 35-70                    | LEU/HEU     | 1957               |
| OPAL    | Australia       | 290                   | 1000-1500                   | No data                  | LEU/LEU     | 2007               |
| OSIRIS  | France          | 180                   | 1200                        | 10-20                    | LEU/HEU     | 1966               |
| SAFARI-1| South Africa    | 305                   | 2500                        | 10-30                    | LEU/HEU     | 1965               |
| RA-3    | Argentina       | 230                   | 200                         | < 2                      | LEU/LEU     | 1967               |
| RSG-GAS | Indonesia       | 147                   | 150                         | < 2                      | HEU/LEU     | 1967               |
Table 2. Reactor core parameters

| Parameter          | Value                                      |
|--------------------|--------------------------------------------|
| Reactor type       | AHR                                        |
| Reactor power      | 200 kW (thermal)                           |
| Enrichment         | 19.75 wt%                                  |
| Chemical           | UO$_2$(NO$_3$)$_2$ liquid                   |
| Temperature        | 80°C                                       |
| Reactor height (cm)| 122                                        |
| Core diameter (cm) | 63.4                                       |
| Reactor vessel     | Stainless steel - 304                      |
| Vessel thickness (cm)| 3.0                                      |
| Reflector          | Beryllium (radial)                         |
| Reflector thickness (cm)| 3.0                                    |

Since there are many kind of stainless steel, in this report we used stainless steel 304 for the reactor vessel material. The characteristics of the 304 SS are shown in Table 3.

Table 3. Reactor vessel densities [8]

| Stainless Steel Nuclide | Atom/barn.cm x 10$^{-2}$ |
|-------------------------|--------------------------|
| Cromium (Cr)            | 1.74                     |
| Mangan (Mn)             | 1.52                     |
| Besi (Fe)               | 5.81                     |
| Nikel (Ni)              | 0.851                    |

The mathematical model of the AHR core is shown in Figure 1.

Figure 1. Geometrical model of AHR. Left: Side view. Right: Top view

The colors in the Figure 1 are code for materials:
- Green: Air surrounding the AHR core
- Blue: Stainless steel: the AHR core vessel
- Red: Fuel UO$_2$(NO$_3$)$_2$ solution
- Yellow: Reflector Be
3. Results and Discussion

In Table 4 we show the values of $k_{\text{eff}}$ for each fuel densities. It can be seen clearly that the criticality of the AHR depends on the uranium concentration present in the fuel solution. The $k_{\text{eff}}$ increases monotonically with the uranium concentration, ranging from 0.90685 up to 1.05885. It is generally accepted that the $k_{\text{eff}}$ value of any reactor should be around 1.052. Accordingly, we decided to use the concentration of uranium was 108 gr-U/L as the best choice. We apply this value to calculate the production of $^{99}$Mo in various interval time from 1 up to 40 days. The results are presented in Figure 2.

| Enrichment 19.75% |
|-------------------|
| No | gr-U/L | $k_{\text{eff}}$ |
|-----|--------|------------------|
| 1   | 75     | 0.90685          |
| 2   | 80     | 0.93446          |
| 3   | 85     | 0.95826          |
| 4   | 90     | 0.98107          |
| 5   | 95     | 1.00473          |
| 6   | 100    | 1.0232           |
| 7   | 105    | 1.04189          |
| 8   | 106    | 1.04614          |
| 9   | 107    | 1.05003          |
| 10  | 108    | 1.0517           |
| 11  | 109    | 1.05552          |
| 12  | 110    | 1.05885          |
| 13  | 115    | 1.07653          |
| 14  | 120    | 1.09139          |

Figure 2. $^{99}$Mo production in interval various time
It can be seen from the figure 2, the $^{99}$Mo production increases over the operation time and reached saturated after ten days operation. In other words, if the reactor is operated up to 40 days, the production of $^{99}$Mo does not grow any more. The rate of $^{99}$Mo production is in balance with its decays. Thus the $^{99}$Mo should be extracted from the fuel solution before day-10th. More importantly, this simulation showed that the 200 kW AHR could produce $^{99}$Mo approximately $1.96 \times 10^3$ Ci 6-days weekly.

After the reactor has been operated over period time, the concentration of the fissile nuclide $^{235}$U decreases. It means the criticality of the reactor became lower than the original value. The $k_{\text{eff}}$ of the AHR should not be lower than 1 at any time. Therefore, it is instructive to calculate the $k_{\text{eff}}$ values over period time of operation. The calculated $k_{\text{eff}}$ is presented on Figure 3. It can be seen, although the $k_{\text{eff}}$ decreases however its value is still higher than 1. It means after the reactor has been operated for 40 days, the operator should not add the fuel.

![Figure 3. Multiplication factor during operation](image-url)

4. Conclusion
The simulation of AHR by MCNPX code shows the reactor was critical during operation. The concentration of uranium in fuel solution shows 108 gr U/L impact to critical of reactor, with the multiplication factor about 1.0517. It implies that we have successfully simulated the weekly production $^{99}$Mo. Our AHR model when fueled with 108 gr U/L $\text{UO}_2(\text{NO}_3)_2$ solution and operated at 200 kW yields about $1.96 \times 10^3$ 6-days Ci.

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