Production of 99Mo via photofission reaction in natural-uranium-bearing molten salt targets

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Abstract

This study proposes a method for 99Mo production via electron accelerator irradiation of a natural-uranium-bearing liquid molten salt target, with advantages including low nuclear proliferation risk, online extraction capability, and low construction costs. The approach primarily produces 99Mo through photofission of uranium (95%), specifically 238U(γ,f). Secondary neutrons, originating from photonuclear interactions or fission processes, contribute minimally (5%) to 99Mo production owing to their high energies and low fission cross-sections. Key parameter analyses revealed that fluoride salt systems exhibit higher 99Mo yield. Their performance stems from high bremsstrahlung energy loss rate and superior photon yield, making them optimal molten salt target materials. To maximize photofission and photoneutron cross-sections while minimizing high-energy gamma ray shielding requirements, an electron beam energy range of 40–80 MeV is recommended. To suppress local hot spots and prevent molten salt boiling, flow conditions were introduced to enhance convective heat transfer, effectively reducing the peak temperature. At a flow velocity of 0.5 m/s and under 80 MeV energy conditions, the maximum system temperature is only 808.9 K, which is significantly lower than the boiling point of 1773 K. Under optimized parameters, the maximum annual production capacity of 99Mo reaches 4486.49 Ci,sufficient for millions of diagnostic procedures and equivalent to 16.37% of China' s projected demand for 2030. This method provides a viable pathway for stable, large-scale 99Mo production.

Full Text

Preamble

This work proposes a method for producing 99Mo through electron accelerator irradiation of natural-uranium-bearing liquid molten salt targets, offering advantages that include low nuclear proliferation risk, online extraction capability, and reduced construction costs. The approach primarily generates 99Mo via uranium photofission (95%), specifically the 238U(γ,f) reaction. Secondary neutrons originating from photonuclear interactions or fission processes contribute minimally (5%) to 99Mo production due to their high energies and low fission cross-sections. Key parameter analyses reveal that fluoride salt systems exhibit higher 99Mo yields, stemming from their high bremsstrahlung energy loss rate and superior photon yield, making them optimal molten salt target materials. To maximize photofission and photoneutron cross-sections while minimizing high-energy gamma ray shielding requirements, an electron beam energy range of 40–80 MeV is recommended. To suppress local hot spots and prevent molten salt boiling, flow conditions were introduced to enhance convective heat transfer, effectively reducing peak temperature. At a flow velocity of 0.5 m/s under 80 MeV conditions, the maximum system temperature reaches only 808.9 K, significantly lower than the boiling point of 1773 K. Under optimized parameters, the maximum annual production capacity of 99Mo reaches 4486.49 Ci, sufficient for millions of diagnostic procedures and equivalent to 16.37% of China' s projected demand for 2030. This method provides a viable pathway for stable, large-scale 99Mo production.

Keywords: 99Mo, Electron accelerator, Molten salt, Natural uranium, Photofission

Introduction

As a pivotal medical radioisotope, 99mTc is extensively utilized in clinical diagnostics [1, 2], accounting for approximately 80% of global nuclear medicine radioisotope consumption [3]. Since 99mTc is primarily derived from the $\beta-$ decay of molybdenum-99 (99Mo, T1/2 65.92 h), the stability of 99Mo supply directly determines market availability of 99mTc. According to estimates by the Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA), global demand for 99Mo reached 5.5$×10^{5}$ Ci in 2020 and grows at over 10% annually, attaining 8.8$×10^{5}$ Ci by 2030 [4]. Furthermore, according to China' s "Medium-to-Long-Term Development Plan for Medical Isotopes (2021-2035)," annual demand for 99Mo has exceeded 1.6$×10^{4}$ Ci since 2019, with a steady 5% annual growth rate.

Currently, global supply of medical 99Mo is predominantly produced through research reactors in Europe, Canada, Australia, and South Africa [5, 6]. Due to significant nuclear proliferation risks associated with highly enriched uranium (HEU, ^{235}U enrichment >90%), its application is restricted [7, 8], prompting

most research reactors to transition to irradiating low enriched uranium (LEU, ^{235}U enrichment <20%) [9]. However, most research reactors have exceeded 40 years of operating time and will face shutdown and decommissioning over the next decade [2, 10], potentially triggering a severe "99Mo shortage crisis" that would critically impact 99Mo-importing nations. To sustain supply, the High Flux Reactor (HFR) in Petten, Netherlands—originally scheduled for shutdown in 2022—had its closure delayed [11], but in 2024, pipeline deformation above the reactor vessel prevented restart after routine maintenance, once again disrupting the global 99Mo supply chain [12]. Meanwhile, Belgium' s BR2 reactor and South Africa' s Safari-1 reactor are also extending operational cycles to address shortages [13, 14].

