Discovery and Verification of a Formula Between Uranium Concentration and Reactivity in Thermal Spectrum Molten Salt Reactors

Authors: Yu, Changqing, Dr. Guifeng Zhu, Jia, Dr. Shuyang, Dr. Yang Zou (邹杨), Yan, Dr. Rui, Guo, Dr. Jian, Dr. Yafen Liu, Zhou, Dr. Bo, Zhao, Dr. Xuechao, Zhu, Dr. Guifeng

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Abstract

Knowing the precise relationship between fuel loading and reactivity helps guide the smooth progress of reactor criticality extrapolation and online refueling in molten salt reactors (MSRs). This study aims to explore and explain the linear relationship between reactivity and the reciprocal of uranium concentration in thermal spectrum MSRs. By applying the neutron balance theory, we analyzed neutron absorption by various nuclides under several single lattice models with varying fuel concentrations. Our findings reveal a simple linear correlation between reactivity and the reciprocal of uranium concentration, which is successfully explained from the perspective of nuclear reaction cross-sections that adhere to the 1/v law in a thermal neutron spectrum. Furthermore, we identified the single-group neutron absorption cross-sections of structural materials and carrier salts exhibit an approximate linear relationship with the single-group fission cross-section of 235U, and the reciprocal of the fission cross-section of 235U exhibits an approximate linear relationship with uranium concentration. This linear relationship will deviate as the volume fraction of molten salt continues to increase since more neutron will be captured in the resonance energy spectrum. But it remains valid within a 25% volume fraction of molten salt, and still demonstrates its broad applicability in the physical design and operation of thermal molten salt reactors.

Full Text

Preamble

Discovery and Verification of a Formula Between Uranium Concentration and Reactivity in Thermal Spectrum Molten Salt Reactors

Changqing Yu[1], Guifeng Zhu[1,2,†], Shuyang Jia[1,3], Yang Zou[1,2,‡], Rui Yan[1,2], Jian Guo[1,2], Yafen Liu[1,2], Bo Zhou[1,2], and Xuechao Zhao[1]

[1]Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China
[2]University of Chinese Academy of Sciences, Beijing 100049, China
[3]ShanghaiTech University, Shanghai 201210, China

Understanding the precise relationship between fuel loading and reactivity is essential for guiding the smooth execution of reactor criticality extrapolation and online refueling in molten salt reactors (MSRs). This study aims to explore and elucidate the linear relationship between reactivity and the reciprocal of uranium concentration in thermal spectrum MSRs. By applying neutron balance theory, we analyzed neutron absorption by various nuclides under several single-lattice models with varying fuel concentrations. Our findings reveal a simple linear correlation between reactivity and the reciprocal of uranium concentration, which can be successfully explained from the perspective of nuclear reaction cross-sections that adhere to the 1/v law in a thermal neutron spectrum. Furthermore, we identified that the single-group neutron absorption cross-sections of structural materials and carrier salts exhibit an approximate linear relationship with the single-group fission cross-section of ^{235}U, and the reciprocal of the fission cross-section of ^{235}U exhibits an approximate linear relationship with uranium concentration. This linear relationship will deviate as the volume fraction of molten salt continues to increase since more neutrons will be captured in the resonance energy spectrum. However, it remains valid within a 25% volume fraction of molten salt and demonstrates broad applicability in the physical design and operation of thermal molten salt reactors.

Keywords: Molten salt reactor, Reactivity, Uranium concentration, Cross-sections, Linear

Introduction

At the Generation IV International Forum (GIF), internationally renowned nuclear energy experts reached a consensus on the technological direction of fourth-generation reactor types, selecting six candidate reactor systems [1, 2]. Among these six Generation IV candidates, the molten salt reactor is the only one that employs liquid fuel, where the molten salt serves both as fuel and coolant [3–5]. As a liquid-fueled reactor, the fuel loading process and methodology in MSRs differ significantly from those in pressurized water reactors (PWRs) [6–8]. PWRs involve placing pre-fabricated fuel assemblies into the core in batches, with different fuel assembly types or structures arranged in various regions, leading to spatial heterogeneity in nuclear fuel distribution [9]. In contrast, MSRs typically add nuclear fuel to the core, which becomes uniformly distributed throughout the entire fuel salt loop as the salt circulates [10].

During reactor loading and criticality extrapolation processes, safe monitoring of reactor status is crucial to ensure controlled operation. Among the key param-

eters for monitoring the physical state of the reactor, the variation of reactivity with fuel loading amount is paramount. Due to the liquid fuel characteristics of MSRs, reactivity exhibits different physical relationships with loading amount compared to solid-fueled reactors.

Research on molten salt reactors dates back to the mid-20th century, with the most notable example being the Molten Salt Reactor Experiment (MSRE). MSRE achieved initial criticality on June 1, 1965, and remains the longest-operating molten salt reactor to date [11]. The initial loading process can be summarized as follows: Initially, 4560 kg of carrier salt (64.75% LiF-30.09% BeF_2-5.16% ZrF_4) and 236 kg of depleted uranium feed salt (73% LiF-27% $^{238}UF_4$) were thoroughly mixed outside the reactor vessel and then transferred through piping into the reactor vessel. Based on calculations, a certain amount of high-enriched uranium feed salt (73% LiF-27% UF_4, ^{235}U 93 wt%) was added after being similarly mixed outside the reactor vessel and then injected into the reactor vessel. This process was repeated multiple times until extrapolation results indicated the need to add approximately 1 kg of high-enriched uranium fuel. Using the feeding port of the pump, fuel capsules containing 150 g of high-enriched uranium feed salt (73% LiF-27% UF_4, ^{235}U 93 wt%) were gradually introduced. Solid molten salt flowed out from the openings of the fuel capsules after melting at high temperatures. Through pump operation, it was mixed into the fuel salt inside the reactor vessel, completing the initial critical loading process [11, 12]. Throughout the entire feeding process, the nuclear fuel was quickly and thoroughly mixed into the fuel salt system, effectively increasing the uranium concentration in the fuel salt.

