Analysis of ^{99}Mo Production Capacity in Uranyl Nitrate Aqueous Homogeneous Reactor using ORIGEN and MCNP

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ABSTRACT

^{99m}Tc is a very useful radioisotope in medical diagnostic procedure. ^{99m}Tc is produced from ^{99}Mo decay. Currently, most of ^{99}Mo is produced by irradiating ^{235}U in the nuclear reactor. ^{99}Mo mostly results from the fission reaction of ^{235}U targets with a fission yield about 6.1%. A small additional amount is created from ^{98}Mo neutron activation. Actually ^{99}Mo is also created in the reactor fuel, but usually we do not extract it. The fuel will become spent fuel which is a highly radioactive waste. ^{99}Mo production system in the aqueous homogeneous reactor offers a better method, because all of the ^{99}Mo can be extracted from the fuel solution. Fresh reactor fuel solution consists of uranyl nitrate dissolved in water. There is no separation of target and fuel in an aqueous homogeneous reactor where target and fuel become one liquid solution, and there is no spent fuel generated from this reactor. Simulation of the extraction process is performed while reactor in operation (without reactor shutdown). With an extraction flow rate of 3.6 L/h, after 43 hours of reactor operation the production of ^{99}Mo is relatively constant at about 98.6 curie/hour.

INTRODUCTION

^{99m}Tc is a very useful radioisotope in medical diagnostic procedure; it is used in nearly 80% of all nuclear medicine procedures [1]. ^{99m}Tc is produced from ^{99}Mo decay. Because of the short half-life of ^{99m}Tc (6.0058 hours) it is not transported to hospitals around the world, but ^{99}Mo which has a longer half life (65.94 hours) is delivered instead. The total production and consumption of ^{99}Mo in the world is approximately 400 TBq/week [2]. Global demand for ^{99m}Tc is expected to grow at an average annual rate of 3–8% [3].

The reduction of uranium enrichment from ~90% to ~19.8% demands modifications on process operation to compensate for the resulting loss in output [4]. The reduction of uranium enrichment is due to nuclear non-proliferation.

Currently most of ^{99}Mo is being produced in research and test reactors by the irradiation of targets containing enriched fissile material ^{235}U [5]. ^{99}Mo is extracted using an acidic process [6]. This process generates waste. Figure 1 shows that ^{99}Mo is mostly produced from the fission reaction of ^{235}U targets with a fission yield about 6.1%, while a small additional amount is created from the neutron activation of ^{98}Mo which is itself created from ^{235}U target fission reaction. Actually ^{99}Mo is also created in the reactor fuel, but it is not usually extracted. The fuel will instead become a highly radioactive waste known as spent fuel.

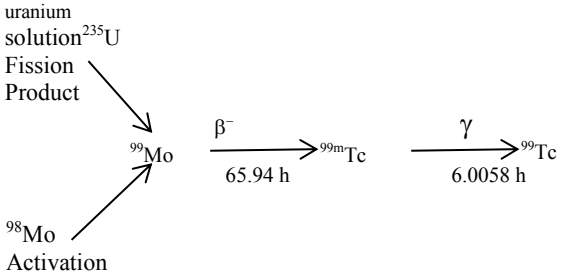

Fig. 1.^{99}Mo production.

^{99}Mo production system in the aqueous homogeneous reactor (AHR) offers a better method, because all of the ^{99}Mo can be extracted from the reactor solution. One such aqueous homogeneous reactor was built in the past; it was operated almost daily as a neutron source from 1951 until its deactivation in 1974, after 23 years of safe, reliable operation [7]. Fresh reactor fuel solution consists of uranyl nitrate dissolved in water. The production of ^{99}Mo in the AHR follows the same nuclear reactions shown in Fig. 1. However, there is no separation of target and fuel in the AHR, as the target and the fuel become one liquid solution. Therefore, there is no

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spent fuel generated from this reactor after ^{99}Mo extraction from the reactor solution; the remains of the extraction results will be returned to the reactor core as the fuel solution for further operation.

