Neutronics analysis of a subcritical blanket system driven by a gas dynamic trap-based fusion neutron source for 99Mo production postprint

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Abstract

Gamma-emitting radionuclide 99mTc is globally used for the diagnosis of various pathological conditions owing to its ideal single-photon emission computed tomography (SPECT) characteristics. However, the short half-life of 99mTc (T1/2 = 6h) makes it difficult to store or transport. Thus, the production of 99mTc is tied to its parent radionuclide 99Mo (T1/2 = 66h). The major production paths are based on accelerators and research reactors. The reactor process presents the potential for nuclear proliferation owing to its use of highly enriched U (HEU). Accelerator-based methods tend to use deuterium-tritium (D-T) neutron sources but are hindered by the high cost of tritium and its challenging operation. In this study, a new 99Mo production design was developed based on a deuterium-deuterium (D-D) gas dynamic trap fusion neutron source (GDT-FNS) and a subcritical blanket system (SBS) assembly with a low-enriched U (LEU) solution. GDT-FNS can provide a relatively high neutron intensity, which is one of the advantages of 99Mo production. We provide a Monte Carlo-based neutronics analysis covering the calculation of the subcritical multiplication factor (ks) of the SBS, optimization design for the reflector, shielding layer, and 99Mo production capacity. Other calculations, including the neutron flux and nuclear heating distributions, are also provided for an overall evaluation of the production system. The results demonstrated that the SBS meets the nuclear critical safety design requirement (ks<0.97) and maintained a high 99Mo production capacity. The proposed system can generate approximately 157 Ci 99Mo for a stable 24 h operation with a neutron intensity of 1$×$1014 n/s, which can meet 50% of China's demand in 2025.

Full Text

Preamble

Neutronics Analysis of a Subcritical Blanket System Driven by a Gas Dynamic Trap-Based Fusion Neutron Source for 99Mo Production

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Abstract

The gamma-emitting radionuclide 99mTc is globally utilized for diagnosing various pathological conditions due to its ideal single-photon emission computed tomography (SPECT) characteristics. However, the short half-life of 99mTc ($T_1/_2$ = 6 h) makes storage and transport challenging. Consequently, 99mTc production is tied to its parent radionuclide 99Mo ($T_1/_2$ = 66 h). The major production pathways rely on accelerators and research reactors, but reactor-based processes pose nuclear proliferation risks due to their use of highly enriched uranium (HEU). Accelerator-based methods typically employ deuterium-tritium (D-T) neutron sources, yet these are hindered by high tritium costs and operational complexities. This study proposes a novel 99Mo production design based on a deuterium-deuterium (D-D) gas dynamic trap fusion neutron source (GDT-FNS) coupled with a subcritical blanket system (SBS) assembly using a low-enriched uranium (LEU) solution. The GDT-FNS can provide relatively high neutron intensity, offering advantages for 99Mo production. We present a Monte Carlo-based neutronics analysis covering the calculation of the subcritical multiplication factor (k) of the SBS, optimization of the reflector and shielding layer designs, and assessment of 99Mo production capacity. Additional calculations, including neutron flux and nuclear heating distributions, are provided for comprehensive evaluation of the production system. The results demonstrate that the SBS meets nuclear critical safety design requirements (k < 0.97) while maintaining high 99Mo production capacity. The proposed system can generate approximately 157 Ci of 99Mo during stable 24-hour operation with a neutron intensity of 1$×10ˆ{14}$ n/s, which can meet 50% of China's projected demand in 2025.

