Nuclear Safety Analysis of Molten Salt Reactor Criticality and 7Li Abundance Variation

Authors: Yang Zhen, Dai Zhimin, Zou Yang, Yao Yongkui, Zhimin Dai

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Abstract

Molten salt reactors represent one of the six Generation IV reactor types, utilizing liquid nuclear fuel in contrast to conventional solid-fuel reactor designs. To ensure effective core design management and nuclear safety regulation, it is necessary to analyze the relationship between 7Li enrichment and nuclear criticality parameters. This study establishes a model based on molten salt reactors with engineering experience, employing software simulation calculations to analyze the influence of fuel salts with varying 7Li enrichments on molten salt reactor reactivity and to examine the patterns of nuclear criticality parameter variations. Through iterative calculations, the 7Li enrichment value at molten salt reactor criticality is rapidly and accurately determined. The final conclusion indicates that molten salt reactor reactivity increases with increasing 7Li enrichment in the fuel salt, and that the rate of reactivity change in molten salt reactors is also related to 7Li enrichment. Based on the analytical results of this study, in-depth discussions are conducted, summarizing relevant management requirements from legal and regulatory perspectives and proposing key points for attention from a regulatory review standpoint.

Full Text

Nuclear Safety Analysis of Molten Salt Reactor Criticality with Changes in 7Li Abundance

YANG Zhen[1,2,3], DAI Zhimin[1], ZOU Yang[1], YAO Yongkui[3]

[1]Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China
[2]University of Chinese Academy of Sciences, Beijing 100049, China
[3]North-Western China Regional Office of Nuclear and Radiation Safety Inspection, National Nuclear Safety Administration, Lanzhou 730020, China

Abstract

Molten salt reactors represent one of the six internationally recognized Generation IV reactor designs, distinguished by their use of liquid nuclear fuel rather than conventional solid fuel. To support effective core design management and nuclear safety regulation, it is essential to analyze the relationship between 7Li abundance and nuclear criticality parameters. This study models a molten salt reactor based on engineering practice and employs software simulation to analyze how fuel salts with different 7Li abundances affect reactor reactivity and to identify patterns in nuclear criticality parameter variations. Through iterative calculations, the study rapidly and accurately determines the 7Li abundance value at reactor criticality. The results demonstrate that molten salt reactor reactivity increases with increasing fuel salt 7Li abundance, and that the rate of reactivity change is also related to 7Li abundance. Building on these findings, the paper discusses relevant management requirements from a legal and regulatory perspective and proposes key points for regulatory attention.

Keywords: Molten salt reactor; Fuel salt; 7Li abundance; Nuclear criticality; Nuclear safety

Introduction

Molten salt reactors differ fundamentally from existing light water reactors, heavy water reactors, and high-temperature gas-cooled reactors in that the core fuel consists of high-temperature liquid molten salt that simultaneously serves as the primary coolant, enabling operation at atmospheric pressure with graphite as the moderator. To date, the only molten salt reactor with actual power operation experience is the Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) in the United States. The MSRE was an 8 MW(thermal) reactor in which molten fluoride salts circulated through graphite channels at 1200°F (648.89°C) to demonstrate key characteristics of molten salt reactor technology. Consequently, this study uses the MSRE as a reference, with initial critical fuel salt composition of LiF-BeF2-ZrF4-UF4 (molar ratio 65%-29.1%-5%-0.9%).

According to China's Regulations on the Control of Nuclear Materials, the maximum designed mass enrichment of 235U for civilian nuclear materials is 20%. Therefore, the model in this study adopts a 235U mass enrichment of 20% (enrichment level of 20.2%) rather than the 33% enrichment used in the original MSRE design. Analysis of neutron microscopic absorption cross-sections (in barns, where 1 b = 10^{-28} m^2) reveals that at room temperature (20°C or 293.15 K) with neutron energy of 0.0253 eV, the thermal neutron cross-sections are: 19F at 0.0095 b, 6Li at 938.041 b, 7Li at 0.0454 b, 9Be at 0.0092 b, natural Zr at 0.158 b, 235U at 680.9 b, and 238U at 2.70 b. At MSRE operating temperature of 1200°F (922 K), corresponding to neutron energy of 0.0795 eV, linear interpolation of National Nuclear Data Center database yields: 19F at 0.0054 b, 6Li at 529.822 b, 7Li at 0.0256 b, 9Be at 0.0057 b, natural Zr at 0.104 b, 235U at 338.2 b, and 238U at 1.53 b.

As a graphite-moderated thermal neutron reactor, the molten salt reactor's neutron spectrum approximates a fission neutron spectrum in the high-energy region, follows a 1/E relationship in the moderation region, and approximates a Maxwellian distribution in the thermal region. Consequently, besides 238U resonance absorption and its Doppler effect in the moderation region, 6Li competes with 235U for neutron absorption in the thermal region. The ratio of neutron absorption capability between 6Li and 235U in the thermal region varies little with temperature, with absorption cross-section ratios ranging from approximately 1.38 to 1.57 in the energy range below Ec2.