The use of fuel solution reactors for the production of medical isotopes is potentially advantageous because of their characteristics such as: low cost; small critical mass (low power); simple fuel handling, processing and purification characteristics; and inherent passive safety [8]. The void volume created by bubbles in the solution core will introduce a strong negative reactivity feedback [9].

EXPERIMENTAL METHODS

We use MCNP5 for reactor criticality analysis and ORIGEN2.2 for nuclide inventory analysis. MCNP is a general-purpose Monte Carlo N–Particle code which can be used for neutron, photon, electron, or coupled neutron/photon/ electron transport and possesses the capability to calculate eigenvalues for critical systems [10]. ORIGEN is a computer code system for calculating the buildup, decay, and processing of radioactive materials [11]. We couple both MCNP5 and ORIGEN2.2. We started by making MCNP input which consists of fuel, reactor vessel and reflector, then ran the input. The fuel solution height was gradually increased until the reactor became supercritical (MCNP output). After the reactor became supercritical we started to extract ^{99}Mo in a close loop. We extracted the ^{99}Mo from the fuel and then returned the remains of the extraction in the reactor core. The ^{99}Mo extraction was simulated in ORIGEN2.2. The simulation flow chart is shown in Fig. 2.

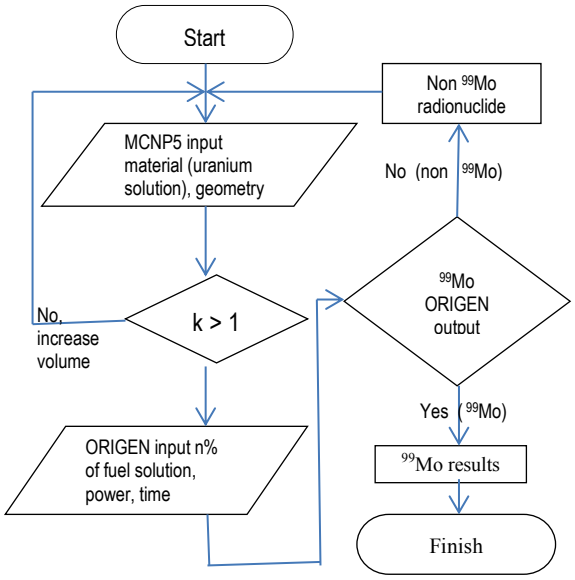

Fig. 2. Simulation flow chart.

The inputs of ORIGEN 2.2 are power (200 kW), time (one hour), and n % of fuel solution, where n % depends on the ratio of the fuel solution extracted per hour to all of the fuel solution; the ratio depends on the extraction flow rate and the volume of the fuel solution.

The ^{99}Mo extraction is capable of diverting 0.1 to 1.0 mL/second flow of uranyl nitrate solution [12]. The extraction flow rate that we used in this simulation is 1.0 mL/second (Table 1).

Table 1. Extraction flow rate.

	Flow rate(mL/s)	Flow rate(L/h)
Extraction flow rate	1	3.6

The reactor core parameters are shown in Table 2. The fuel solution consists of 20% enrichment uranyl nitrate dissolved in water. The atom densities of the fuel solution are shown in Table 3. The reactor vessel atom densities are shown in Table 4.

Table 2. Reactor core parameters.

Parameter	Value
Reactor power (thermal)	200 kW
Fuel solution	Uranyl nitrate
Enrichment	20%
Inner core diameter (cm)	30
Reactor height (cm)	100
Reactor vessel	Stainless steel-304
Vessel thickness (cm)	0.5
Reflector (radial)	Beryllium
Reflector thickness (cm)	30

Table 3. Atom densities in fresh fuel.