Keywords: Gas dynamic trap, Fusion neutron source, Molybdenum-99, Low-enriched uranium, Subcritical blanket system

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1. Introduction

According to the World Nuclear Energy Association (WNA) [1], over 10,000 hospitals worldwide use radioisotopes for disease diagnosis and treatment, with approximately 90% of radioisotopes employed in diagnostic procedures. Among these, 99mTc is the most commonly used, benefiting from its ideal characteristics for single-photon emission computed tomography (SPECT). Further analysis indicates that 99mTc constitutes approximately 80% of nuclear medicine procedures, with 85% utilized for diagnostic scans (updated April 2022) [1]. However, 99mTc production ($T_1/_2 = 6$ h) is intrinsically linked to its parent radionuclide 99Mo ($T_1/_2 = 66$ h), making 99Mo production research an essential priority.

The 99mTc production process involves two steps: (1) 99Mo production via mechanisms illustrated in the schematic diagram of Fig. 1 [Figure 1: see original paper] (neutron-fission, gamma-fission, neutron-gamma, gamma-neutron, etc.), and (2) 99mTc separation following beta decay from 99Mo [2]. Although both 99Mo and 99mTc have short half-lives, the 66-hour half-life of 99Mo allows adequate time for transportation, unlike the 6-hour half-life of 99mTc that hinders its direct transport. Consequently, 99mTc production kinetics depend on 99Mo production; under normal circumstances, 99Mo is transported to target countries immediately after production, and its decay product (99mTc) is rapidly extracted for use in hospitals or nuclear medicine centers.

[Figure 1: see original paper]

The global shortage of 99Mo has recently been attributed primarily to aging nuclear reactors and their decommissioning [3-5]. A notable example is Canada's National Research Universal (NRU) reactor, which produced approximately 40% of the world's 99Mo supply before ceasing production on October 31, 2016 [3]. Currently, most 99Mo isotopes are produced through fission in HEU reactors, including the Belgian Reactor 2 (BR-2), High Flux Reactor (HFR) in the Netherlands, LVR-15 Reactor in the Czech Republic, Maria Research Reactor in Poland, Open Pool Australian Light Water Reactor (OPAL), and South African Fundamental Atomic Research Installation (Safari-1) [6]. Additional details regarding 99Mo production, specific reactors, and target materials are listed in Table 1 . Most of these reactors face not only aging and decommissioning issues but also significant nuclear proliferation risks.

[FIGURE:1 Schematic diagram of the 99mTc production process]

To address both the 99Mo shortage and HEU proliferation concerns, scientists have proposed alternative production methods that can be categorized into three approaches: (1) 235U(n,f)99Mo reactions in low-enriched uranium (LEU) reactors [7,8]; (2) solid target irradiation using accelerators, such as the neutron capture 98Mo(n,γ)99Mo reaction [9-13], 100Mo(n,2n)99Mo reaction [14,15], 100Mo(p,2n)99Mo reaction [16], 100Mo(γ,n)99Mo reaction [17], and photon-induced 238U fission 238U(γ,f)99Mo [18]; and (3) LEU solution fission via 235U(n,f)99Mo reactions in subcritical systems [19-23]. Among these, the third method represents the most efficient and reliable production approach and has become a prime candidate for 99Mo production due to several advantages: (1) compared to accelerator-based solid target irradiation, the LEU solution can be effectively recovered and reused, significantly reducing radioactive waste generation; (2) the LEU fission method offers high production efficiency at low cost; and (3) compared to HEU fission, the LEU solution in subcritical systems avoids criticality accidents and prevents nuclear proliferation while being relatively easier to license for construction and operation.

In 2021, Han et al. [22,23] proposed a subcritical 99Mo production system driven by an accelerator-based D-T neutron source, where accelerated deuterium ions bombard a tritium target to generate neutrons via deuterium-tritium (D-T) fusion reactions. The LEU solution target is then irradiated by these neutrons to induce 235U fission through the 235U(n,f)99Mo reaction. Although this method eliminates HEU requirements, it suffers from the disadvantages of tritium usage, including high costs and licensing difficulties for tritium ownership and operation.