The international community is committed to developing new sustainable 99Mo production methods. In 1992, Chopela and Ball [15, 16] from Babcock & Wilcox (B&W) proposed the Medical Isotope Production Reactor (MIPR), which has attained commercial viability with small-scale industrial implementation [17-20]. However, MIPR faces significant power fluctuations during operation due to void, chemical, and temperature effects, risking control instabilities [21]. Molten salt reactors (MSRs) use molten salt mixtures as both fuel and primary coolant, offering significant advantages for isotope production such as high efficiency and online extraction capabilities [22, 23]. Consequently, global research increasingly focuses on MSR-based 99Mo production with online extraction systems. Studies have confirmed the technical feasibility and promise of MSRs for isotope generation [24, 25], revealing unique behaviors of fission products (e.g., Mo) that spontaneously migrate from molten salt phase into gas phase, with at least 50% of Mo fragments accumulating in the aerosol phase above the salt surface [26, 27]. This behavior forms the foundation for 99Mo production in MSRs, leading to further evaluations of online extraction feasibility [22, 28]. Nevertheless, MSR-based isotope production remains predominantly in theoretical validation and exploratory stages, with critical scientific and technical challenges requiring resolution.

In 2009, the OECD/NEA convened a meeting in Paris to address global 99Mo/99mTc supply security, establishing the High-level Group on Medical Isotopes to ensure sustainable global access [6]. Among various technological approaches, accelerator-based production routes—either generating 99Mo or directly yielding 99mTc—are recognized as viable medium-to-long-term alternatives due to regulatory flexibility, non-proliferation benefits, and low infrastructure/operating costs [29]. Accelerator-based production primarily employs two approaches: direct irradiation of production targets with primary particles (e.g., protons, electrons, deuterons, α-particles) or irradiation of converter targets to generate secondary particles bombarding production targets. Viable pathways include: ^{100}Mo(p,2n)99mTc, ^{100}Mo(p,pn)99Mo, ^{100}Mo(γ,n)99Mo, ^{96}Zr(α,n)99Mo, ^{100}Mo(n,2n)99Mo, ^{98}Mo(n,γ)99Mo, ^{235}U(n,f)99Mo, U/Th(p,f)99Mo and ^{238}U(γ,f)99Mo, among others [2, 9, 30-32]. Fig. 1 [Figure 1: see original paper] summarizes these accelerator-based production technologies [6].

Proton beam irradiation of natural uranium or thorium-232 (^{232}Th) can produce 99Mo via (p,f) reaction, but cross-sections are low [2]. Alternatively, proton irradiation of ^{100}Mo generates 99mTc directly through (p,2n) reaction or produces 99Mo via (p,pn) reaction, achieving high yields with lower proton energy requirements (E <30 MeV). However, the short half-life of 99mTc (T1/2 6.01 h) restricts long-distance transportation, limiting supply to surrounding areas. Notably, the competing reaction ^{100}Mo(p,2n)99gTc produces significant quantities of 99gTc, compromising radiopharmaceutical purity and imaging efficacy [33, 34]. Secondary particle methods utilize protons or electrons hitting converter targets to generate neutrons/photons that irradiate ^{100}Mo or ^{98}Mo to produce 99Mo. Nevertheless, molybdenum-based routes face three critical challenges: (a) low specific activity (0.35–15 Ci/g) due to molybdenum carrier component, requiring complex separation processes—significantly below clinical generator requirements (20–500 Ci/g) [35, 36]; (b) prohibitive target costs owing to low yields from natural molybdenum, requiring enriched targets (^{98}Mo or ^{100}Mo at 500–1500 USD/g) [29, 37]; (c) minuscule utilization efficiency (only 0.001% to 0.0001% of Mo atoms are transformed), mandating costly recovery systems and increasing operational expenditures [29, 35, 37, 38].

