

CONFERENCE PRE-PRINT

NUMERICAL SIMULATION OF COMPOSITIONAL
REDISTRIBUTION DRIVEN BY ISOTOPOLOGUE
FRACTIONATION DURING SOLIDIFICATION
OF D-T FUEL IN ICF TARGETS

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Abstract

In inertial confinement fusion, compositional homogeneity of cryogenic deuterium-tritium fuel target is critical for fusion performance. However, isotopologue fractionation causes a radial concentration gradient within the solid D-T layer and alters the equilibrium composition of the central D-T gas. This study investigates isotopologue fractionation of D-T mixtures under various conditions of solidification, analyses the factors that affect the homogeneity of target. A comprehensive numerical framework integrating heat transfer, species transport, and phase transition was developed using ANSYS Fluent to simulate the compositional distribution in D-T mixtures during solidification process. The simulation results show that D atom distribution in the solid layer exhibits a radial mole fraction gradient; D atom content in the central gas is significantly in excess. The detrimental effects of composition inhomogeneity on ICF performance in multiple stages were examined. This study provides a quantitative simulation framework for understanding isotopologue fractionation in ICF target and offers theoretical guidance for design and fabrication of high-performance, homogeneous solid D-T layers.

1. INTRODUCTION

In inertial confinement fusion (ICF), a typical cryogenic deuterium-tritium (D-T) fuel target consists of a spherical multilayered structure with an ablator shell, a solid D-T fuel layer, and a central D-T vapor core [1], as shown in Fig. 1(a). Minor structural imperfections in target can significantly degrade implosion performance [2]. However, compositional inhomogeneity in the D-T fuel remains a critical but underexplored factor. The most common technique for forming a solid D-T layer is β -layering, in which heat from tritium beta decay drives repeated sublimation that redistribute D-T to form the solid layer [3]. During phase transition, isotopologue fractionation can produce a radial concentration gradient across the solid D-T layer (Fig. 1(b)). Such a gradient compromises fuel homogeneity, thereby impairing implosion uniformity and fusion performance [4].

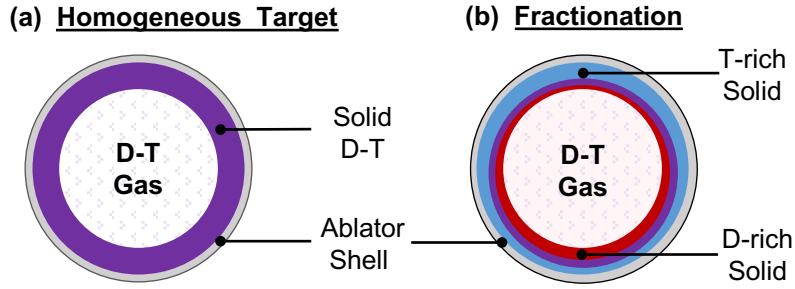


FIG. 1. Schematic of the effect of isotopologue fractionation in ICF target. (a) Target with homogeneous fuel. (b) Target with isotopologue fractionation.

However, there is currently no established quantitative method for characterizing the composition distribution within the D-T target. Experiments on hydrogen-deuterium (H-D) mixtures have demonstrated isotopologue fractionation during solidification [5,6]. Since tritium is radioactive and difficult to handle, numerical simulation is an indispensable tool for analysing fractionation behaviour in D-T fuel. This study developed a three-dimensional (3D) model for cryogenic D-T target to simulate the compositional distribution of fuel during solidification, and to evaluate the impact of compositional inhomogeneity on target performance. This work advances understanding of isotopologue fractionation mechanisms and offers theoretical support for designing and fabricating high-quality, homogeneous solid D-T layer.

