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 1.96×10^3 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, especialy 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

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[6]. General equation base on Monte Carlo calculation in this software have been constructed as shown in equation (1) [7].

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	Commis-sioning 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
MARIA	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

$$\frac{dX_i}{dt} = \sum_{j=1}^{N} l_{ij}\,\lambda_j X_j + \emptyset \sum_{k=1}^{N} f_{ik}\sigma_k X_k - (\lambda_i + \emptyset\sigma_i + r_i)X_i + F_i, \qquad i = 1, \ldots, N \tag{1}$$

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*;
λ_j = radioactive decay constant;
$\emptyset$ = position and energy averaged neutron flux;
f_{ik} = fraction of neutron absorption by nuclide *k* which leads to formation of nuclide *i*;
σ_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*.

$$\phi = \frac{6.242 \times 10^{18}.P}{\sum_{i=1}^{N} X_i^f\ \sigma_i^f\ R_i} \tag{2}$$

where
$\emptyset$ = instantaneous neutron flux (n.cm^{-2}. s^{-1})
P = power (MW)
X_i^f = amount of fissile nuclide *i* in fuel (g. atom)
σ_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 critycallity of the AHR were calculated for each concentration solutions.

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.

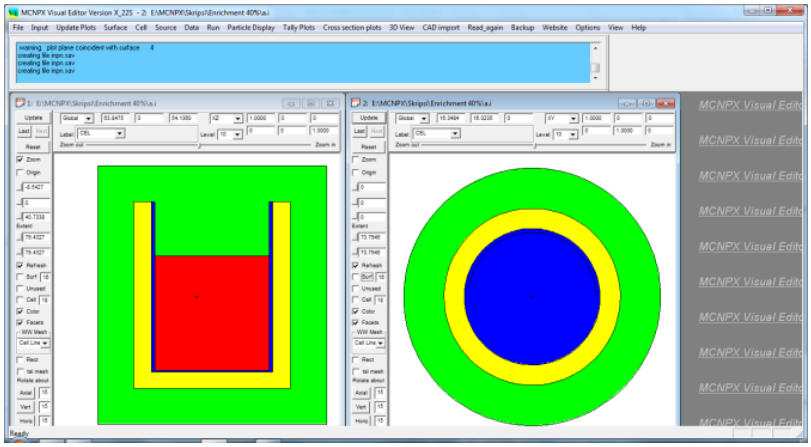


Figure 1. Geometrical model of AHR. Left: Side view. Right: Top view

The colors in the Figure 1 are code for materials:

- Air surounding the AHR core
- Stainless steel: the AHR core vessel
- Fuel $UO_2(NO_3)_2$ solution
- Reflector Be

3. Results and Discussion

In Table 4 we show the values of k_{eff} for each fuel densities. It can be seen clearly that the criticality of the AHR depends on the uranium concentration presents in the fuel solution. The k_{eff} increases monotonically with the uranium concentration, ranging from 0.90685 up to 1.05885. It is generally accepted that the k_{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.

Table 4. The k_{eff} values of fresh fuel densities

Enrichment 19,75%		
No	gr-U/L	k_{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

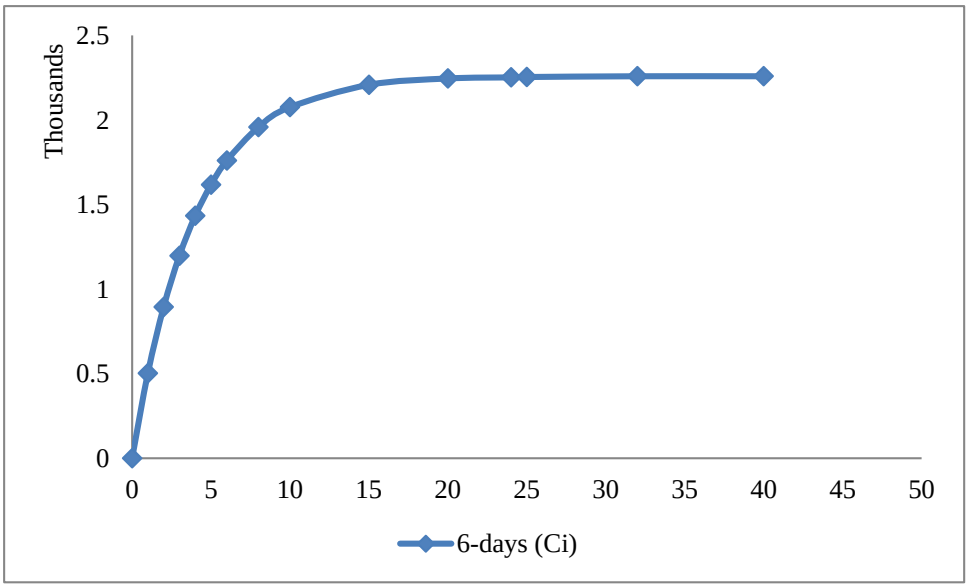


Figure 2. ^{99}Mo production in interval various time

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 it's 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 1.96×10^3 Ci 6-days weekly.

After the reactor has been operated over periode time, the concentration of the fissile nuclide ^{235}U decreases. It means the criticallity of the reactor became lower than the original value. The k_{eff} of the AHR should not be lower than 1 at any time. Therefore, it is instructive to calculate the k_{eff} values over periode time of operation. The calculated k_{eff} is presented on Figure 3. It can be seen, althought the k_{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.

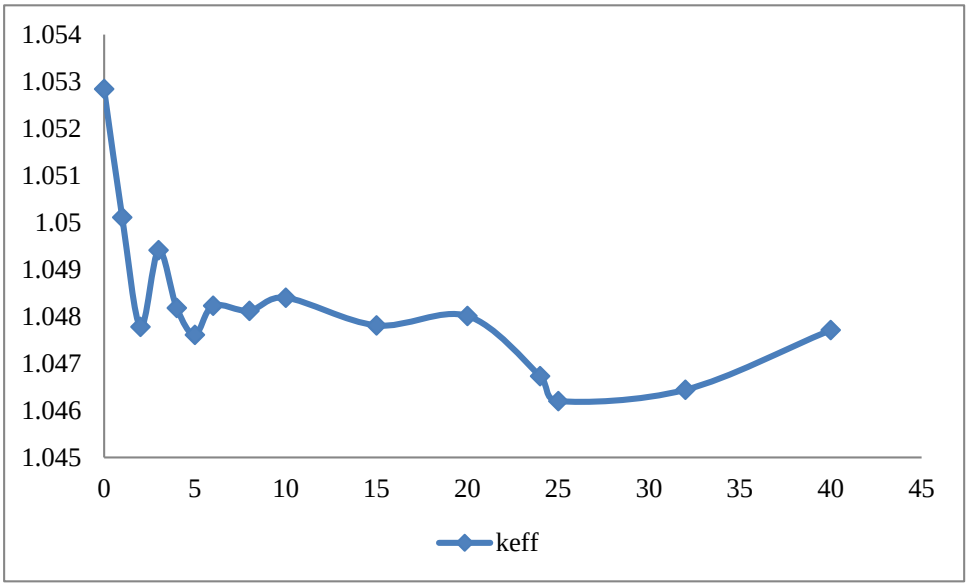


Figure 3. Multiplication factor during operation

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 $UO_2(NO_3)_2$ solution and operated at 200 kW yields about 1.96×10^3 6-days Ci.

5. References

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