The large neutron absorption cross-section of 6Li leads to increased tritium production upon neutron capture, affecting critical fuel loading and increasing effluent emissions. The tritium production depends on neutron flux, 6Li density in the core, and operating time. The reaction is: 6Li + n → T + 4He. While using 100% pure 7Li would be ideal for safety and environmental protection, natural lithium contains only 92.41% 7Li and 7.59% 6Li. Isotopic separation to purify 7Li is costly, with global annual production capacity limited to ton-scale quantities, making 100% 7Li purity unrealistic with current industrial capabilities. Therefore, the impact of 6Li on reactivity must be considered to understand safety characteristics and patterns, which benefits regulatory review, licensing, and inspection processes.

The fuel salt temperature in the core is set at 648.9°C, with the core approximated as a cylindrical vessel containing graphite at a certain packing fraction. For liquid-fuel molten salt reactors, neutron moderation and nuclear fission reactions occur primarily in the graphite channel region, and core filling can reduce the influence of fuel salt outside the graphite structure. While fuel salt circulation removes some delayed neutron precursors, reducing critical reactivity depending on flow velocity and external piping length, neglecting this effect in keff calculations is conservatively safe. Due to strong negative temperature feedback, operation exhibits inherent self-regulating safety characteristics, making temperature fluctuations less concerning. This study focuses on initial fuel loading and criticality conditions to prevent instantaneous introduction of excessive positive reactivity, with other operating conditions analyzed qualitatively. Therefore, this research does not consider reactivity from fuel salt outside the graphite structure, effects of fuel salt flow on delayed neutrons, temperature feedback effects, or external graphite configuration, as these are not central to the research focus, though they must be addressed in detailed engineering design and operation.

1.1 Fuel Salt Parameters

Based on MSRE data, the density of LiF-BeF2-ZrF4-UF4 fuel salt is approximately $2.26 \times 10^3\ \mathrm{kg \cdot m^{-3}}$. The relationship is given by:

$$\frac{0.65 \cdot m_{\mathrm{LiF}} + 0.291 \cdot m_{\mathrm{BeF2}} + 0.05 \cdot m_{\mathrm{ZrF4}} + 0.009 \cdot m_{\mathrm{UF4}}}{N_A} \cdot \rho = 2.26 \times 10^3$$

where N_A is Avogadro's constant (6.022×10^{23} mol^{-1}), and m_{LiF}, m_{BeF2}, m_{ZrF4}, m_{UF4} are the molecular weights of the respective compounds. Calculation yields a total cation density N_0 of approximately 3.25958×10^{28} m^{-3}. The total density of all anions and cations in the fuel salt is:

$$N_{\mathrm{total}} = 8.04464 \times 10^{28}\ \mathrm{m}^{-3}$$

1.2 Core Geometry Parameters

The MSRE core had a diameter of 54 inches (1.372 m) and a moderator height of 64 inches (1.626 m), constructed from graphite rods with 2-inch (0.051 m) square cross-sections. This design could accommodate 260 graphite rods. The core fuel salt mass is calculated as:

$$V_{\mathrm{total}} = \frac{\pi D^2 H}{4} = 3.14 \times \frac{1.626 \times D^2}{4}$$

$$V_{\mathrm{fuel}} = H \times \left(3.14 \times \frac{D^2}{4} - 260 \times 0.051^2\right) = 1.30\ \mathrm{m}^3$$

$$m_{\mathrm{fuel}} = 1.3 \times 2.26 \times 10^3 = 2945\ \mathrm{kg}$$

where V_{total} is total core volume, D is core diameter, H is core height, a is fuel salt channel diameter or side length, V_{fuel} is core fuel salt volume, m_{fuel} is total fuel salt mass, and ρ is fuel salt density. Based on neutron moderation/reflection characteristics and conservative safety assumptions, graphite is assumed to fit seamlessly against the reactor vessel inner wall, with liquid fuel salt flowing only through internal channels surrounded by graphite.

Initially referencing the MSRE moderator region fuel salt mass, the analysis examines reactivity variation for 225 channels (channel radius 3.4 cm). Unlike water reactors, molten salt reactors use graphite as both core structure and fuel salt channels, with graphite structures immersed in fuel salt, lacking a reflector layer like water reactors and experiencing higher neutron leakage, though benefiting as heterogeneous reactors in resonance escape probability. Therefore, key design parameters are graphite channel diameter and spacing, followed by appropriate core height-to-diameter ratio. To ensure criticality with 20% enriched 235U fuel salt, this study designs a core with 2 m diameter (compared to 1.372 m for original MSRE), fuel salt channel cross-section area of 0.80 m^2, and fuel salt loading volume of 1.3 m^3 (2945 kg), with other parameters unchanged.