Loading nuclear fuel and achieving initial criticality represents the most crucial step before reactor power operation, ensuring reactor safety and controllability. During reactor loading and criticality extrapolation processes, establishing the relationship between reactivity and the amount of nuclear fuel loaded forms the physical basis for designing loading schemes. The reciprocal neutron count rate extrapolation method is commonly used for critical loading extrapolation [13]. This method involves plotting the reciprocal of the neutron counting rate against the loading amount of nuclear fuel. Extrapolating this curve to its intersection with the horizontal axis allows estimation of the loading amount required to achieve criticality. The physical principle of this method is based on source multiplication theory [14]. According to source multiplication theory, the effective neutron multiplication factor k ff can be approximated as:

$$k\ ff = 1 - N_0/N$$

where N_0 represents the neutron detector count rate before loading nuclear fuel into the reactor, and N represents the count rate after loading a certain amount of nuclear fuel. As more nuclear fuel is loaded, the corresponding neutron count rate (N) increases. As the reactor approaches criticality, k ff approaches 1, and the corresponding neutron count rate N tends toward infinity. The reciprocal extrapolation method of neutron count rate approximates a linear relationship between k ff and the amount of fuel loaded. The curve typically exhibits con-

cavity because this proportional relationship is a conservative approximation; as the amount of nuclear fuel loading increases, the reactivity introduced per unit of fuel loading decreases. While conservative approximations are generally beneficial for reactor criticality safety, they can sometimes lead to disadvantageous judgments about reactor status.

During reactor startup in nuclear physics experiments, reactivity measurement is one of the primary means to characterize reactor status. Methods for measuring reactivity mainly include the source multiplication method [15, 16], inverse kinetics method [17], and period method [18]. These methods all require processing neutron signals captured by neutron detectors to obtain reactivity parameters. However, these measurement methods face considerable difficulties in achieving precise measurements under subcritical conditions due to factors such as the influence of external neutron source worth and spatial effects of control rods. To obtain more precise measurement results, some scholars resort to complex correction methods based on theoretical calculations [19, 20]. If the relationship between reactivity and nuclear fuel loading can be determined, it would enable direct calculation of reactor reactivity based on the amount of fuel loaded.

Due to the liquid fuel characteristics of MSRs, the reactor can achieve online refueling during operation [5, 21]. By adding nuclear fuel online to the fuel salt circuit, excess reactivity can be adjusted. Currently, the online refueling process is primarily simulated through coupled neutron transport and burnup codes. In simulation calculations, adjustment of the nuclear fuel addition rate is typically used to achieve k ff regulation, employing methods such as the secant method [22] and linear approximations of k ff with burnup depth or fuel addition rate [23]. These methods typically require multiple iterations of neutron transport calculations to update the feed rate of nuclear fuel.

Therefore, establishing a relationship between reactivity and nuclear fuel loading is not only related to reactivity measurement but also plays a key guiding role throughout the entire process of loading and criticality extrapolation, as well as in criticality search calculations related to fuel addition. In general, k ff is associated with various factors such as the amount of nuclear fuel, temperature, and spatial configuration of the reactor [24]. Establishing a simple theoretical relationship is typically challenging. However, as mentioned earlier, the feeding process in an MSR primarily involves increasing the uranium (or other nuclear fuel) concentration in the fuel salt without altering the reactor' s structure. Therefore, for MSRs, under the same operational conditions, there should exist a corresponding relationship between the uranium concentration in the fuel salt and reactor reactivity. Based on this idea, through analysis of the relationship between k ff and uranium concentration, we find a highly linear relationship between reactivity and the reciprocal of uranium concentration. Further research is needed to explore the theoretical basis and applicability of this linear relationship.

The objective of this paper is to establish the relationship between uranium

concentration in MSR fuel salts and reactivity through theoretical research and to verify the principles and applicability of this linear relationship. The main research content includes: Section 2 introduces the reactor model; Section 3 describes the discovery process of the linear relationship between 1/k ff and the reciprocal of uranium concentration; Section 4 involves principle verification based on the graphite-moderated single-lattice model using neutron balance theory to solve the relationship between k ff and uranium concentration step by step, establishing a theoretical explanation for the linear relationship between the reciprocal of k ff and the reciprocal of uranium concentration; Section 5 verifies the applicability of the linear relationship based on the graphite-moderated core model over a broader range of fuel loading, as well as under different molten salt volume fractions, various uranium enrichments, and ^{232}Th/^{233}U fuel; Section 6 provides a summary and discussion of this paper.

II. Reactor Model

This study focuses on the relationship between reactor reactivity and uranium concentration in molten salt reactors, using a graphite-moderated model as the subject of investigation. The reactor core utilizes graphite as a moderator and consists primarily of fuel salt channels, graphite moderators, graphite reflectors, and reactor vessel components, as shown in [Figure 1: see original paper]. The lattice structure is hexagonal prismatic, with a salt volume fraction of 15%, meaning that the fuel salt occupies 15% of the total volume. The core active region has consistent dimensions of 200 cm in diameter and height. The moderator and reflector graphite have a density of 2.3 g/cm^3, with a reflector thickness of 30 cm. The reactor uses a fuel salt composed of ^{7}LiF-^{33}BeF$_2$-xUF$_4$ (mol%). By adjusting the proportion of UF$_4$, the uranium loading in the fuel salt is changed, simulating the fuel addition process of the reactor. Additionally, the fuel salt can also contain a certain proportion of ThF$_4$ to simulate thorium loading, which will be analyzed in Section 5.4. The density of molten salts is calculated from the densities of unit salts [25] by volume-weighted average [26]. The parameters of the fuel salt are shown in .

