Innovative Compact Molten Salt Reactor (ICMSR) Analysis for Mo-99 Production

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Abstract. *Mo-99 isotope production calculation in the ICMSR (Innovative Compact Molten Salt Reactor) with the computer code MCNP6 has been carried out. ICMSR is a conceptual design of the MSR type reactor that uses NaF-ThF$_4$-UF$_4$ fuel with an enrichment of ^{235}U of 19.75%. This reactor operates in thermal neutron spectrum with a graphite moderator. ICMSR is a power reactor that produces Mo-99 as a by-product. Calculations carried out for 12 days of operation show that the reactor condition is still critical so that there will be no intervention from refueling. The total Mo-99 produced until the 12th day is 9.118 x 10^6 Ci. Mo-99 can be extracted from the reactor as long as the power reactor is operating so it will be economically advantageous.*

Keywords: ^{99}Mo, isotope production, ICMSR, MCNP, criticality

Introduction

Almost all hospitals in the world use radioisotopes in medicine. Most of it is for diagnosis. Technetium-99m ($T_{1/2} = 6.02\ h$) is the most widely used radioisotope [1]. More than 80% of radio-diagnostic procedures in the world use this isotope [2, 3, 4]. Technetium-99m can be generated from the decay of the parent nuclide Molybdenum-99 ($T_{1/2} = 66\ h$) [5]. The Mo-99 will decay to Tc-99m which then decays to its ground state Tc-99 by emitting 143 keV $\gamma-$rays which are known to have the same wavelength as diagnostic x-ray devices. The Tc-99m is ideal for diagnostic because of its short half-life so it is safe for patients [6]. However, this also makes it difficult to supply to medical centers. To solve the problem of sending Tc-99m to end user, a generator is used. A generator is a system that contains a radioactive parent nuclide with relatively long half-life that decays into short-lived daughter nuclides [7]. The generator used here refers to the Mo-99 generator.

The five main supply countries of Mo-99 are Canada, Belgium, France, Netherlands and South Africa. Most isotopes are generated from research reactors [8]. In 2008, shortages of Tc-99m began to emerge as the two Mo-99 reactors that supply 66% of the world's supply was shut down for maintenance. That reactor is National Research Universal (NRU) in Canada and Petten Nuclear Reactor (HFR) in Netherlands [9, 10]. Discontinuation of isotope supply from both reactors led to many delays and cancellations of medical procedures. The situation is getting worse: firstly, the reactor is old, secondly, using highly enriched uranium (HEU) which is not compatible with nonproliferation [11].

In order to meet the need of the domestic market, several research and development were carried out. One of the parties authorized to produce Mo-99 is PT INUKI. However, the production is not

sufficient to meet national needs [12]. From the results of the research reactor (RSG GAS), the production capacity of Mo-99 is 300 Ci per batch (week) [13]. The development of reactors to produce radioisotopes is also being carried out, i.e SAMOP and CAMOLYP [12, 14, 15].

An alternative technology that can support the fulfillment of the growing demand for Mo-99 is Molten Salt Reactor (MSR) [16]. The big advantage of using MSR for the production of Mo-99 is the possibility of online extraction [17]. In this study, we propose Innovative Compact Molten Salt Reactor (ICMSR). ICMSR is a power reactor, not a special reactor to produce Mo-99. However, during its operation, this reactor produces by-products, one of which is Mo-99. So this research was conducted to calculate and find out how much amount of Mo-99 can be produced from reactor operation for support reactors such as SAMOP and CAMOLYP to supply this isotope.

Theoretical Background

The production of Mo-99 in general can be carried out in two ways, namely: fission fragments from U-235 and neutron capture reactions from Mo-98. When the U-235 is hit by thermal neutrons, the atomic nucleus to become unstable and lose its shape, then split into smaller elements while releasing heat energy and releasing 2-3 new neutrons. One of those elements is Mo-99. In addition, neutron capture by M-98 can also produce Mo-99 accompanied by $\gamma -$ray emission. However, the production of Mo-99 from capture reaction is not used for commercial scale production because it is inefficient [18]. Thus, the production of this isotope depends on the fission reactions in nuclear reactors.