Isotope	atom/barn.cm
^{235}U	1.26504531144E-04
^{238}U	5.07525204789E-04
^{16}O	3.34878465916E-02
^{14}N	1.26805947187E-03
^{1}H	5.68312174084E-02

Table 4. Stainless steel-304 [13].

Nuclide	atom/barn.cm
Chromium	1.74×10^{-2}
Manganese	1.52×10^{-3}
Iron	5.81×10^{-2}
Nickel	8.51×10^{-3}

We used MCNP5 for criticality calculation. The geometrical model of the reactor on the Visual Editor of MCNP5 is shown in Fig. 3.

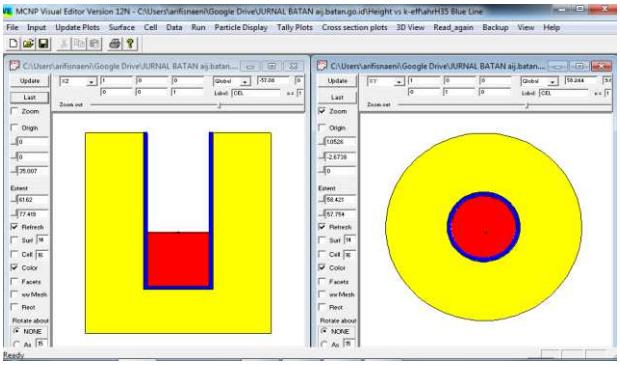

Fig. 3. Geometrical model of the reactor, from the side of the reactor (left) and from the top of the reactor (right). The reactor consists of uranyl nitrate solution (red), reactor vessel (blue) and beryllium reflector (yellow).

RESULTS AND DISCUSSION

The first step we performed was to obtain the condition where k_{eff}> 1 was barely achieved, or, in other words, to determine the height of the uranyl nitrate solution because k_{eff} is a function of fuel solution height. Using MCNP we made a variation of uranyl nitrate solution height. The result of the simulation is shown in Table 5.

Table 5. Fuel solution height vs k_{eff}.

No	Fuel solution height (cm)	k_{eff}
1	10	0.58154
2	15	0.75903
3	20	0.86390
4	25	0.93485
5	30	0.98914
6	35	1.02461
7	40	1.05394
8	45	1.06629
9	50	1.08568
10	55	1.10217
11	60	1.11266
12	65	1.11608
13	70	1.12535

From the result we decided to use k_{eff} = 1.02461 for which the height of uranyl nitrate solution in the core is 35 cm. Since the inner diameter of the core is 30 cm, the volume of the fuel solution is 24.7275 L. The extraction flow rate is 3.6 L/h. Therefore, the ratio of the fuel solution which is extracted per hour to all fuel solution in the core is:

$$\text{Ratio} = \frac{3.6}{24.7275} = 0.1459$$

Thus, 14.59% of all fuel solution is extracted per hour. ORIGEN2.2 simulates the fuel extraction. Of the ^{99}Mo in the fuel, 14.59% is extracted as the ^{99}Mo result per hour, while the other nuclide and 85.41% of ^{99}Mo remains in the core as the fuel solution. In this simulation this result becomes ORIGEN2.2 and MCNP5 input for the next hours. The resulting ^{99}Mo has to be decayed for one hour because this extraction process takes one hour. The ^{99}Mo result after the decay can be seen in Fig. 4.

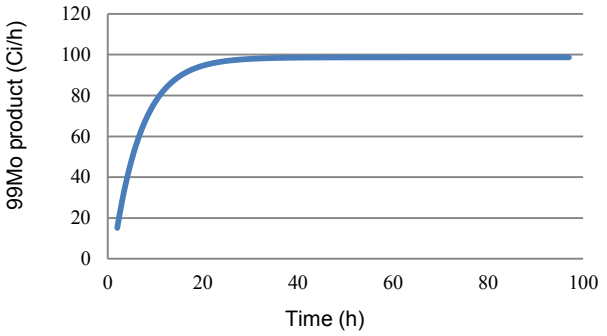

Fig. 4. ^{99}Mo production capacities per hour.