Building on the MIPR concept, the United States proposed an accelerator-driven subcritical solution reactor for isotope production [39, 40]. However, this method requires high-intensity, long-term stable accelerator neutron sources, posing significant challenges including considerable R&D complexity, prohibitive accelerator infrastructure costs, and high neutron production costs. In 2021, Han et al. [40, 41] proposed an accelerator-based deuterium-tritium (D-T) neutron source subcritical system for producing 99Mo, but this method requires tritium, which is costly and presents licensing challenges. Consequently, Han et al. [9] further introduced a novel design based on a LEU subcritical blanket system (SBS) driven by a Gas Dynamic Trap-based Fusion Neutron Source (GDT-FNS) utilizing deuterium-deuterium (D-D) fusion neutrons to induce ^{235}U fission. Alternatively, 99Mo can be produced by α-particle irradiation of ^{96}Zr via (α,n) reaction, though enriched ^{96}Zr targets increase production expenses [42].

Ruth [43] proposed photofission of ^{238}U targets using electron accelerators for 99Mo production, emphasizing that while ^{238}U(γ,f) yield is significantly lower than ^{235}U(n,f) production, this is outweighed by safer materials. Naik et al. [44] validated ^{238}U(γ,f) as a viable production route. Concurrently, Naik [45], Thierens [46], and Schmitt [47] measured cumulative 99Mo fission yields from ^{238}U at bremsstrahlung energies of 10 MeV, 25 MeV, and 48 MeV as 4.835%, 6.480%, and 6.600%, respectively. The cumulative yield increases with incident electron energy. In contrast, fast-neutron-induced fission of ^{238}U achieves a 99Mo cumulative yield of 6.168% [48], demonstrating comparable efficiency between photofission and fast-neutron-induced fission. Collectively, these studies establish the technical viability of 99Mo production via ^{238}U(γ,f) reaction. With its inherently high ^{238}U abundance (99.27%), natural uranium emerges as a strategically advantageous target material that eliminates en-

richment requirements, substantially reduces fabrication costs, and minimizes proliferation risks. Naik et al. [44] irradiated a 1 g natural uranium target for 24 hours with bremsstrahlung photons generated from a 10 MeV, 4 kW electron beam (10 Hz repetition rate) on tantalum foil, achieving 99Mo production of (0.309$±$0.050) mCi—confirming natural uranium' s practical utility.

Current accelerator-based 99Mo production predominantly employs solid targets, facing persistent challenges including complex fabrication, cutting, and dissolution processes; high costs; inability for online extraction; and inadequate heat dissipation. To overcome these limitations, this paper integrates the synergistic advantages of liquid molten salts and electron accelerators to propose 99Mo production via electron beam irradiation of natural-uranium-bearing molten salt targets. This approach utilizes the ^{238}U(γ,f) reaction, offering significant advantages: elimination of HEU to reduce proliferation risks; avoidance of solid-target processing through fluid targets; online extraction and simplified reprocessing for high specific activity; continuous irradiation to enhance uranium utilization; direct irradiation without converter targets, reducing costs and complexity; inherent flow-based cooling to enable higher beam intensities; and lower fabrication costs with economical molten salts.

A distinctive feature of molten salt targets compared to solid targets is their fluidity, which introduces numerous new challenges for target system design, including target size and structural design, molten salt circulation systems, thermal management and temperature control, and material corrosion compatibility. To evaluate feasibility, this paper employs Monte Carlo methods to systematically simulate and analyze critical operational parameters including molten salt composition, target geometry, incident electron beam energy, temperature distribution, and online extraction efficiency. Based on parameter analysis, the theoretical annual production capacity was quantitatively projected and comparatively assessed against China' s demand. These findings provide essential references for engineering implementation of this methodology.

Materials and Methods

A. Monte Carlo Model

FLUKA has been validated as a reliable tool for modeling radioisotope production [49–51]. Consequently, this study utilizes FLUKA to simulate and analyze 99Mo generation via electron irradiation of molten salts. Complementarily, coupled electron-photon transport and bremsstrahlung analysis was conducted using pyPENELOPE [52], while temperature distribution analysis was performed using ANSYS Fluent software [53].

The schematic of 99Mo production via electron beam irradiation of a molten salt target is shown in Fig. 2 [Figure 2: see original paper], comprising two core components: an electron beam and a molten salt target assembly. The electron beam features a radius of 1 cm, an energy range of 20–160 MeV, and an intensity of 1 mA, incident along the positive Z-axis. The molten salt target

assembly consists of two concentric layers: an inner molten salt region and outer cladding. The internal cavity is filled with molten salt (Fig. 2b [Figure 2: see original paper]), while the outermost cladding is a 0.5 cm thick nickel-based alloy. To conduct thermal analysis, the model further incorporates molten salt flow, with inlets and outlets configured accordingly (Fig. 2c [Figure 2: see original paper]). Along the Y-axis, molten salt enters from the bottom and exits from the top.