Molybdenum is considered a noble metal in molten salt so it is easily reduced in molten [19]. The reactor that uses molten salt is the Molten Salt Reactor (MSR). The ability of MSR to produce Mo-99 has been carried out and shows good results [20].

The accumulation of each isotope in a nuclear reactor depends on fission, decay, activation and extraction [17]. This can apply to any isotope, not just Mo-99.

Firstly is fission. The rate equation for the fission reaction can be written as:

$$\hat{R}^f \times Y_i = FP \ generation \ rate \tag{1}$$

Where, $\hat{R}^f = \frac{power \ density}{energy \ release/fission}$, Y_i is the fission product yield per fission. The subscript i refers to the isotope of interest.

Secondly is decay. The decay equation can be divided into two, namely the decay of the parent isotope and the decay of the isotope of interest. The generation of isotopes from parent isotopes can be written as:

$$\sum_p \beta_p \lambda_p N_p = decay \ of \ parent \ isotopes \tag{2}$$

Where, β is the branch fraction, λ is the decay constant and N is the number density. The subscript p refers to the parent isotopes. This equation is especially important for isotopes which have short half-lives. This isotope will remove and decay into its daughter isotopes. The decay of

the isotope of interest is also needed if isotope unstable and decay. This equation can be written as:

$$-\lambda_i N_i = decay\ of\ the\ isotope\ of\ interest \tag{3}$$

Where, λ is the decay constant and N is the number density. The subscript i refers to the isotope of interest.

Thirdly is activation. Activation occurs when a neutron is absorbed by an isotope. Activation of a nuclear reaction depends on the type of isotope, the energy of the neutron and the isotopic number density. In reactors where the fuel is considered homogeneous and has uniform neutron energy such as MSR, the activation equation can be approximated by the following equation

$$\hat{R}_i^x = N_i\sigma_i^x\phi = reaction\ rate \tag{4}$$

Where, $\hat{R}$ is reaction rate, σ is microscopic cross section, ϕ is neutron flux. The superscript x refers to nuclear reaction of interest.

The neutron flux in this equation can be eliminated by looking at the relationship between the reaction rates of two different isotopes by assuming that the flux is mono energetic.

$$\frac{\hat{R}_i^x}{N_i\sigma_i^x} = \frac{\hat{R}_j^y}{N_j\sigma_j^y} = \phi \tag{5}$$

So the activation equation can be rewritten as follows:

$$\hat{R}_i^x = \frac{N_i\sigma_i^x}{N_j\sigma_j^y}\hat{R}_j^y \tag{6}$$

The nuclear reaction we want to observe on activation is an isotope capture reaction in a fission fuel, so the equation becomes:

$$\hat{R}_i^c = \frac{N_i\sigma_i^c}{N_{fuel}\sigma_{fuel}^f}\hat{R}^f \tag{7}$$

Where, superscript c refers to capture reaction and superscript f refers to fission reaction.

Activation can do two things: generate isotopes and remove isotopes. Equation (7) refers to removal of the isotope due to activation. Meanwhile, the generation of isotopes produced from the activation product precursors can be written by replacing the subscript i with the subscript a. The total activation equation is as follows:

$$-\frac{N_i\sigma_i^c}{N_{fuel}\sigma_{fuel}^f}\hat{R}^f + \frac{N_a\sigma_a^c}{N_{fuel}\sigma_{fuel}^f}\hat{R}^f = total\ activation\ density \tag{8}$$

Later the isotope of interest will be extracted, the extraction of isotopes such as Mo-99 which is classified as a noble metal fulfills the equation

$$-k_c A N_i = extraction \tag{9}$$

Where, k_c is the mass transfer coefficient, A is the surface area available for extraction and N is number density.