Fuel salt parameters. Parameters: Fuel salt (mol%), Temperature, Salt density (g/cm^3), ^{7}Li abundance, ^{235}U enrichment. Value: ^{7}LiF-^{33}BeF$_2$-xUF$_4$, 900 K, LiF: 1.81-0.00049 • (T-848.2) BeF$_2$: 1.96-0.000015 • (T-552) UF$_4$: 6.485-0.00092 • (T-1036) ThF$_4$: 6.058-0.000759 • (T-1110), 99.95 at%, 20 wt%

The theoretical analysis based on the single-lattice model and the validation within the core model are derived from simulations performed using the SCALE 6.1 code [27]. SCALE 6.1 can provide reactor k ff, neutron energy spectra, and single-group cross-sections for various fuel compositions. It can simulate the variation of parameters such as k ff in a reactor during the fuel salt loading process as the uranium concentration increases. In this paper, during the criticality calculation, the number of particles is set to 20,000, and the selected database is the 238-group ENDF/B-VII library.

III. Discovery of the Linear Relationship

Before commencing the study presented in this paper, research initially focused on the reactor loading and criticality extrapolation process based on the reactor model depicted in Figure 1: see original paper, particularly on reactivity calculations during loading. As mentioned earlier, the simulation involves incrementally increasing the mole percentage of UF_4 in the fuel composition ^{7}LiF-$^{33}BeF_2$-xUF_4 (mol%), where x represents the amount of UF_4. Due to the liquid fuel characteristics of the MSR, each batch of nuclear fuel, once added, is quickly mixed and evenly distributed throughout the entire fuel channels of the reactor [28, 29]. Therefore, there should be a corresponding relationship between reactor reactivity and the amount of fuel added, without the need to consider spatial arrangement as in PWRs. Simulations using SCALE 6.1 can determine k ff after each batch of nuclear fuel is added, and attempts are made to identify the relationship between uranium concentration in the fuel salt or uranium loading and k ff.

During the extrapolation process for fuel addition, a practical approximation is employed where the increase in k ff is assumed to be linearly related to the amount of fuel added. Therefore, we initially analyze the relationship between k ff and the concentration of ^{235}U, as shown in Figure 2: see original paper. Here, M $_{235}$ refers to the atomic number density of ^{235}U in the fuel salt (units: atoms/b · cm). It can be observed that there is actually a concave curve relationship between k ff and M $_{235}$, approximating a linear relationship only when k ff approaches 1.

To establish the relationship between k ff and M $_{235}$, neutron balance theory is employed for analysis. According to neutron balance theory, the effective multiplication factor k ff of a reactor system can be defined as the ratio of neutron production rate to neutron loss rate [30]:

k ff = neutron production rate / neutron loss rate (absorption + leakage)

The effective multiplication factor k ff is influenced by the system' s material composition, structure, and neutron leakage. The mathematical expression is written as follows:

k ff = R_f / (R_a + L)

In Eq. (3), represents the average number of neutrons emitted per fission, R_f is the fission reaction rate, R_a is the total absorption reaction rate (including fuel salt, graphite, and structural materials), and L is the neutron leakage rate. Eq. (3) involves the solution of neutron flux and reaction cross-sections, requiring neutron transport calculations. From a qualitative perspective, the reaction rate of uranium is directly proportional to the concentration of ^{235}U. Assuming further that the neutron absorption reaction rate R_c of materials other than uranium isotopes is constant, Eq. (3) can be approximated and rewritten as:

k ff $c_{01}M_{235}$ / ($c_{02}M_{235}$ + R_c + L)

In Eq. (4), c_{01} and c_{02} are both constants. Taking the reciprocal of both sides of Eq. (4) and assuming that and L are also constants, it can be rewritten in the following form:

1/k ff a • (1/M $_{235}$) + b

Eq. (5) suggests that 1/k ff may exhibit an approximate linear relationship with 1/M $_{235}$. To verify whether Eq. (5) holds, Figure 2: see original paper is transformed by swapping the horizontal axis to 1/M $_{235}$ and the vertical axis to 1/k ff, as shown in Figure 2: see original paper. A linear fit is performed on 1/k ff and 1/M $_{235}$, yielding a linear determination coefficient R^2 as high as 0.99997. Therefore, although Eq. (5) is derived from a rough qualitative analysis of Eq. (3), the results shown in Figure 2: see original paper suggest that Eq. (5) may be valid. Eq. (5) indicates that reactor reactivity can be quickly obtained through uranium loading or uranium concentration, which would be a beneficial discovery for the reactor loading process.

Due to the liquid fuel characteristics of MSRs, nuclear fuel loading is equivalent to increasing the uranium concentration in the fuel salt. Based on the qualitative analysis from Eq. (2) to Eq. (5) and the linear regression analysis of Figure 2: see original paper, we have discovered that reactor reactivity or 1/k ff exhibits a linear relationship with the reciprocal of uranium concentration. However, during actual fuel loading, the neutron absorption reaction cross-sections of various nuclides within the reactor are changing, and the assumption in Eq. (4) that c_{01}, c_{02}, and R_c are constants may not hold true, necessitating further research.

Moving forward, this paper utilizes neutron balance theory to examine neutron energy spectrum and reaction cross-section variations in response to changes in uranium concentration. It progressively solves Eq. (3) and theoretically validates the linear relationship proposed in Eq. (5).