This study proposes a novel 99Mo production design based on an LEU subcritical blanket system (SBS) driven by a gas dynamic trap-based fusion neutron source (GDT-FNS). Instead of conventional D-T fusion, the system employs deuterium-deuterium (D-D) fusion neutrons to induce 235U fission. Beyond avoiding HEU, our proposed 99Mo production system offers numerous advantages, including a compact structure, high neutron source intensity, and absence of tritium consumption, resulting in low capital costs. To ensure design safety and process optimization for the SBS 99Mo production pathway, we performed a neutronics analysis of the production system using the SuperMC Monte Carlo particle transport code. This analysis provides a comprehensive evaluation of the production system, including subcritical multiplication factor (k) analysis, neutron flux characterization, and heat deposition assessment. A set of optimized production system parameters was obtained while maintaining high production rates and safety standards, encompassing geometric dimensions, material compositions, and LEU solution concentrations.

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2. Model and Method

The LEU solution SBS driven by GDT-FNS primarily consists of the GDT-FNS and the 99Mo SBS, as illustrated in the schematic diagram of Fig. 2 [Figure 2: see original paper]. The SBS is positioned in the high neutron flux region of the GDT, forming a fan-shaped blanket structure. Detailed descriptions are provided in Sections 2.1 and 2.2.

[Figure 2: see original paper]

2.1 Gas Dynamic Trap-Based Fusion Neutron Source (GDT-FNS)

A GDT is an axisymmetric magnetic mirror device [24,25]. Under specific magnetic field configurations, warm plasma constrained within the GDT vacuum chamber undergoes frequent collisions that produce fusion reactions, providing either D-D or D-T fusion neutron sources characterized by "high flux at both ends and low flux in the middle." This neutron source architecture offers advantages of high neutron flux, large experimental space, compact structure, and low construction cost.

The GDT-FNS not only meets requirements for fusion materials and component testing but also enables applied nuclear technology research utilizing its high neutron flux, such as medical isotope production, neutron radiography, neutron irradiation (breeding), and low-dose neutron effects on cells.

In this study, we employed the GDT-FNS design developed by the Hefei Institute of Physical Science [26,27] to analyze the neutronics of a solution-based LEU SBS. Quasi-monoenergetic neutrons with approximately 2.5 MeV energy were generated through D-D reactions in the central vacuum system (CVS). As shown in Fig. 2, the GDT-FNS primarily comprises a neutral beam injection system (NBI), CVS, magnetic coils (MC), and a high neutron flux region. The main parameters for D-D operation are listed in Table 2 . Due to the axisymmetric characteristics of the GDT-FNS, SBS units with flexible configurations can be arranged in the high neutron flux region to meet increasing 99Mo production demands.

The plasma parameters of the GDT-FNS system were simulated using the 1-D DOL code [28], which is based on a nonstationary numerical model describing the confinement of two different plasma components. During simulations, a pure deuterium beam was injected into the central vacuum chamber, yielding axial distributions of the D-D neutron generation rate as shown in Fig. 3 [Figure 3: see original paper]. The results reveal two high neutron flux regions located between axial positions of -700 to -600 cm and 600 to 700 cm within the GDT-FNS. This important finding motivated the placement of the SBS in these high neutron flux regions.

[Figure 3: see original paper]

2.2 Subcritical Blanket System (SBS)

The 99Mo production SBS is positioned in the high neutron flux region of the GDT-FNS, forming a fan-shaped blanket structure. The primary material components include the LEU solution, solution container, reflector, and shielding layer. A schematic of the SBS 99Mo production model is shown in Fig. 4 [Figure 4: see original paper]. Based on preliminary analysis and considering FNS design constraints, certain geometric and material parameters were fixed, including dimensions and specific material compositions. The SBS length was set to 100 cm with thickness less than 100 cm. The LEU solution thickness ranged between 30-50 cm, and the solid angle of the LEU solution relative to the neutron source central axis was between $\pi/4$ and $\pi/3$. Variable parameters requiring optimization in this study include the thicknesses of the LEU solution, reflector, and shielding, as well as material types for the reflector and shielding layers. For the LEU solution, UO_2SO_4 solution was selected with uranium concentrations varying from 60 g/L to 150 g/L and a ^{235}U enrichment of 19.75%. The lower concentration limit (60 g/L) was chosen to achieve substantial 99Mo output, while the upper limit (150 g/L) corresponds to the saturated concentration of UO_2SO_4 at room temperature.