Thus, a graphite core is designed with a honeycomb structure: diameter $D = 2$ m, height $H = 1.626$ m, fuel salt flow channel radius of 0.0337 m, and 225 such channels. Each channel and surrounding graphite structure can be divided into a lattice with 0.0934 m side length, as shown in Figure 2 [Figure 2: see original paper].

By setting 7Li abundance, the corresponding k_{eff} can be calculated using SCALE6.1 (Standardized Computer Analyses for Licensing Evaluation). Developed and maintained by ORNL, SCALE6.1 is a modular code package for reactor physics analysis, criticality safety, and radiation shielding calculations, based on multigroup cross-section three-dimensional Monte Carlo neutron transport theory. The system includes databases, cross-section processing programs, radiation transport codes, and fuel burnup/activation analysis capabilities. The generic neutron reaction cross-section library includes 238-ENDF/B-V and 44-ENDF/B-V neutron libraries; this calculation employs the 238-ENDF/B-V library.

To find the 7Li abundance at $k_{\text{eff}} = 1$, a point-by-point evaluation from 0 to 1 would be computationally prohibitive. Therefore, this study innovatively applies an iterative segmentation method to progressively narrow the k_{eff} range until reaching the desired error tolerance. Starting with initial 7Li abundance of 100% and maintaining constant core loading and geometry, the "golden section" method iteratively approximates the 7Li abundance at criticality using SCALE6.1 calculations.

Based on SCALE6.1 criticality analysis results for this model, the maximum k_{eff} standard deviation is 0.00104, making $|k_{\text{eff}} - 1| < 0.001$ an appropriate convergence criterion. A maximum of 15 iterations prevents infinite loops from calculation errors, achieving 7Li abundance precision at the 10^{-6} level, sufficient for this analysis. The calculation process is shown in Figure 3 [Figure 3: see original paper].

2 Impact of 7Li Abundance on Fuel Loading Uranium Concentration

Natural lithium contains 92.41% 7Li and 7.59% 6Li. This study sets the upper 7Li abundance limit at 100% and lower limit at 90% for iterative calculations using SCALE6.1 to determine the effective multiplication factor k_{eff}. Basic parameters are first calculated as shown in Table 2 .

2.1 Upper Limit Initial 7Li Abundance

100% 7Li abundance serves as the upper calculation limit. For a core with 2 m diameter, 1.626 m height, and channel radius of 0.034 m, k_{eff} is 1.075, sufficient for criticality. At this condition, total fuel salt volume is 1.30 m^3, total mass is 2938 kg, total uranium mass is 150 kg, and total 235U mass is 30 kg, as calculated in Table 3 .

2.2 Lower Limit Initial 7Li Abundance

90% 7Li abundance serves as the lower calculation limit, yielding $k_{\text{eff}} = 0.074$, as shown in Table 4 .

2.3 Iterative 7Li Abundance Calculations

Following the method in Figure 3 [Figure 3: see original paper], the first iteration shows that at 7Li abundance P_1, $k_{\text{eff}} = 1.075 > 1$, while at P_2, $k_{\text{eff}} = 0.074 < 1$. The iterative parameter (abundance) is set to 0.120.

The second iteration proceeds similarly: at 7Li abundance P_1, $k_{\text{eff}} = 1.075 > 1$; at $P_3 = 96.2\%$, $k_{\text{eff}} = 0.120 < 1$, yielding an iterative parameter of 0.203.

Subsequent iterations continue (for comparison, values are displayed with appropriate significant figures):

$$P_4 = P_3 + 0.618 \times (P_1 - P_3) = 0.98541$$

Results for various 7Li abundances are summarized in Table 5 .

3 Data Fitting and Application Discussion

Using the MSRE design as reference, this study determines the 7Li abundance required for criticality in a 225-channel graphite core with 20% 235U enrichment. The resulting reactivity variation patterns and safety parameters provide valuable reference for reactor nuclear design and regulatory oversight.

The relationship between k_{eff} and 7Li abundance is shown in Figure 4 [Figure 4: see original paper]. The trend indicates that k_{eff} changes more rapidly as 7Li abundance increases. For the designed core, 7Li abundance near 99.97% is reasonable, while abundance above 99.98% ensures criticality. At 100% 7Li abundance, $k_{\text{eff}} = 1.075$, with supercritical margin exceeding the delayed neutron fraction, potentially leading to prompt criticality or reactor period protection issues. Conversely, using natural or below 99% 7Li abundance may prevent criticality due to reduced neutron utilization of 235U.