IV. Principle Validation

To investigate the relationship between reactivity and uranium concentration using neutron balance theory, we utilize the single lattice cell model illustrated in Figure 1: see original paper. The single lattice model omits intricate core configurations, thereby eliminating the need to account for neutron absorption by structural materials [31, 32]. Furthermore, to neglect the influence of neutron leakage, a white reflective boundary condition is applied to the single lattice structure. During calculations in SCALE 6.1, the concentration range of UF_4 is 0.02-1 mol%, corresponding to a k ff range of 0.12-1.42, covering a sufficiently wide range. Therefore, the neutron absorption reaction in Eq. (3) includes only the fuel salt and graphite moderator materials, with a leakage rate (L) equal to zero. The equation can be further expanded as:

k ff = (σ $_{235}$ M $_{235}$ + σ $_{238}$ M $_{238}$) / [σ $_{235}$ M $_{235}$ + σ $_{238}$ M $_{238}$ + Σ σ M]

In the equation, σ $_{235}$, σ $_{238}$, σ $_{235}$, σ $_{238}$, M $_{235}$, and M $_{238}$ respectively

denote the microscopic single-group cross sections for fission and absorption of ^{235}U and ^{238}U, as well as their atomic number densities. The terms σ and M represent, respectively, the single-group absorption cross-sections and atomic number densities of nuclides other than ^{235}U and ^{238}U, where i includes ^{6}Li, ^{7}Li, ^{9}Be, ^{19}F, and ^{12}C. Due to the approximate proportionality between the average neutron flux density in the graphite moderation region and the fuel region, the single-group cross-section of ^{12}C underwent neutron flux equivalence treatment.

Upon careful observation of Eq. (6), the enrichment of ^{235}U is 20 wt%, establishing a fixed proportional relationship between M $_{235}$ and M $_{238}$. Additionally, during the fuel loading process, changes in the concentrations of other nuclides in the fuel salt can be considered negligible aside from uranium. Regarding the average number of fission neutrons ( ), since the fissile material is uranium with fixed enrichment, it also approximates a constant. Therefore, the primary variables on the right-hand side of the equation are the single-group cross-sections of each nuclide and the uranium concentration. Next, we examine the variation of single-group cross-sections with uranium concentration, incorporating the neutron energy spectrum, and gradually simplify Eq. (6) accordingly.

A. Variations in Energy Spectrum and Cross-Sections

The single-group cross-sections are primarily related to the neutron energy spectrum, calculated as follows:

$$\bar{\sigma} = \sigma(\mathrm{E})\ (\mathrm{E})\mathrm{dE}\ /\ \ (\mathrm{E})\mathrm{dE}$$

The term σ(E) represents the nuclear reaction cross-section for energy distribution. This value remains constant for a given nuclide and reaction type, typically determined through experimental measurements. This article utilizes cross-section library data from the ENDF/B-VII database. (E) represents the neutron flux distribution in the region where the nuclide is present, i.e., the neutron energy spectrum. The neutron energy spectrum is influenced by various factors such as reactor structure, uranium concentration, and temperature. It is a result of neutron transport and can be calculated using the SCALE 6.1 program.

Figure 3: see original paper shows the normalized neutron energy spectra in the fuel region at different uranium concentrations, along with the cross-sections of key neutron absorption reactions in a single lattice. For fissionable isotopes, the primary neutron absorption reactions of ^{235}U and ^{238}U are fission (n,f) and capture (n,γ) reactions, respectively. The primary reaction type for ^{6}Li is tritium production through the (n,T) reaction. The main nuclear reaction type for other light nuclei is capture absorption (n,γ) reactions. Additionally, for the nuclides ^{9}Be and ^{19}F, there is also a significant proportion of (n,α) absorption reactions in the high-energy region, which absorb neutrons to produce ^{4}He particles.

Upon observation, it is noted that although the neutron flux decreases in the thermal neutron region (E $<$ 1 eV) with increasing uranium concentration, there

is a corresponding increase in other energy regions. However, overall, the neutron flux in the thermal neutron region remains significantly higher compared to the fast neutron region, indicating a predominantly thermal neutron spectrum. Furthermore, although the neutron flux in different energy regions varies with uranium concentration, the overall shape characteristics of the spectrum remain essentially unchanged. The variation of reaction cross-sections for major nuclides with energy is shown in Figure 3: see original paper. For the two fissionable nuclides, ^{235}U and ^{238}U, their fission and capture cross-sections in the thermal neutron region both follow the 1/v law, meaning σ(E) is proportional to $1/\sqrt{E}$. In the intermediate energy region, they exhibit strong resonance peaks, particularly noticeable in the case of ^{238}U. Other light nuclei' s neutron absorption cross-sections, except for the reactions ^{9}Be(n,α) and ^{19}F(n,α), follow the 1/v law across larger energy ranges.

Based on the analysis of neutron energy spectrum and neutron absorption reaction cross-sections, the following three main conclusions can be drawn:

1. The neutron energy spectrum is a thermal neutron spectrum, where the thermal neutron flux (E < 1 eV) is significantly higher than other neutron flux components.
2. The neutron absorption cross-sections of major nuclides in the thermal neutron energy region follow the 1/v law, meaning σ(E) is proportional to $1/\sqrt{E}$.
3. In the high-energy neutron range, isotopes such as ^{235}U, ^{238}U, and ^{19}F exhibit prominent resonance absorption peaks. Additionally, ^{9}Be and ^{19}F have significant (n,α) neutron absorption cross-sections in the fast neutron energy range.