[Figure 4: see original paper]

2.4.1 Calculation Program and Uncertainty

The neutronics parameters of the SBS were calculated using Super Monte Carlo Simulation Program (SuperMC) version 3.2 [29] coupled with ENDF-VII cross-section libraries. The steady-state neutronics parameters of the SBS primarily included k , neutron energy spectrum, activity of produced 99Mo, and heat deposition. In this study, 10 million statistical particles were used for each neutronics calculation, yielding statistical uncertainties below 1%, except for energy deposition calculations where uncertainties were below 3%.

2.4.2 Subcritical Multiplication Factor

The critical safety state of a subcritical system can be characterized by the subcritical multiplication factor k when the uranium fission system has an externally driven neutron source. Parameter k is defined as the ratio of fission neutrons to total neutrons in the system [30,31], as shown in equation (1):

$$k_s = \frac{\nu R_f}{S + \nu R_f}$$

where S is the intensity of the externally driven neutron source [n/s], R_f is the fission reaction rate [fission/s], and ν is the average number of neutrons generated per fission reaction. Parameter k is typically required to be less than 0.98 to ensure operational safety of a subcritical system [32-36]. The k calculation was performed using a general source card (SDEF) with the energy spectrum obtained from the first wall of the GDT-FNS high neutron flux region, which served as the external driver neutron source.

2.4.3 Activity and Specific Activity of Produced 99Mo

To evaluate 99Mo production efficiency and uranium utilization, we defined the total activity of 99Mo produced by the SBS in one day (24-hour operation) as A [Ci/day], and the daily 99Mo produced per unit mass of ^{235}U as the specific activity (SA) [Ci/kg/day]. The 99Mo inventory increases through ^{235}U fission and decreases through radioactive decay. The number of 99Mo nuclides $N(t)$ at time t [s] evolves according to equation (2):

$$\frac{dN(t)}{dt} = Y_{99}\Sigma_f\Phi - \lambda N(t)$$

where $N(t)$ is the number of 99Mo nuclides at time t [s], λ is the 99Mo decay constant, Y_{99} is the 99Mo fission yield (0.061), Σ_f is the macroscopic fission cross-section of ^{235}U [barns], and Φ is the neutron flux [cm^{-2}s^{-1}].

By defining $A(t) = \lambda N(t)$ and integrating equation (2), we obtain the activity equation (3), which is consistent with the generalized activity equation [37]:

$$A(t) = Y_{99}\Sigma_f\Phi(1 - e^{-\lambda t})$$

However, in an SBS, the activity equation can be modified to yield equation (4):

$$A(t) = Y_{99}N_{235}\bar{\sigma}_f\frac{S}{V}(1 - e^{-\lambda t})$$

In this formulation, the neutron flux Φ is replaced by S/V, where S is the external neutron source intensity [n/s] from the GDT-FNS high neutron flux region and V is the volume. Σ_f equals $N_{235}\bar{\sigma}_f/V$, where $\bar{\sigma}_f$ [barns] is the average microscopic fission cross-section of a ^{235}U atom. N_{235} is the total number of ^{235}U atoms in the SBS, equal to mN_A/M, where N_A is Avogadro's constant (6.02$×10^{23}$), M is the relative atomic mass of ^{235}U (235), and m is the mass of ^{235}U [g] in the LEU solution. In this study, $\bar{\sigma}_f$ was calculated using tally card 4 and multiplier card FM4 in the input file. By substituting these values into equation (4), the daily 99Mo production in the SBS can be evaluated.