B. Reaction Models and Cross-Sections

Under electron beam irradiation, incident electrons initially generate bremsstrahlung photons through interactions with target atoms. These photons subsequently induce direct (γ,f) reactions with uranium to produce 99Mo, or alternatively undergo photonuclear reactions (γ,xn) to generate secondary neutrons that initiate (n,f) reactions on uranium atoms, yielding additional 99Mo. This reaction cascade is schematically illustrated in Fig. 3 [Figure 3: see original paper]. Consequently, 99Mo in molten salt originates primarily from fission of both ^{235}U and ^{238}U, encompassing direct fission products and decay of short-lived precursor nuclei. Key precursor nuclei include ^{99}Nb (T1/2=15.0 s) and ^{99}mNb (T1/2=2.5 min). Given their negligible half-lives compared to 99Mo, fission-generated precursors are assumed to fully decay to 99Mo during irradiation.

Fig. 4 [Figure 4: see original paper] displays the (γ,f) reaction cross-sections for ^{235}U and ^{238}U extracted from the ENDF/B-VIII.1 database [54]. Within the 10–20 MeV photon energy range, both isotopes exhibit significant (γ,f) cross-sections, with ^{235}U peaking at 0.33 b (Eγ=14 MeV) and ^{238}U peaking at 0.16 b (Eγ=14.5 MeV), indicating feasibility with relatively low electron/photon energies. Neutron-induced fission also contributes to 99Mo production, where secondary neutrons are predominantly generated via photonuclear reactions. Fig. 5 [Figure 5: see original paper] illustrates the (γ,xn) cross-sections for ^{235}U and ^{238}U, both peaking within 10–20 MeV (^{238}U: 0.73 b at 14 MeV; ^{235}U: 0.52 b at 14.5 MeV). The comparable photofission and photonuclear reaction cross-sections between ^{235}U and ^{238}U demonstrate low ^{235}U enrichment requirements, enabling use of natural uranium.

C. Computational Methods for Online Extraction

99Mo predominantly exists in molten salt as ionic species, noble metallic states, and fluoride compounds [22, 55]. Noble metallic 99Mo adheres to bubble liquid-gas interfaces, enabling online extraction via bubble methods. For instance, in molten salt reactors, helium is injected into the molten salt, circulates, and returns to the pump bowl. Helium exists as dissolved atoms and bubbles; 99Mo enters helium bubbles, adheres to liquid-gas interfaces, and flows to the pump bowl. High-velocity jet spraying within the pump bowl produces salt mist that migrates to the degassing module. The off-gas undergoes sequential physical-chemical processes (collection, cooling, separation, adsorption, filtration, purifi-

cation, and isotopic separation) to produce radiopharmaceutical-grade 99Mo [22].

The 99Mo production method proposed herein leverages liquid properties and bubble-based online extraction, simplifying processes and reducing costs. However, the physical behavior of 99Mo in molten salts—including ionic-to-metallic transition kinetics, governing constraints, and conversion ratios—remains incompletely understood [22]. Thus, calculations assume all 99Mo exists in noble metallic states. Online extraction is modeled using pseudo-decay kinetics with a pseudo-decay factor λi. For radionuclide i with inventory Ni and extraction efficiency P (s^{-1}), the differential extracted quantity dNi within infinitesimal time is expressed as: dNi = NiP dt. Therefore, the pseudo-decay factor is: λi = P.

Throughout the irradiation period, the 99Mo production rate is assumed constant at Y (Bq/s). Consequently, temporal evolution of 99Mo activity without online extraction is described by:

$dA(t)/dt = Y - \lambda A(t),\ A(0) = 0$

where λ denotes the decay constant of 99Mo (s^{-1}) and A(t) represents activity (Bq) at time t. Integration gives:

$A(t) = (Y/\lambda)(1 - e^{(-\lambda t)})$

From this equation, as t approaches infinity:

$Amax = Y/\lambda$

Amax denotes saturation activity where production and decay reach equilibrium. Thus, $Y = \lambda Amax$. When extraction efficiency P is introduced, the rate of change of activity A(t) is governed by three components: irradiation production rate Y, radioactive decay rate $\lambda A(t)$, and pseudo-decay rate (extraction efficiency) $\lambda iA(t)$:

$dA(t)/dt = Y - (\lambda + \lambda i)A(t) = Y - (\lambda + P)A(t),\ A(0) = 0$

Integration yields the activity function:

$A(t) = Y/(\lambda + P)\ (1 - e^{(-(\lambda+P)t)})$

The cumulative extracted activity is:

$Q(t) = {}_0\ P\ A(t\ )dt\ = (PY/(\lambda + P)^2)[(\lambda + P)t - 1 + e^{(-(\lambda+P)t)}]$

Irradiation Parameters Analysis

The 99Mo yield is primarily determined by electron energy, beam intensity, irradiation time, molten salt composition, and dimensions. Since per-particle yield remains constant, total yield increases proportionally with beam intensity. Consequently, subsequent simulations fix beam intensity at 1 mA to focus on other parameters. Composition analysis reveals that LiF-UF4 salt achieves optimal

99Mo yield (5.33$×10^{9}$ Bq) under 40 MeV electron irradiation for 1 hour in a 5 cm × 6 cm × 10 cm volume. For LiF-UF4 systems, electron energies of 40–80 MeV are most efficient. Evaluations of 99Mo spatial distributions under 40 MeV electrons and LiF-UF4 salt show that optimized geometry (74 cm × 74 cm × 46 cm) yields 8.78$×10^{9}$ Bq after 1 hour of irradiation. Finally, to ensure localized boiling does not occur, maximum molten salt temperature was evaluated, showing that increasing flow velocity to 5 m/s under 80 MeV conditions results in a maximum temperature of only 808.9 K, effectively avoiding boiling risk and ensuring safe, stable operation.

A. Molten Salt Composition and Electron Energy Analysis

Molten salt composition critically influences 99Mo yield. Compared to enriched molybdenum targets, molten salts offer significantly lower costs—base salts (e.g., LiF-BeF2) cost 71.23 USD/kg [56], and natural uranium is approximately 200 USD/kg [57]. This cost advantage enables substantial reductions in target fabrication expenses. This section investigates 99Mo yields across various molten salt compositions, with parameters detailed in Table 1 . The system employs 99.95% ^{7}Li enrichment, 97% ^{37}Cl enrichment, natural uranium, and 923 K operating temperature.

A 40 MeV electron beam irradiates the molten salt target assembly with dimensions of 5 cm × 6 cm × 10 cm. After 1 hour of irradiation, 99Mo yields for various salts are presented in Table 2 . LiF-UF4 salt achieves the highest yield at 5.33$×10^{9}$ Bq, followed by NaF-UF4 at 4.55$×10^{9}$ Bq. NaCl-MgCl2-UCl3 demonstrates the lowest yield (1.89$×10^{9}$ Bq). Notably, while NaCl-UCl3 salt contains the highest U-content, its 99Mo yield remains comparatively low. Conversely, LiF-BeF2-UF4 salt—despite having the lowest U-content —achieves higher yields than both NaCl-KCl-UCl3 and NaCl-MgCl2-UCl3 systems. Crucially, fluoride salts universally exhibit higher 99Mo yields than chloride salts.

As shown in Table 2 , 95% of 99Mo originates directly from photofission (encompassing both direct fission and decay of fission products), indicating that bremsstrahlung photon yield governs 99Mo production. For electrons, the bremsstrahlung energy loss rate satisfies:

$$S = (4\alpha N_A)/(A X_0) \ln(183/Z^{1/3})$$

where E denotes electron energy, represents material density, and X_0 is the radiation length:

$$X_0 = (716.4 \cdot A)/(Z(Z+1)\ln(287/\sqrt{Z})) \ g/cm^2$$

For compounds or mixtures, the radiation length X_0 is calculated as:

$$1/X_0 = \Sigma(w_i/X_{0i})$$

At 40 MeV electron energy, radiation energy loss rates in different molten salts are quantified in Table 2 . Concurrently, bremsstrahlung photon spectra gener-

ated by 40 MeV electrons in various salts were simulated using pyPENELOPE (Fig. 6 [Figure 6: see original paper]). Results reveal that LiF-UF4 salt exhibits the highest bremsstrahlung photon yield, whereas NaCl-MgCl2-UCl3 gives the lowest, directly correlating with radiation energy loss rates: LiF-UF4 displays the maximum rate, followed by NaF-UF4, while NaCl-MgCl2-UCl3 shows the minimum, accounting for superior 99Mo production in LiF-UF4 salt.