Based on these three conclusions and Eq. (7), we can qualitatively infer that the single-group neutron absorption cross-sections of major nuclides may exhibit an approximate linear relationship with the single-group fission cross-section of ^{235}U. However, due to strong resonance absorption peaks in isotopes such as ^{235}U and ^{238}U, as well as (n,α) reactions in ^{9}Be and ^{19}F, deviations from a linear relationship may occur. To further illustrate this issue, Figure 3: see original paper shows the normalized neutron reaction rates versus energy distribution curves for major nuclides under critical conditions, with uranium comprising 0.36 mol%. From Figure 3: see original paper, it can be seen that in the thermal neutron energy region (E < 1 eV), the neutron reaction rates of the major nuclides are ranked as follows: ^{235}U(n,α) > ^{235}U(n,f) > ^{6}Li(n,α) > ^{12}C(n,α) > ^{7}Li(n,α) > ^{19}F(n,α) > ^{238}U(n,α) > ^{9}Be(n,α). For major nuclides such as ^{235}U, ^{6}Li, ^{12}C, and ^{7}Li, the neutron absorption reaction rates are significantly higher in the thermal neutron region compared to other energy ranges. When calculating k ff using Eq. (6), it is essential to consider primarily the neutron reaction rates in the thermal neutron region. In the resonance energy region, both ^{238}U and ^{235}U exhibit significant absorption reactions. For ^{238}U, resonant absorption in the energy range of 1 eV to 1 keV constitutes approximately 75% of its total absorption, yet this accounts for only about 2% of the overall material

absorption. In addition, due to the (n,α) reactions of ^{9}Be and ^{19}F, these two nuclides have a certain proportion of neutron absorption reaction rates in the fast neutron energy region, accounting for approximately 45% and 19% of their total absorption, respectively. However, these reaction rates are significantly smaller than the absorption reaction rates of nuclides such as ^{235}U and ^{6}Li in the thermal neutron region. Therefore, for several nuclides that contribute significantly to neutron absorption reactivity, the focus is primarily on thermal neutron absorption reactions, where the neutron absorption cross-sections follow the 1/v law. Although some nuclides also contribute to neutron absorption reactions in the resonance and fast neutron energy regions, these contributions are relatively small.

To verify the above analysis, we use SCALE 6.1 to calculate the single-group neutron absorption cross-sections for major nuclides and analyze their linear relationships with the ^{235}U fission cross-section. [Figure 4: see original paper] shows the linear relationship between the single-group neutron absorption cross-sections of ^{235}U, ^{238}U, ^{6}Li, ^{7}Li, ^{9}Be, ^{19}F, and ^{12}C relative to the single-group fission cross-section of ^{235}U. The corresponding coefficient of determination (R^2) for each linear fit is provided. [Figure 4: see original paper] demonstrates that the neutron absorption cross-sections of various nuclides show a strong linear relationship with the fission cross-section of ^{235}U. This linear relationship can be summarized as follows:

$$\sigma = c_1 \sigma_{235} + c_2$$

In the equation, σ represents the single-group neutron absorption cross-section of nuclide i, σ_{235} denotes the single-group fission cross-section of ^{235}U, where c_1 and c_2 represent the slope and intercept of the linear fit for nuclide i, which includes nuclides ^{235}U, ^{238}U, ^{6}Li, ^{7}Li, ^{9}Be, ^{19}F, and ^{12}C. Meanwhile, [Figure 4: see original paper] shows that the coefficient of determination (R^2) for the nuclides ^{238}U and ^{9}Be is relatively smaller. This is primarily due to ^{238}U exhibiting a strong resonance absorption peak in the intermediate neutron energy region, and the nuclide ^{9}Be having a relatively large (n,α) absorption cross-section in the fast neutron energy region, as depicted in Figure 3: see original paper or Figure 3: see original paper. An increase in uranium concentration results in a harder neutron energy spectrum, enhancing resonance and (n,α) absorption, thereby deviating from the linear relationship. However, since the single-group neutron absorption cross-sections of these nuclides are significantly smaller compared to major nuclides like ^{235}U and ^{6}Li, the use of Eq. (8) for linear approximation will not greatly affect the calculation of k ff in Eq. (6).

Thus, we can convert all single-group absorption cross-sections in Eq. (6) into the fission cross-section of ^{235}U. Therefore, Eq. (6) can be further simplified to:

$$k_{ff} = (c_{2351} + wc_{2381})\sigma_{235} M_{235} / [(c_{2351} + wc_{2381})\sigma_{235} M_{235} + \Sigma (c_1 \sigma_{235} + c_2) M]$$

The constant w in the denominator of Eq. (9) is associated with the enrichment (wt) of ^{235}U, and its value equals (1-wt)\$×235/(*wt*×\$238). Meanwhile, as

shown in Figure 3: see original paper, the fission cross-section of ^{238}U is lower by four orders of magnitude compared to that of ^{235}U. Therefore, an approximate treatment can neglect the contribution of ^{238}U' s fission. Moreover, compared to the fission cross-section of ^{235}U, the intercept of the linear fit of the absorption cross-sections of ^{235}U and ^{238}U amounts to approximately 1% of σ_{235}, which can be neglected. The linear relationship between the absorption cross-sections of ^{235}U and ^{238}U and the fission cross-section of ^{235}U is approximately proportional. Therefore, Eq. (9) can be further simplified to:

$$k_{ff} = (c_{2351} + wc_{2381})\sigma_{235} M_{235} / [(c_{2351} + wc_{2381})\sigma_{235} M_{235} + \Sigma c_1 \sigma_{235} M]$$

In Eq. (11), the slopes c_1 and intercepts c_2 associated with the linear fit, where i includes ^{235}U and ^{238}U along with other nuclides, as well as M , are constants. Thus, k ff has been simplified to depend solely on the fission cross-section σ_{235} of ^{235}U and its concentration M_{235}. Next, by analyzing the relationship between the fission cross-section σ_{235} and the concentration M_{235}, Eq. (11) will be further streamlined.