2.4.4 Neutron and Gamma Flux Calculation

Neutron and gamma flux can be calculated using tally 4 [38] by specifying different particle types [Neutron (N) or Photon (P)] through equation (5):

$$\Phi(\vec{r}, E, t) = \int_{4\pi} \psi(\vec{r}, \vec{\Omega}, E, t) d\vec{\Omega}$$

where Φ is the neutron or gamma flux at the point detector [particles/cm^2], ψ is the angular flux [particles/(cm$^3 \cdot$ sr $\cdot$ MeV $\cdot$ s)], $\vec{r}$ is the position vector [cm], E is the incident particle energy [MeV], $\vec{\Omega}$ is the direction vector, and t is time [s]. To evaluate the neutron flux distribution in the SBS, we introduce the heterogeneity coefficient K_H, defined as the ratio of the maximum core thermal neutron flux to the average value. K_H is generally required to be less than 1.4 in reactor design [39].

2.4.5 Nuclear Heat Deposition

Nuclear heat deposition was calculated using tally 6 (T6) combined with a superimposed mesh tally card (FMESH). The T6 calculation yields the average energy deposition in each computational cell, as shown in equation (6) [38]:

$$E_{dep} = \frac{1}{m} \int_E N \sigma_t(E) H(E) \Phi(E) dE$$

where E_{dep} is the total energy deposition in the cell [MeV/g], N is the atom density (10^{24} atoms/cm^3), m is the cell mass [g], $\sigma_t(E)$ is the microscopic total cross-section [barns], and $H(E)$ is the heating number [MeV/collision].

3.1 Neutron Spectrum of the High Neutron Flux Region of GDT-FNS

The neutron spectrum is a critical parameter of the GDT-FNS that affects fission reaction efficiency. In this study, the neutron generation rate (Fig. 3) was used as input data to calculate the neutron spectrum. The neutron spectrum in the plasma tube of the high-neutron-generation-rate region was obtained using the SuperMC program, with statistical errors maintained below 1%. The spectral distributions are shown in Fig. 5 [Figure 5: see original paper]. The results demonstrate that neutrons with approximately 2.5 MeV energy constitute the highest proportion, as expected from D-D reactions producing neutrons with an average energy of 2.5 MeV. This calculated neutron spectrum was subsequently used as the external driver neutron source for SBS neutronics design.

[Figure 5: see original paper]

3.2 Preliminary Design of the SBS

The SBS 99Mo production system aims to meet 50% of China's projected 99Mo demand by 2025. China currently relies heavily on 99Mo imports, with demand continuing to grow. The estimated medical 99Mo demand in China was approximately 16,000 6-day Ci in 2019 [40]. Considering an annual growth rate of 5% [5], the projected 99Mo demand in 2025 will be approximately 21,500 6-day Ci (equivalent to 59 6-day Ci per day). These estimates account for decay losses during 99Mo separation and purification, where approximately 80% of the originally produced 99Mo is lost during the 6-day period, and an additional 10% [41] cannot be extracted through chemical processes. Therefore, the daily 99Mo demand is estimated to be 298 Ci in 2025 and 447 Ci in 2035.

When determining the preliminary SBS design, the following constraints were considered: (1) To ensure nuclear critical safety of the LEU solution SBS, k must be less than 0.98 [32-36]. However, accounting for neutron source fluctuations, measurement uncertainties, and other factors, k was limited to less than 0.97 to provide sufficient safety margins. (2) Uranium concentration was constrained between 60-150 g/L, with the upper limit determined by UO_2SO_4 solubility at room temperature. (3) Specific activity (SA) should be maximized.