So that the desired isotope accumulation equation in the reactor can be written:

$$\frac{dN_i}{dt} = \hat{R}^f\, Y_i - \lambda_i N_i + \sum_p \beta_p \lambda_p N_p - \frac{N_i \sigma_i^c}{N_{fuel} \sigma_{fuel}^f} \hat{R}^f + \frac{N_a \sigma_a^c}{N_{fuel} \sigma_{fuel}^f} \hat{R}^f - k_c A N_i \tag{10}$$

Materials and Methods

The type of MSR proposed in this study is the conceptual design of ICMSR (Innovative Compact Molten Salt Reactor). ICMSR is a power reactor with $NaF - ThF_4 - UF_4$ fuel. ICMSR uses 19.75% uranium enrichment to avoid proliferation. The core of this reactor is a HeteType core, which is a heterogeneous graphite fuel and moderator layout, allowing molten salt to flow through many graphite blocks. The HeteType core design has the advantage that it requires the smallest initial fuel loading and the lowest radiotoxicity of the remaining spent fuel in long term operation [16]. In the ICMSR design, the heat exchanger (HE) is integrated with the core, the aim is to reduce fuel inventory in the core and reduce the size of the cooling system. ICMSR is equipped with a noble and volatile gas extraction system. Noble gases can be removed by purging an inert gas into the molten salt. With the extraction system, the utility of the neutron is higher because neutron poisons such as Xe-135 no longer exist. Higher neutron utility means more collisions with uranium-235, resulting in higher production of fission products such as Mo-99. ICMSR parameter and geometry is show in Table 1 and Figure 1.

Table 1. Input parameter of ICMSR

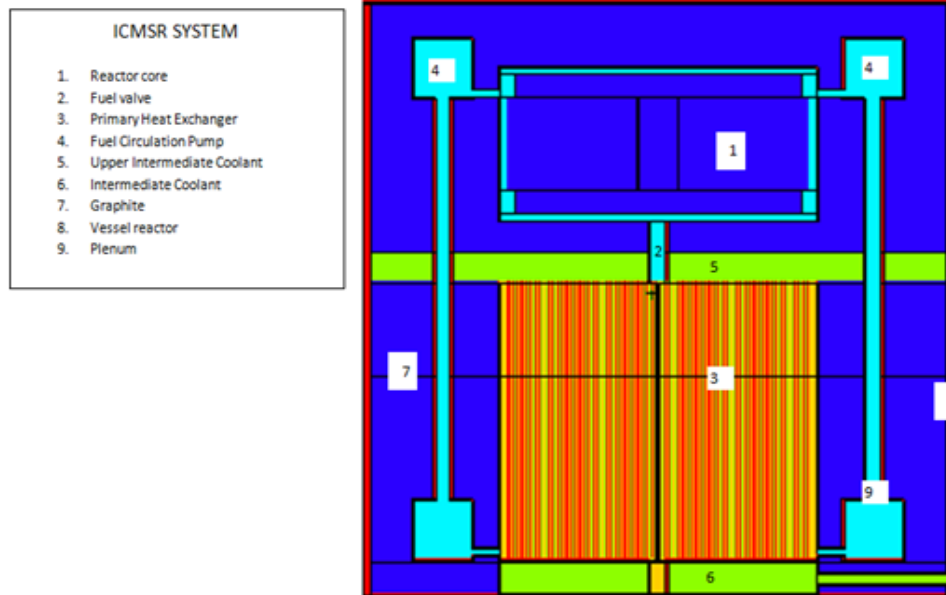| Parameter	Value (unit)
Power	75 MWe; 187.5 MWth
Temperature (during operation)	900K
Fuel	NaF-ThF_4-UF_4
U-235 enrichment	19.75%
Core diameter; high	220 cm; 246 cm
Moderator	Graphite
Moderator diameter; high	28 cm; 150 cm
Intermediate coolant	NaF-KF
Vessel reactor	Hastelloy-N
Vessel diameter; high	400 cm; 10.51 m