B. Relationship Between Fission Cross-Section and Uranium Concentration

In Eq. (6), we express k ff as a function of cross-sections and nuclide concentrations and establish the relationship between neutron absorption cross-sections in various nuclides and the fission cross-section of ^{235}U in Eq. (8). Therefore, under the condition where the concentration of non-fissile nuclides is approximately constant, there are currently two quantities related to k ff on the right side of Eq. (11): M_{235} and σ_{235}. Next, we further establish the relationship between M_{235} and σ_{235}.

From Eq. (7), it can be seen that the calculation of the single-group cross-section is directly related to the neutron spectrum, indicating that the neutron spectrum determines the magnitude of the cross-section. Moreover, changes in the spectrum are directly caused by fuel loading, implying an inherent connection between the spectrum and uranium concentration. As uranium concentration increases, neutron absorption strengthens, reducing the moderator capacity of the fuel salt and resulting in spectrum hardening, as shown in Figure 3: see original paper. Establishing the relationship between neutron spectrum and uranium concentration is a challenging task that requires solving for the neutron spectrum. If we use the free gas model, the neutron spectrum follows a Maxwell-Boltzmann distribution. However, due to the continuous generation of fission neutrons and absorption by the medium during the neutron moderation process, the neutron spectrum shifts toward higher energies [33]. The approach in this work does not involve seeking analytical solutions for the neutron energy spectrum and subsequently using Eq. (7) to solve for the fission cross-section. Instead, neutron transport calculations are directly employed to obtain the energy spectrum. Single-group cross-sections are processed to estab-

lish their relationship with uranium concentration. This aspect of the work can be accomplished using SCALE 6.1. A fitting analysis has been conducted to examine the relationship between σ_{235} and M_{235}, and the results indicate that $1/\sigma_{235}$ and M_{235} exhibit a highly linear relationship, with a linear determination coefficient R^2 reaching as high as 0.99998. This linear relationship can be described as:

$$1/\sigma_{235} = c_3 M_{235} + c_4$$

In Eq. (12), c_3 and c_4 are respectively the slope and intercept of the linear fit, which are constants. The results of the linear fit are shown in [Figure 5: see original paper]. Qualitatively, for Eq. (12), as the amount of uranium increases, the neutron energy spectrum hardens, and the single-group fission cross-section decreases. The linear relationship may be related to the 1/v law for the fission cross-section in the thermal neutron energy region, and the theoretical methods based on the energy spectrum still need further exploration.

Substituting Eq. (12) into Eq. (11) allows for further simplification of Eq. (11), resulting in:

$$k_{ff} = M_{235} / (a + bM_{235})$$

Here, a and b are constants: $a = (\Sigma c_2 M)/$ and $b = (c_3 \Sigma c_1 M + c_{2351} + wc_{2381})/$. In Eq. (13), k_{ff} depends solely on the concentration of ^{235}U. Taking the reciprocal of both sides of Eq. (13) yields Eq. (5), demonstrating that $1/k_{ff}$ or reactivity is linearly related to the reciprocal of uranium concentration.

It can be observed that from Eq. (6) to Eq. (13), we have made several approximations, which are summarized as follows:

1. The single-group absorption cross-section of the main nuclides is linearly related to the single-group fission cross-section of ^{235}U, as shown in [Figure 4: see original paper] or Eq. (8). The absorption cross-sections of ^{235}U and ^{238}U approximately have a proportional relationship to the ^{235}U fission cross-section.
2. $1/\sigma_{235}$ exhibits a linear relationship with M_{235}, as shown in [Figure 5: see original paper] or Eq. (12).
3. The average number of neutrons per fission, , is constant.
4. Changes in concentrations of other nuclides besides uranium are excluded.

V. Validation of Applicability

In the previous section, we employed the single lattice model depicted in Figure 1: see original paper to explain the basis for the validity of Eq. (5). To neglect the influence of neutron leakage, the single lattice model adopted a white reflective boundary condition. By examining the variation of single-group cross-sections during changes in uranium concentration and using reasonable approximations, we successfully explained the linear relationship between reactivity and the reciprocal of uranium concentration. However, as shown in Figure

1: see original paper, in the typical core structure of a reactor, apart from the core' s active region, there are also reflector layers and various structural materials. These materials absorb neutrons to varying degrees and alter the neutron energy spectrum. Nevertheless, since these structures do not vary with uranium concentration changes, similar to the single lattice structure, these structures can be physically equivalent to a graphite moderation structure. Therefore, Eq. (5) or Eq. (13) should still hold in the core model. To verify this conclusion, we simulate and compute the variation of k ff with uranium concentration in the core model.

A. Verification of the Linear Relationship

Firstly, based on the core model in Figure 1: see original paper, we verify the linear relationship between 1/k ff and 1/M $_{235}$ under the condition that the volume fraction (VF) and uranium enrichment (wt) of the molten salt channel in the single-lattice model are the same (i.e., VF equals 15% and wt equals 20%). Simultaneously, the uranium concentration is expanded from 0.02-1 mol% to 0.02-3 mol%. The purpose of this section is to demonstrate the applicability of the aforementioned linear relationship within the core model.

The method involves using SCALE 6.1 to calculate the k ff of the reactor at different uranium concentrations and to establish the relationship between reactivity and uranium concentration. As shown in Figure 6: see original paper, the reciprocal of the effective multiplication factor k ff, i.e., 1/k ff, exhibits a highly linear relationship with the reciprocal of uranium concentration (1/M $_{235}$). The fitted coefficient of determination R^2 is 0.99995, very close to 1. Furthermore, with ^{235}U concentrations ranging from 1.4$×10^{-6}$ to 2.0$×10^{-4}$ atoms/b · cm, the k ff values span from 0.08 to 1.43, covering nearly all possible fuel loading scenarios. This verifies the conclusion that reactivity, derived from the single-lattice model, is linearly related to the reciprocal of uranium concentration.