Bremsstrahlung photons not only induce photofission but also generate secondary neutrons, while uranium fission produces additional neutrons. Spatial distributions of secondary neutrons from 40 MeV electrons in various molten salts are illustrated in Fig. 7 [Figure 7: see original paper]. As penetration depth Z increases, electron energy gradually diminishes, causing cumulative growth in bremsstrahlung photon yield and flux that elevates secondary neutron production. Beyond a critical depth, complete electron energy depletion coupled with photon attenuation diminishes neutron yield. Consequently, LiF-UF4 exhibits the highest secondary neutron yield due to maximal photon production. Fig. 8 [Figure 8: see original paper] displays the normalized energy spectrum of secondary neutrons in LiF-UF4, wherein fast neutrons dominate with a mean energy of 1.23 MeV. At this energy, neutron fission cross-sections for ^{235}U and ^{238}U are low (1.21 b and 0.04 b, respectively), resulting in poor neutron utilization efficiency as most neutrons leak from the target, contributing only 5% to total 99Mo production.

The effects of electron energy on 99Mo yield in LiF-UF4 are further investigated (Fig. 9 [Figure 9: see original paper]). Photofission cross-sections of uranium peak within the 5–20 MeV photon energy range. Bremsstrahlung photons exhibit a continuous spectrum where maximum photon energy approaches incident electron energy with low intensity, while peak intensity occurs near half the electron energy [59, 60]. To optimize photofission and photonuclear cross-sections, electron energies must exceed 40 MeV. While 99Mo yield increases with electron energy, growth rates progressively diminish. However, excessively high energy generates energetic γ-rays with reduced reaction cross-sections, increased shielding complexity, and undesirable side-reactions producing radiochemical impurities. Consequently, the 40–80 MeV range balances yield efficiency and engineering feasibility.

B. Molten Salt Dimensions and Temperature Analysis

With a fixed electron beam spot size (e.g., 1 cm), molten salt target dimensions critically impact 99Mo yield. Undersized targets underutilize beam flux, whereas oversized targets increase salt consumption and costs. FLUKA simulations reveal 99Mo spatial distributions via residual nuclei calculations under 40 MeV, 1 mA electron irradiation for 1 hour (Fig. 10 [Figure 10: see original paper]). The electron beam propagates along the positive Z-direction, perpendicular to the XY-plane. In the XY-plane, residual nuclei exhibit a concentric distribution from the beam point, with density decreasing radially. Along the XZ-plane, residual nuclei diminish exponentially along Z, becoming negligible

beyond 90 cm. Statistical analysis confirms that 98% of residual nuclei concentrate within $±$37 cm (X/Y) and 46 cm (Z). Thus, optimal dimensions of 74 cm × 74 cm × 46 cm achieve a peak yield of 8.78$×10^{9}$ Bq.

For accelerator targets, a core challenge is effectively removing substantial heat generated by beam energy deposition to prevent local overheating [61, 62]. To evaluate temperature distribution in a molten salt target (LiF-UF4, 74 cm × 74 cm × 46 cm), the three-dimensional energy deposition distribution from an electron beam (40/80 MeV, 1 mA) was calculated using FLUKA. The energy deposition data were imported into ANSYS Fluent for thermal-hydraulic calculations to obtain the steady-state temperature field. During operation, beam energy deposition can form hot spots, posing boiling risks, thus requiring efficient heat removal. However, due to the large target volume, beam-deposited heat is insufficient to maintain the entire region in a molten state, necessitating additional heating devices to prevent solidification. To simplify the computational model, a constant temperature boundary condition of 773 K was applied to the molten salt region in Fluent.