The initial calculation model conditions were: plasma tube inner diameter of 35 cm; LEU solution inner and outer radii between 37-74 cm; LEU solution container wall thickness of 1 cm; LEU solution solid angle between $\pi/4$-$\pi/3$; top and bottom reflector thickness of 5 cm; back reflector composed of Be material with 8 cm thickness; shielding layer thickness of 8 cm using W, B, and polyethylene (PE) composite (mass ratio 4:3:3). Parameters including k , activity A, and SA were calculated by varying uranium concentration and solid angle, with results listed in Table 3 .

As shown in Table 4 , Case 5 satisfies the critical safety condition (k < 0.97) with an activity A of 156 Ci, which closely approaches 50% of China's projected medical 99Mo demand for 2025 (149 Ci). Additionally, Case 5 exhibits the highest SA, indicating the most efficient ^{235}U utilization compared to other cases. Based on these results, Case 5 was selected as the preliminary scheme for subsequent optimization, including uranium concentration refinement and reflector/shielding material and sizing optimization.

3.3 Impact of Uranium Concentration on SBS Performance

To investigate the influence of various uranium concentrations on SBS 99Mo production performance, detailed calculations were performed using Case 5 conditions from Table 4 while varying uranium concentration. The LEU solution volume was fixed at 155.5 L with ^{235}U enrichment at 19.75%. The effects of varying uranium concentration on k and daily 99Mo production A are shown in Fig. 6 [Figure 6: see original paper].

[Figure 6: see original paper]

The results demonstrate that both k and A exhibit increasing dependence on uranium concentration. Specifically: (1) As expected, k shows strong dependence on uranium concentration, increasing sharply with concentration. At

110 g/L uranium concentration, k exceeds the design safety limit of 0.97. (2) While daily 99Mo production A increases gradually with uranium concentration, a sharp increase occurs above 90 g/L. (3) The most favorable condition for nuclear critical safety occurs when uranium concentration is just below 105 g/L, where k remains below 0.97. Therefore, the uranium concentration was set to 105 g/L in the final design model.

3.4 Reflector Optimization Design

The reflector, positioned outside the LEU solution, reflects neutrons back into the uranium solution to minimize leakage and improve neutron utilization. Reflector material selection and dimensions significantly influence 99Mo production efficiency and associated k values. Common reflector materials include Be metal, BeO, graphite (GR), heavy water (D_2O), zirconia (ZrO_2), among others. Using Case 5 parameters with an 8 cm back reflector thickness while holding other parameters constant, k values were calculated for various reflector materials, as shown in Fig. 7 [Figure 7: see original paper].

[Figure 7: see original paper]

The k values obtained with different reflector materials show a maximum for Be, indicating that beryllium provides the best reflection performance under the selected conditions. Therefore, Be was selected as the reflector material for the SBS design. Subsequent optimization calculations for k and A were performed by varying Be reflector thickness, with results shown in Fig. 8 [Figure 8: see original paper].

[Figure 8: see original paper]

The calculations demonstrate that k and A increase with Be reflector thickness, emphasizing the beneficial effect of improved neutron utilization on 99Mo production. However, the optimum reflector thickness is approximately 10 cm; beyond this thickness, only marginal increases in A are observed while k increases more significantly. Therefore, to balance capital cost, minimize overall SBS dimensions, and enhance critical safety, a 10 cm reflector thickness was selected as optimal.

3.5 Shielding Layer Optimization Design

A shielding layer is employed to reduce neutron and gamma radiation in the environment. Different materials exhibit varying shielding capabilities for neutrons and gamma rays. Eight material types were selected for shielding evaluation: Fe, Pb, W, Fe/B (weight ratio 1:1), W/B (weight ratio 1:1), Fe/PE (weight ratio 1:1), W/PE (weight ratio 1:1), and W/B/PE (weight ratio 4:3:3). Shielding performance was evaluated using an 8 cm thickness, with corresponding k values and average neutron fluxes outside the shielding layer calculated. The results are shown in Fig. 9 [Figure 9: see original paper].