To further illustrate the accuracy of the aforementioned linear relationship, Figure 6: see original paper presents the statistical error in calculating k ff directly by the SCALE 6.1 program, as well as the relative deviation between k ff derived from the linear relationship in Figure 6: see original paper and the results calculated by SCALE 6.1 (i.e., the difference between the k ff values obtained by the two methods). From Figure 6: see original paper, it can be observed that the error in calculating k ff by SCALE 6.1 is essentially maintained between approximately 30 to 50 pcm. Although the relative deviation between the k ff calculated through the linear relationship in Figure 6: see original paper and that of SCALE 6.1 fluctuates more significantly, within the range of k ff from 0.7 to 1.1, the deviation is essentially within $±$100 pcm. Therefore, Figure 6: see original paper directly demonstrates the accuracy of calculating k ff through the linear relationship shown in Figure 6: see original paper, within the allowable error range.

B. Impact of Molten Salt Volume Fraction (VF)

In the process of deriving the linear relationship, the most important approximation used is that the neutron absorption cross-sections of all nuclides are linearly related to the fission cross-section of ^{235}U, represented by approximation condition (1). The main rationale for this approximation is that in graphite-moderated molten salt reactors, the neutron energy spectrum is thermal, which diminishes the impact of resonance absorption and fast neutron absorption reactions. When the fuel composition is the same, the neutron spectrum is primarily influenced by the proportion of molten salt volume within the graphite channels. This section discusses primarily the applicability of the aforementioned linear relationship under different volume fraction conditions.

The verification method involves keeping the enrichment of ^{235}U constant at 20 wt% and calculating the relationship between k ff and uranium concentration at different VF values using SCALE 6.1. The range of VF is from 5% to 40%.

First, observe how the neutron energy spectrum changes as VF varies. [Figure 7: see original paper] shows the normalized neutron flux distribution as a function of energy for different VF values. At different VF values, with the uranium loading remaining constant, the corresponding concentration of ^{235}U is 3.81$×10^{-5}$ atom/barn·cm. For VF=15%, k ff=1. [Figure 7: see original paper] shows that the neutron energy spectrum undergoes significant changes as VF varies. The main effect is that as VF increases, the neutron flux decreases in the thermal region (E < 1 eV), while correspondingly, the neutron flux increases in the intermediate and fast neutron energy regions. Combining the neutron absorption cross-sections of various nuclides from Figure 3: see original paper, it can be seen that the relative increase in neutron flux in the resonance or fast neutron energy regions leads to a relative decrease in neutrons that conform to the 1/v law. As a result, this will inevitably lead to degradation in the linearity between the absorption cross-sections of various nuclides and the fission cross-section of ^{235}U, meaning that approximate condition (1) no longer holds, ultimately causing deviation from a linear relationship between reactivity and $1/M_{235}$.

Figure 8: see original paper shows the local curves of the relationship between 1/k ff and $1/M_{235}$ at different VF values. It can be observed that as VF increases, the curve gradually bends, with a more pronounced bend at higher VF values. Correspondingly, as VF increases, the determination coefficient R^2 of the linear fit between 1/k ff and $1/M_{235}$ gradually decreases, meaning it deviates increasingly from the linear relationship. Figure 8: see original paper directly illustrates that an increase in VF, or the hardening of the neutron energy spectrum, leads to deviation from the linear relationship between reactivity and the reciprocal of uranium concentration. This deviation is a gradual change with VF variation, and the applicability of the linear relationship does not have a strict boundary; it is mainly related to error requirements. Generally speaking, for a graphite-moderated molten salt thermal neutron reactor, the optimal

design range for VF is 10-20% [31, 34, 35]. Within this range, the determination coefficient remains above 0.9999, indicating a highly linear relationship between reactivity and the reciprocal of uranium concentration.

C. Impact of Uranium Enrichment

As shown in Figure 3: see original paper, ^{238}U has strong resonance capture cross-sections, and its varying content in the fuel salt may result in different relationships between reactivity and uranium concentration. Furthermore, to validate the applicability range of the linear relationship in Eq. (3), we continue studying the relationship between reactor reactivity and uranium concentration under various uranium enrichments. The verification method involves keeping VF constant at 15% and using SCALE 6.1 to calculate the relationship between k ff and uranium concentration under different enrichments of ^{235}U. In the study, the enrichment range of ^{235}U in UF_4 ranges from 4 to 20 wt%, and the effect of enrichment variation on the density of UF_4 is neglected during calculation.