Figure 1. Configuration of modular ICMSR system

The fuel depletion analysis was carried out for 12 days. The scenario of fuel depletion by removing gas fission products is carried out with the OMIT command. The analysis will only carried out at a power of 75 MWe (thermodynamic efficiency = 40%) where this value corresponds to the operating power of the ICMSR. Although in this reactor operation all data on isotope depletion information is available, this paper will only focus on the production of Mo-99. The flowchart of this study is shown in Figure 2.
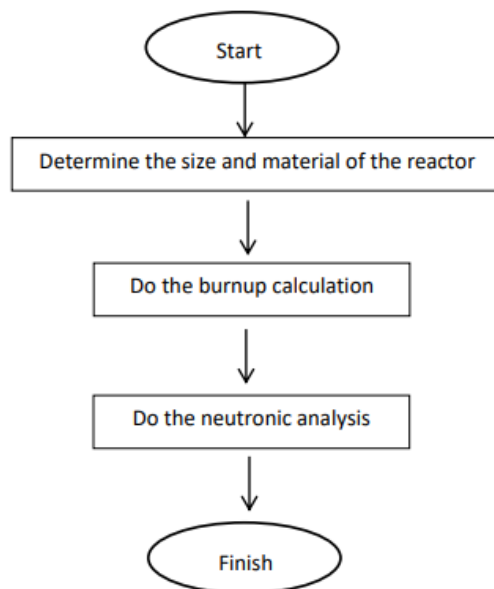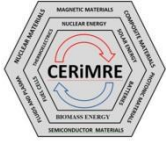

Figure 2. Flowchart of the study

The method used for calculation of the Mo-99 isotope is a computation method. This study has used MCNP6. The MCNP (Monte Carlo N Particle) is a computer program that developed since 1963 at Los Alamos National Laboratory (LANL), United States. MCNP6 is the latest version of MCNP which has been successfully developed by LANL. MCNP6 is the result of merging MCNP5 and MCNPX into a single computer code program that contains the features of both. In addition, MCNP6 also includes new calculation capabilities such as calculation of reactor kinetic parameters, unstructured Tally FMESH, and sensitivity analysis card [21].

The monte carlo method is a calculation method that theoretically imitates a statistical or random process. This method is usually used to solve complex problems, which can no longer be solved by deterministic calculation methods. This method is implemented by simulating every single probabilistic event that occurs in a process, one by one in sequence. The probability distribution that occurs in each event is calculated randomly. In this method, it takes a lot of repetition, so that the whole phenomenon being simulated can be fully and realistically described. Multiplication factor calculation in MCNP can be done with several estimators [22]:

a. Collision estimator

$$k_{eff}^{C} = \frac{1}{N} \Sigma_i W_i \left[\frac{\Sigma_k f_k \bar{v}_k \sigma_{fk}}{\Sigma_k f_k \sigma_{Tk}} \right] \qquad (11)$$

Where, i is summed over all collision in a cycle where fission is possible; k is summed over all nuclides of the material involved in the ith collision; σ_{Tk} is total microscopic cross section; σ_{fk} is microscopic fission cross section; $\bar{v}_k$ is average number of prompt or total neutrons produced per fission by the collision nuclide at the incident energy; f_k is atomic fraction for nuclide k; N is nominal source size for cycle; W_i is weight of particle entering collision.

b. Absorption estimator

$$k_{eff}^{A} = \frac{1}{N} \Sigma_i W_i \bar{v}_k \frac{\sigma_{fk}}{\sigma_{ck} + \sigma_{fk}} \qquad (12)$$

Where, i is summed over each analog absorption event in the kth nuclide.

c. Track length estimators

$$k_{eff}^{TL} = \frac{1}{N} \Sigma_i W_i \rho \, d \, \Sigma_k f_k \bar{v}_k \sigma_{fk} \qquad (13)$$

Where; i is summed over all neutron trajectories; ρ is atomic density in the cell; d is the trajectory track length from the last event.
        The combination average of the three estimators is:

$$k_{eff}^{best} = \frac{\Sigma_{i,i \neq j \neq k} k_{eff}^{i} * \left(C_{jj} * C_{kk} - C_{jj} * C_{ik} - C_{kk} * C_{ij} + C_{jk} * (C_{ij} + C_{ik} - C_{jk}) \right)}{\Sigma_{i,i \neq j \neq k} C_{jj} * C_{kk} - C_{jj} * C_{ik} - C_{kk} * C_{ij} + C_{jk} * (C_{ij} + C_{ik} - C_{jk})} \qquad (14)$$

Where:
i = C, A, and TL

C_{ij} = covariance = $\frac{1}{M}\sum_m \quad k_{effm}^i k_{effm}^j - \left(\frac{1}{M}\sum_m k_{effm}^i\right)\left(\frac{1}{M}\sum_m k_{effm}^j\right)$

m is the index for the mth active cycle and M is the number of active cycles.