[Figure 9: see original paper]

The results indicate that different shielding materials have minimal influence on k , with only a 0.0028 difference between maximum (0.9695) and minimum (0.9667) values. W/B/PE demonstrates the best neutron shielding performance, as expected because boron provides excellent thermal neutron absorption, polyethylene serves as an effective neutron moderator, and tungsten offers good gamma shielding. Considering combined neutron and gamma shielding performance and avoiding toxic lead, the W/B/PE composite was selected as the shielding material.

The influence of varying W/B/PE thickness on k and shielding performance was further investigated, with results shown in Fig. 10 [Figure 10: see original paper]. The k shows no significant change with increasing shielding thickness because the shielding material contains boron, which absorbs neutrons. However, both average neutron flux and gamma flux outside the shielding layer decrease with increasing shielding thickness.

[Figure 10: see original paper]

According to shielding design requirements, to limit radiation impact from thermal neutron-activated products, thermal neutron flux should be less than 1×10^{5} cm^{-2}s^{-1} and gamma flux should be less than 4×10^{10} cm^{-2}s^{-1} [42,43]. The results demonstrate that at a W/B/PE thickness of 15 cm, the average neutron flux outside the shielding layer is 4.31×10^{4} cm^{-2}s^{-1}, while the average gamma flux is 2.10×10^{7} cm^{-2}s^{-1} with k = 0.9685. These values satisfy both shielding design requirements and nuclear critical safety margins.

3.6 Neutron Flux Distribution in SBS

The radial and axial neutron flux distributions in the SBS were calculated, with results shown in Fig. 11 [Figure 11: see original paper] (a) and (b), respectively. The neutron flux in the uranium fission zone is on the order of 10^{11} n/cm$^2\cdot$s (average of 3.73×10^{11} n/cm$^2\cdot$s; peak value of 4.94×10^{11} n/cm$^2\cdot$s). The neutron flux decreases rapidly after passing through the reflector layer (i.e., in the shielding layer), confirming the effectiveness of the shielding materials. In the axial direction, the neutron flux remains relatively constant across the uranium solution but decreases sharply at both ends of the reflector layer. The average neutron flux was calculated to be 3.88×10^{11} n/cm$^2\cdot$s, with a maximum of 4.72×10^{11} n/cm$^2\cdot$s. As defined in Section 2.4.4, K_H can be calculated as $4.72\times10^{11}/3.88\times10^{11} = 1.22$, which meets design requirements ($K_H < 1.4$). This demonstrates that the radial and axial neutron flux distributions in the SBS are relatively uniform, which benefits both efficient ^{235}U utilization and safe system operation.

[Figure 11: see original paper]

The neutron energy spectrum characteristics of the uranium solution layer, outer uranium container, reflector layer, and shielding layer were calculated, as shown in Fig. 12 [Figure 12: see original paper]. The results reveal two thermal neutron peaks in the thermal region (10^{-8}–10^{-6} MeV) and a fast neutron peak at approximately 2.5 MeV. The thermal neutron peaks arise from H_2O in the uranium solution acting as a neutron moderator, thermalizing many neutrons. The fast neutron peak appears because the external neutrons driving the SBS are primarily 2.5 MeV neutrons from D-D reactions, with additional fast neutrons produced by ^{235}U fission.

[Figure 12: see original paper]

3.7 Nuclear Heat Distribution in SBS

To determine nuclear heat distribution in each SBS component, tally card T6 combined with FMESH was used to calculate nuclear heat deposition. The visualization function of SuperMC was employed to display results, as shown in Fig. 13 [Figure 13: see original paper] (a) and (b).