Fig. 11 [Figure 11: see original paper] shows the steady-state temperature distribution at Z=0.2 cm under 40 MeV and 80 MeV electron beam irradiation. The electron beam is incident vertically with a spot radius of only 1 cm, and due to the short electron range, energy is primarily deposited near the injection point, forming a local hot spot. The maximum temperature point is located at approximately (X=0 cm, Y=0 cm, Z=0.2 cm). Molten salt flows upward (inlet at bottom, outlet at top) and continuously removes heat from the hot spot vicinity, creating a temperature gradient band along the flow direction. Variation of maximum temperature with inlet flow velocity is shown in Fig. 12 [Figure 12: see original paper]. As flow velocity increases from 0.1 m/s to 0.5 m/s, maximum temperature decreases from 977.3 K to 805.0 K (40 MeV) and from 1011.0 K to 808.9 K (80 MeV). At the same flow velocity, the 80 MeV beam induces higher maximum temperature than the 40 MeV beam, but this difference gradually diminishes as flow velocity increases. Importantly, all maximum temperatures remain well below the boiling point (above 1773 K [63]), indicating safe operation. The heat dissipation performance of flowing molten salt targets is significantly superior to solid targets, attributed to distinct thermal-hydraulic mechanisms. Solid targets rely mainly on molecular thermal vibration for heat conduction—a single, less efficient path. In contrast, flowing molten salt targets incorporate additional forced convection, significantly enhancing heat transfer through convective exchange. Continuous flow efficiently carries away heat, preventing local overheating and ensuring stable, efficient heat transport [64, 65]. This allows molten salt targets to meet dissipation requirements through their own flow, eliminating need for additional cooling devices, simplifying system structure, and reducing costs.

Online Extraction Computational Analysis

Current accelerator-based 99Mo production predominantly employs solid targets (e.g., metallic molybdenum, molybdenum oxide, uranium oxide) requiring complex fabrication: homogenizing powders via dissolution-oxidation processes with a carrier matrix, followed by mechanical compaction. Isotope extraction requires multiple steps—target disassembly, sectioning, dissolution, wet or dry separation and purification—yielding a complex workflow [5]. Therefore, leveraging 99Mo's existence as insoluble particles in molten salt and the fluid properties of liquid molten salts, this paper adopts a bubbling method for online extraction analysis.

Based on optimization results from Section III (40 MeV electron energy, LiF-UF4 molten salt, 74 cm × 74 cm × 46 cm), Fig. 13 [Figure 13: see original paper] illustrates 99Mo yield variation with irradiation time. The yield increases progressively with irradiation time, while decay also increases. Consequently, production and decay eventually reach equilibrium, saturating yield at 8.53$×10^{11}$ Bq. Utilizing this saturated yield and Eq. (6), the 99Mo production rate Y under these conditions is calculated as 2.49$×10^{6}$ Bq/s.

Fig. 14 [Figure 14: see original paper] and Fig. 15 [Figure 15: see original paper] display residual 99Mo activity in molten salts, cumulative extraction quantity, equilibrium time, and saturated extraction yields at different extraction efficiencies. Higher extraction efficiencies reduce residual 99Mo activity, accelerate equilibrium attainment, and lower equilibrium activity levels. At an extraction efficiency of 10^{-3} s^{-1}, equilibrium is achieved in 65 minutes. Once 99Mo activity in molten salt saturates, extraction quantity per unit time stabilizes, resulting in linear cumulative yield increase. Saturated extraction yield initially increases with extraction efficiency then stabilizes: at 10^{-4} s^{-1}, it stabilizes at 2.09$×10^{11}$ Bq/day; at 10^{-3} s^{-1}, it increases to 2.15$×10^{11}$ Bq/day. Balancing equilibrium time against saturated yield, the optimal extraction efficiency is determined to be 10^{-4} s^{-1} with equilibrium time at 10.57 hours and saturated yield of 2.09$×10^{11}$ Bq/day.

99Mo Production Estimation and Comparison with China' s Demand

Stable daily production via electron accelerator irradiation of molten salts could support domestic demand. The proposed technology enables continuous operation through accelerator flexibility and molten salt online extraction capabilities. Through systematic optimization of electron energy, target parameters, and temperature control, efficient supply-demand matching and autonomous security for China' s 99Mo supply chain can be achieved.

Using optimized parameters (40/80 MeV electrons, 1 mA beam intensity, LiF-UF4 molten salt 74 cm × 74 cm × 46 cm, 923 K salt temperature, 10^{-4} s^{-1} extraction efficiency), annual 99Mo extraction yields were calculated. Under

these conditions, production rates reached 2.49$×10^{6}$ Bq/s (40 MeV) and 6.35$×10^{6}$ Bq/s (80 MeV). Accounting for cyclic accelerator maintenance (3 days irradiation + 0.5 days maintenance cycles), Fig. 16 [Figure 16: see original paper] illustrates annual 99Mo activity and cumulative extraction dynamics. Each cycle features rising activity during irradiation and rapid decay during maintenance, with cumulative extraction increasing quasi-linearly. Annual yields differ significantly: 6.53$×10^{13}$ Bq (1764.86 Ci) at 40 MeV versus 1.66$×10^{14}$ Bq (4486.49 Ci) at 80 MeV.