Results and Discussion

The burnup calculation with the extraction of noble gas for 12 days has been done. The results showed that during 12 days of operation, the reactor was still in critical condition. This indicates that there is no need to add fuel online at that time. This condition can be achieved with a mole fraction of UF4 of 5.6% with an enrichment of 19.75%. Basically, the higher the two values, the higher the criticality value. But keep in mind, that the reactor will be safe when the criticality value is 1 or the reactivity value is zero and continues to be rated constant. The results show that k_{eff} value is slightly above 1. This supercritical condition is still relatively reasonable where the excess reactivity can be used to increase power if necessary. The results of the criticality value during operation can be seen in Figure 3.
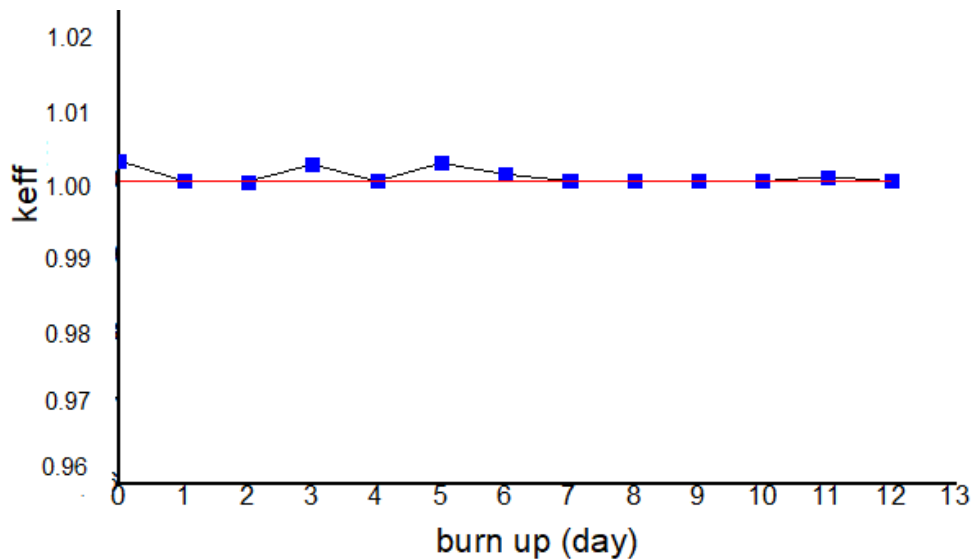

Figure 3.   The criticality levels per day of ICMSR reactor operation

Mo-99 is one of the major radionuclides in FPs. Table 2 shows the accumulation of Mo-99 in the ICMSR for 12 days. On day 0, Mo-99 has not yet formed; meaning there is only fuel in the reactor core. The highest increase in Mo-99 production was on the first day at 2.136 x 10^6 Ci. This production is the result of the fission of fuels. On six days, the resulting average is around 1.245 x 10^6 Ci, while on the 6th to 12th day the average generated is 0.275 x 10^6 Ci. The total Mo-99 produced until the 12th day is 9.118 x 10^6 Ci. In ICMSR, Mo-99 can be extracted during power reactor operation through fission product extraction. So it is very profitable from an economic point of view.

Table 2. Specific production of Mo-99 as by product of ICMSR power reactor

Day	Specific production (Ci)	Day	Specific production (Ci)
1	2.136×10^6	7	7.946×10^6
2	3.794×10^6	8	8.306×10^6
3	5.083×10^6	9	8.597×10^6
4	6.085×10^6	10	8.822×10^6
5	6.863×10^6	11	8.893×10^6
6	7.472×10^6	12	9.118×10^6

Conclusions

The criticality of the ICMSR reactor for 12 days of operation shows the rector is still in a critical condition so that there will be no intervention for refueling or power if Mo-99 extraction is carried out in that time span. Mo-99 production for six days of operation showed a high average of 1.245×10^6 Ci. Meanwhile, on the 6th to 12th day the average generated is 0.275×10^6 Ci. The total Mo-99 produced until the 12th day is 9.118×10^6 Ci. So it can be state that the ICMSR power reactor produces an isotope that is useful for medical purposes, namely Mo-99 as a by product.

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