The relationship between 7Li abundance variation and reactivity coefficient is shown in Table 6 . Near the critical point (99.98% 7Li abundance), the reactivity change rate is maximum (steepest curve slope), meaning small changes in 7Li abundance significantly affect criticality. This region should inform reactor design 7Li abundance selection. Considering $k_{\text{eff}} > 0.9$ (7Li abundance range 99.92% to 100%), interpolation yields a recommended 7Li abundance range of 99.95% to 99.99%.

For 235U thermal fission, the delayed neutron reactivity effect at criticality is 650 pcm. For MSRE with 235U fuel, the measured total worth of three control rods inserted simultaneously was approximately 5590 pcm (theoretical value

~5460 pcm), with a single control rod worth about 2260 pcm (theoretical ~2110 pcm).

1. If 7Li abundance varies between 99.97% and 99.99%, the corresponding reactivity change is 759 pcm, exceeding the delayed neutron fraction. If a modular molten salt reactor is loaded with 99.99% 7Li but designed for 99.97%, with control rod positions adjusted accordingly, prompt criticality could occur with a very small reactor period, creating an uncontrollable and extremely dangerous condition.

2. If 7Li abundance varies between 99.92% and 100%, the reactivity change is 3201 pcm, exceeding the worth of a single MSRE control rod. Loading 99.92% 7Li when designed for near 100% could result in insufficient shutdown margin or inability to reach criticality; conversely, loading near 100% when designed for 99.92% could leave insufficient safety rod shutdown margin. Therefore, for effective reactivity control, molten salt reactor fuel should use 7Li abundance greater than 99.92%.

3. If 7Li abundance varies between 99.78% and 99.97%, the reactivity change is 6976 pcm, exceeding the total worth of all MSRE control rods. Deviation between actual and design 7Li abundance in this range would make the reactor uncontrollable or unable to achieve criticality.

According to the Research Reactor Operating Unit Reporting System, during molten salt reactor operation and experiments from initial fuel loading through decommissioning, if 7Li abundance exceeds design values during initial approach to criticality, causing reactivity insertion rate to exceed limits, introducing reactivity beyond prescribed limits, or resulting in power doubling period shorter than specified; or if 6Li depletion during operation increases 7Li abundance, causing shutdown margin to deviate from limits—operating units must report such events to the National Nuclear Safety Administration and regional supervision stations.

This study examines the relationship between k_{eff} and 7Li abundance for a specific core, developing a golden-section iterative method using SCALE6.1 to determine critical 7Li abundance. From criticality characteristic curve analysis, it proposes regulatory review points from a nuclear safety supervision perspective. For this core, critical safety characteristics are:

1. 6Li exhibits significant neutron absorption effects, making 7Li abundance a critical parameter affecting k_{eff}. Appropriate 7Li abundance selection is essential. Considering only reactor physics criticality characteristics, values above 99.92% are generally appropriate. For similar designs, 7Li abundance of $99.97 \pm 0.02\%$ is recommended.

2. Higher 7Li abundance yields greater k_{eff} after loading. Deviations of actual 7Li abundance from design values—whether high or low—are concerning. Higher abundance may increase reactivity, causing accidental criticality or supercriticality; lower abundance may prevent criticality even when other

loading parameters meet requirements.

3. Near 100% 7Li abundance, the contribution to k_{eff} becomes more pronounced, with the 7Li abundance-k_{eff} relationship curve changing more rapidly. Particularly near critical 7Li abundance (~99.98%), each 0.001% change in 7Li abundance alters reactivity by more than 50 pcm. Since practical applications use 7Li abundance near 100%, preventing steep-edge effects at criticality is necessary.

Regulatory attention should focus on liquid-fuel molten salt reactors as "first-of-a-kind" installations, establishing a nuclear safety experience feedback system and emphasizing testing during commissioning and operation. Before initial fuel loading, operating units must analyze and evaluate the safety impact of 7Li abundance on reactor reactivity control. During approach to criticality, batch fuel loading and conservative extrapolation methods should be used to prevent excessive reactivity insertion rates and limits due to high 7Li abundance. Additionally, excessive 6Li neutron absorption increases tritium effluent emissions, affecting environmental radiation safety. Long-term operation should prevent shutdown margin deviation from 6Li depletion. For licensing, operating units should analyze 7Li abundance impacts, considering laboratory analysis deviations. Regulatory recommendations are summarized as:

1. The National Nuclear Safety Administration and regional supervision stations must prioritize oversight of molten salt reactor 7Li laboratory analysis capabilities and abundance measurements during fuel loading, establishing operating limits based on consequence analysis.
2. Retest results of 7Li abundance before loading must be effectively communicated to commissioning and operation planning, particularly pre-criticality technical programs, to prevent reactivity 失控, serving as an important quality management inspection item.
3. Both regulators and operating units should monitor 7Li abundance changes with burnup, accumulating engineering experience, particularly regarding nuclear safety impacts for high-power, high-burnup reactors like nuclear power plants.

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