As shown in Table 3 , compared with other production methods, this study shows no significant yield advantage—only exceeding the result reported by Villa et al. [66]. However, this study employs natural uranium as raw material, offering higher safety and easier availability compared to schemes requiring LEU. Furthermore, studies by Minato et al. [67], Gao et al. [32], and Bondar et al. [68] all utilize highly enriched molybdenum or zirconium targets, entailing high costs and complex production processes. Therefore, the notable advantages in safety, flexibility, and online production capability sufficiently compensate for lower yield.

In clinical practice, administered 99mTc activity varies with radiopharmaceuticals, disease types, and patient conditions. For instance, 99mTc-MDP (Technetium-99m methylene diphosphonate) employed in bone scintigraphy requires 740–1110 MBq (20–30 mCi) per dose, necessitating 34–51 mCi of 99Mo [69–71]. In contrast, 99mTc-DMSA (Technetium-99m dimercaptosuccinic acid) for renal imaging requires 11.1–111 MBq (0.3–3 mCi) of 99mTc, corresponding to 0.5–5 mCi of 99Mo [72]. Excluding other losses, based on maximum (51 mCi) and minimum (0.5 mCi) 99Mo requirements per dose, a 40 MeV facility can supply approximately 34,605 (high-dose scenario) to 3,529,720 (low-dose scenario) patient doses annually, while an 80 MeV facility can provide roughly 87,970 (high-dose scenario) to 8,972,980 (low-dose scenario) patient doses per year. Furthermore, China' s "Medium-to-Long-Term Development Plan for Medical Isotopes (2021–2035)" projects national 99Mo demand in 2030 will reach 2.74$×10^{4}$ Ci. A single 40 MeV accelerator and an 80 MeV unit could satisfy 6.44% and 16.37% of national demand, respectively. Increasing beam intensity could further reduce required facilities.

To minimize decay losses during transportation and leverage deployment flexibility and safety advantages, deploying 99Mo production facilities in major cities (e.g., Beijing, Shanghai, Shenzhen, Chengdu, Wuhan) is recommended. Integrated with existing ^{18}F distribution networks, this strategy would achieve rapid nationwide supply delivery, covering 95% of the population within the effective distribution radius [73].

Conclusions

This study proposes 99Mo production via electron accelerator irradiation of natural-uranium-bearing liquid molten salts, primarily exploiting the ^{238}U(γ,f)

reaction pathway. Key advantages include low nuclear proliferation risk, online extraction capability, and reduced target fabrication costs. Through systematic feasibility analysis and parameter optimization, the following principal conclusions are drawn:

1. Fluoride-based systems exhibit higher 99Mo yields than chloride-based systems due to higher bremsstrahlung energy deposition rates and photon production.
2. Following comprehensive assessment of yield efficiency, shielding costs, and technical feasibility, an electron energy range of 40–80 MeV is identified as optimal.
3. As inlet molten salt flow velocity increases, maximum temperature inside the target gradually decreases. At 0.5 m/s flow velocity under 80 MeV conditions, maximum system temperature is only 808.9 K, significantly lower than the 1773 K boiling point, ensuring safe operation.
4. Online extraction capability of liquid molten salt targets significantly improves production flexibility. By balancing equilibrium time and saturated yield, the optimal extraction efficiency is 10^{-4} s^{-1}.
5. Maximum annual production capacity reaches 4486.49 Ci at 80 MeV, sufficient for millions of diagnostic procedures and accounting for approximately 16.37% of China' s projected 2030 demand.

This study establishes an innovative technical pathway for 99Mo production, though it primarily focuses on feasibility analysis and critical parameters. Several limitations require further refinement, such as cladding alloy thickness and target configuration. The 0.5 cm thick nickel-based alloy cladding necessitates rigorous evaluation of electron beam attenuation impacts, while the proposed cuboid target geometry warrants comparative assessment against alternatives (e.g., spherical, cylindrical). Future work must integrate shielding design, thermal-hydraulic analysis, and insulation system optimization for systematic structural refinement. Additionally, the bubbling online extraction system demands in-depth investigation aligned with 99Mo physicochemical properties in molten salts. These advancements will establish a more robust foundation for industrial-scale application.

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