After 43 hours of reactor operation the production rate of ^{99}Mo is relatively constant at about 98.6 Ci/h. The ^{235}U in the fuel solution was consumed during reactor operation; the decreasing atom density of ^{235}U in the fuel can be seen in Fig. 5.

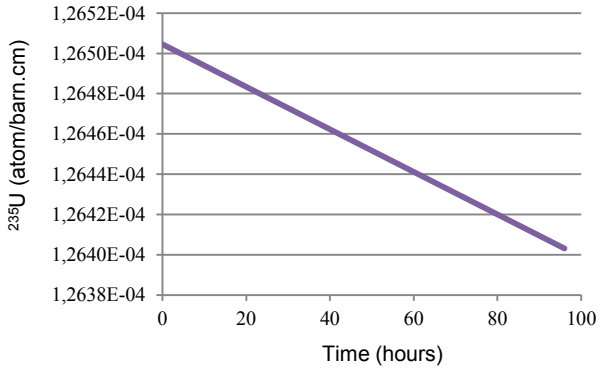

Fig. 5. ^{235}U atom density (atom/barn.cm).

Criticality analysis in MCNP5 shows that the reactor is supercritical for 96 hours of reactor operation. The k_{eff} during reactor operation is shown in Fig. 6.

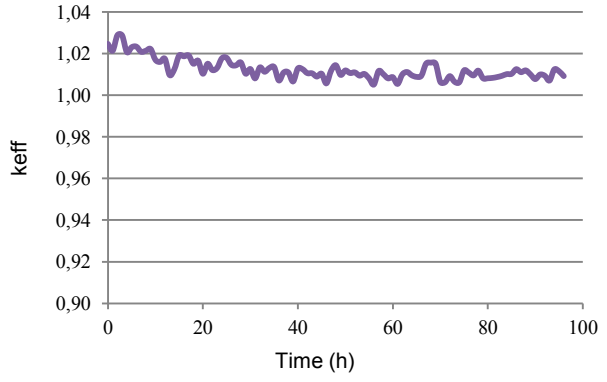

Fig. 6. k_{eff} during reactor operation.

Figure 7 shows the accumulation or total of ^{99}Mo product for 720 hours of reactor operation.

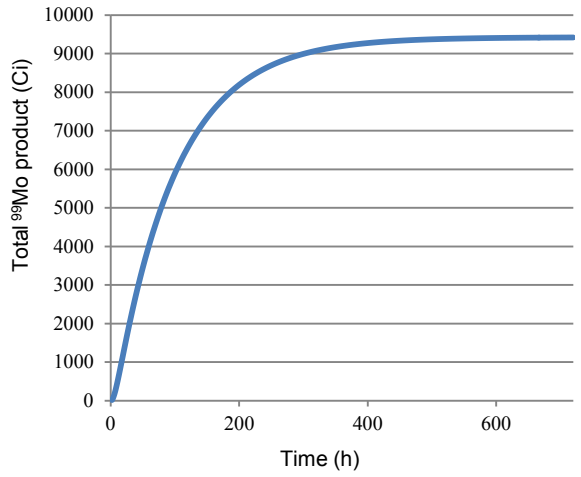

Fig. 7. Total ^{99}Mo Product (Ci).

The product accumulation is increasing with reactor operating time, but after reaching about 400 hours it relatively constant. Because the accumulation result of ^{99}Mo will decrease by decay process, it is better to extract the product as often as possible.

CONCLUSION

Reactor simulation on MCNP5 shows that the reactor is supercritical during reactor operation. The ^{99}Mo extraction is performed during reactor operation with a flow rate of 3.6 L/h. After 43 hours of reactor operation the ^{99}Mo production is relatively constant at about 98.6 Ci/h. Since the accumulation result of ^{99}Mo will decrease by decay process, it is better to extract the result as often as possible.

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