[Figure 13: see original paper]

The results demonstrate that nuclear heat is primarily deposited in the LEU solution, with a maximum heat deposition of 1.53×10^{-1} W/cm^3 at the central position and a minimum of 4.57×10^{-3} W/cm^3 at the edge. The average nuclear heat deposition is 7.01×10^{-2} W/cm^3, corresponding to a total nuclear heat of 10.9 kW. In the reflector layer, average nuclear heat deposition is 1.21×10^{-4} W/cm^3, while in the shielding layer it is 5.57×10^{-5} W/cm^3—approximately 2-3 orders of magnitude lower than in the LEU solution. This difference occurs because nuclear heat primarily originates from fission energy released by ^{235}U fission, whereas heat in the reflector and shielding layers results from neutron and gamma energy deposition, which is significantly lower than fission energy. COMSOL [44] was used to simulate the cooling system, revealing that the fuel solution could boil within 2 hours without cooling. The simulations also demonstrated that supplying cooling water at 22°C with a flow velocity of 1.0 m/s would be sufficient to maintain fuel solution temperature below 90°C.

Based on the aforementioned analysis, the optimized design parameters for 99Mo production by the GDT-FNS-driven SBS are summarized in Table 4.

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4. Conclusions

This study proposes an LEU SBS driven by GDT-FNS for 99Mo production and conducts a neutronics analysis of the production system using the Monte Carlo method (SuperMC code). The analysis includes calculation of the neutron spectrum in the high neutron generation rate region of the GDT-FNS, preliminary design and optimization assessments for various uranium concentrations and their 99Mo production activities, reflector and shielding layer optimization design, and neutron flux and nuclear heat distribution analysis of the SBS.

In all optimization cases, the designed system must meet safety requirements while producing sufficient 99Mo to satisfy 50% of China's projected 99Mo demand in 2025. The preliminary assessment, shown as Case 5 in Table 4, demonstrated favorable conditions with a uranium concentration of 105 g/L in an LEU solution volume of 155.5 L, containing 3.266 kg of uranium at 19.75% enrichment, and achieving a subcritical multiplication factor of 0.9681. Further analysis was performed by fixing the LEU solution volume at 155.5 L and varying the uranium mass (19.75% enrichment), comparing the distribution with daily 99Mo production and its impact on the subcritical multiplication factor. Based on this analysis, the most favorable condition for nuclear critical safety was a uranium concentration of 105 g/L yielding k near 0.97. Additional analyses included shielding and reflector material selection and design optimization. Calculations demonstrated that Be (10 cm thick) and W/B/PE (15 cm thick) were suitable for the reflector and shielding layers, respectively. The main optimized parameters are summarized as follows:

1. The optimal subcritical multiplication factor (k) for the designed SBS is 0.9685, with average neutron and gamma fluxes outside the shielding layer of 4.31$\times10^{4}$ n/cm$^2\cdot$s and 2.10$\times10^{7}$ γ/cm$^2\cdot$s, respectively. The neutron flux and nuclear heating distributions in the SBS are relatively uniform, as indicated by the K_H value of 1.12, which further enhances operational safety.
2. The SBS enables high ^{235}U utilization, producing 48 Ci of 99Mo per kilogram of ^{235}U.
3. A single SBS can produce 157 Ci of 99Mo per day. Since the GDT-FNS has an axisymmetric structure and the SBS solid angle is only $5\pi/18$, multiple SBS units can be simultaneously arranged in the high neutron flux region of the GDT-FNS. According to calculations, two and three such SBS units can meet Chinese market demand by 2025 and 2035, respectively.

The GDT-FNS-driven SBS 99Mo production system offers advantages of high production efficiency, low nuclear waste generation, and low cost. Our study indicates that this system could serve as a potential facility for 99Mo production. However, to enhance feasibility and practicality, further detailed design studies are necessary, including uranium burnup analysis and verification of 99Mo separation and purification techniques.

Author Contributions

The conception and design were originally proposed by Yun-Cheng Han, with contributions from all authors. Material preparation, data collection, and analysis were performed by Hou-Hua Xiong, Qiu-Sun Zeng, and Yun-Cheng Han.

The first draft was written by Hou-Hua Xiong. All authors commented on all versions of the manuscript and approved the final version.

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Note: Figure translations are in progress. See original paper for figures.