Figure 8: see original paper presents the local curves of the relationship between 1/k ff and the reciprocal of uranium concentration at different uranium enrichments, as well as the determination coefficient R^2 obtained through linear fitting. From Figure 8: see original paper, it can be seen that within the range of 4-20 wt% uranium enrichment in the fuel salt, there is a strong linear correlation between 1/k ff and 1/M $_{235}$, without any deviation from the linear relationship as shown in Figure 8: see original paper. Meanwhile, the linear correlation coefficients at different enrichments are all greater than 0.99995, indicating the applicability of this linear relationship across varying enrichment conditions. The determination coefficient obtained from the linear fits at different ^{235}U enrichments exhibits some fluctuation, but since the values are all above 0.99995, the range of fluctuation is within the computational precision of the linear fitting and can be considered due to computational error. Therefore, we can conclude that under various low-enriched uranium conditions, there exists a strong linear relationship between reactivity and the reciprocal of uranium concentration. The reason is that although uranium enrichment varies several-fold, the overall absorption cross-section of ^{238}U is much smaller compared to the fission cross-section of ^{235}U [36]. Therefore, the multiple-fold change in enrichment does not greatly affect the calculation of k ff using Eq. (6). However, changes in volume fraction (VF) have a much more significant impact, altering the neutron spectrum noticeably. This affects the neutron absorption cross-sections of all isotopes, including ^{238}U. Thus, when VF becomes too large, nonlinear deviations occur from the linear relationship. Comparing Figure 8: see original paper and Figure 8: see original paper, it can be observed that the intercept of the linear fit varies with different uranium enrichment levels, while the slope remains almost unchanged. However, under different VF values, both the intercept and slope change. The reasons behind this can be seen in Eq. (14) and Eq. (15). When uranium enrichment changes, the neutron energy spectrum varies only slightly, so c does not change significantly, and consequently, the

slope (a) remains relatively stable. However, the intercept (b) is related to the enrichment level and thus changes accordingly. When the value of VF changes, the neutron energy spectrum undergoes significant changes, and accordingly, the c values also change, resulting in variation of both slope and intercept.

D. ^{232}Th/^{233}U Fuel

Based on the analysis of ^{235}U/^{238}U fuel, it can be seen that the main reason for reactivity being linearly related to the reciprocal of uranium concentration is that, in the thermal neutron energy region, the neutron absorption cross-sections of the primary nuclides follow the 1/v law. The fission cross-section of ^{233}U is similar to that of ^{235}U and also follows the 1/v law. Therefore, the same linear relationship observed in the analysis should also exist for ^{232}Th/^{233}U fuel. To verify this conclusion, the relationship between reactivity and concentration is examined using ^{232}Th/^{233}U fuel. Using the core model shown in Figure 1: see original paper, the fuel salt composition is ^{7}LiF-^{33}BeF$_2$-(0-8)ThF$_4$-xUF$_4$ (mol%). The ThF$_4$ content ranges from 0 to 8 mol%, aiming to validate the applicability of the linear relationship under different thorium loading conditions. Under different thorium loadings, reactivity is adjusted by varying the amount of UF$_4$, i.e., the value of x. The verification results are shown in Figure 8: see original paper. It can be seen that, under different thorium loadings, 1/k ff and 1/M $_{233}$ exhibit a strong linear relationship, demonstrating the applicability of this linear relationship to ^{232}Th/^{233}U fuel.

VI. Conclusion

Due to its liquid fuel characteristics, the graphite-moderated molten salt reactor considers the fuel loading process as equivalent to increasing the uranium concentration in the fuel salt. There exists a corresponding relationship between uranium loading or concentration and reactivity. The paper first investigates the variation of neutron spectrum and reaction cross-sections with uranium concentration using a single lattice cell model. Under certain approximations, it derives a linear relationship between reactivity and the reciprocal of uranium concentration based on neutron balance theory. Subsequently, the applicability of this linear relationship is validated through simulation calculations using a core model. Based on these studies, the following main conclusions are drawn:

1. Due to the thermal neutron spectrum in graphite-moderated molten salt reactors and the fact that neutron absorption cross-sections of various nuclides follow the 1/v law at low energies, the neutron single-group absorption cross-sections of each nuclide approximately exhibit a linear relationship with the single-group fission cross-section of ^{235}U.
2. The reciprocal of the ^{235}U single-group fission cross-section is linearly related to uranium concentration.
3. Reactor reactivity is linearly related to the reciprocal of uranium concentration, a conclusion drawn on the basis of the two aforementioned premises and the main conclusion that this paper aims to demonstrate.

4. In conclusion (3), the linear relationship weakens as the volume fraction of graphite molten salt channels increases. However, within the optimal design range of VF=10-20%, the coefficient of determination (R^2) for this linear relationship remains greater than 0.9999, indicating a high degree of linearity.
5. In conclusion (3), the linear relationship within the range of ^{235}U enrichment from 4-20 wt% is minimally affected by uranium enrichment. At VF=15%, the linear coefficient of determination remains above 0.99995 across different enrichments, demonstrating a highly linear relationship.
6. For $^{232}Th/^{233}U$ fuel, this linear relationship still holds strongly.

The relationship between reactor reactivity and nuclear fuel loading is generally considered complex. When nuclear fuel loading changes, it usually requires complex transport calculations, which are time-consuming and labor-intensive, to determine the impact. During the fuel loading process in experiments, reactors are often in a subcritical or deep subcritical state, making reactivity measurements challenging. This paper establishes a simple relationship between reactivity and uranium concentration through simulation analysis, which can be used for calculating or measuring reactor reactivity. Moreover, it effectively establishes a connection between the amount of nuclear fuel loaded and reactivity, applicable in numerous scenarios such as critical extrapolation during the fuel loading process. This research on the relationship between reactor reactivity and uranium concentration has established a simple linear relationship through theoretical analysis and simulation verification. This relationship has significant application value in both theoretical analysis and engineering experiments of molten salt reactors. Additionally, it should be noted that the reactor model used in this paper is a single-zone structure, meaning all lattices have the same VF. For molten salt reactors with more complex partition designs, the relationship between reactivity and uranium concentration may take different forms and requires further validation.

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Author Contributions

Changqing Yu oversees the conceptualization, writing, and data analysis of the article, while Guifeng Zhu contributes to the funding, conceptualization, and writing. Shuyang Jia makes significant contributions to the revision of articles. Yang Zou, Rui Yan, Jian Guo, Yafen Liu, Bo Zhou, and Xuechao Zhao provide suggestions for analysis and assist with writing revisions.

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