2. COMMON SIMULATION SETTINGS

During nonequilibrium solidification of the D-T mixture, the accumulation of kinetic fractionation leads to macrosegregation. Heavier T_2 molecules preferentially enrich into the solid phase, whereas lighter D_2 molecules are rejected into the remaining gas or liquid phase [7], resulting in a concentration gradient within the solid. Because diffusion in the solid state is extremely slow, the gradient cannot be eliminated, ultimately leading to macrosegregation in solid [8]. Previous studies have successfully simulated macrosegregation in hydrogen isotopologue mixtures during unidirectional solidification using ANSYS Fluent (Ansys, Inc.) [9]. These simulations reproduced experimental results, validating the underlying theoretical framework. Building on this foundation, the present study extends the simulation to a 3D model of a D-T fuel target to analyse the compositional homogeneity.

2.1. Assumptions

To simplify the mathematical model, the following assumptions are introduced:

- The effect of decay heat is neglected.
- The influence of gravity is neglected. Liquid D-T forms a hollow spherical shell.
- Liquid D-T is treated as an incompressible Newtonian fluid, and flow is assumed to be laminar.
- The gas phase is considered a pure substance that does not participate in species transport and undergoes no phase transitions.
- The liquid and solid phases are assumed to have identical thermophysical properties.
- Volume shrinkage during the phase transition is neglected.

2.2. Input parameters

The simulation framework integrates heat transfer, species transport, and phase equilibrium [10]. Thermophysical properties of species and the liquidus and solidus curves of the mixture phase diagram [11] are required as input parameters. The detailed values are available in Ref. [9]. Boundary conditions are defined as follows:

- Initial condition: The D-T mixture is entirely in a homogeneous liquid phase at 19.7 K, with $D_2 : T_2 = 1:1$.
- Wall surface: Heat transfer occurs only through the outer wall surface of the sphere model. The solidification rate is controlled by adjusting the wall temperature, initially set to 19.55 K.

2.3. Numerical solution procedure

A pressure-based solver is used to perform a transient analysis. Relevant physical models include energy equation, laminar flow, species transport, and solidification and melting. The fluid domain consists of a non-interpenetrating gas phase and a liquid phase. The energy equation is solved to obtain the temperature field. Solidification is realized through the solidification and melting model, the mushy zone constant is 100,000. The compositions of the liquid and solid phases are determined using the lever rule.

3. SIMULATION OF COMPOSITIONAL DISTRIBUTION DURING SOLIDIFICATION

3.1. Compositional distribution during unidirectional solidification

Solidification in practice are non-equilibrium processes, and diffusion in the solid phase is too slow to homogenize the composition. The resulting macroscopic inhomogeneity in the solid is referred to as macrosegregation [12]. The slower the solidification rate, the more sufficient the diffusion in the liquid phase, leading to more pronounced macrosegregation [13]. When diffusion in the liquid phase is complete, macrosegregation reaches its maximum. In this case, the Scheil equation relates the solute mole fraction in the solid (x_s) to the initial solute mole fraction (x_0) and the solid fraction (f_s) [14]:

$$x_s = kx_0(1 - f_s)^{k-1}, \quad (1)$$

where k is the partition coefficient. According to Eq. (1), the composition distribution is independent of the overall volume. In the final stage of solidification, k gradually deviates from the constant value assumed initially. Therefore, in the Scheil model, solidification process is considered complete when $f_s = 0.99$ [15]. More accurate values of x_s can be obtained simulation.

The beta-layering of target results in an extremely slow solidification process lasting about 5-14 hours [3]. The time is much longer than that required to reach maximum macrosegregation. Therefore, in the D-T simulation, the compositional distribution can be directly evaluated under the condition of maximum macrosegregation. Fig. 2(a) illustrates unidirectional solidification in a rectangular bar of D₂-T₂ mixture. Due to the extremely narrow liquidus-solidus gap of D₂-T₂ mixture, the partition coefficient k can be approximated as a constant over a small range. A 2D bar model with a length of 100 μm and a width of 10 μm was established (Fig. 2(b)), with the initial mole ratio of D₂ to T₂ set to 1:1. Fig. 2(c) shows that the simulated mole fraction distributions of D₂ is in good agreement with the theoretical predictions, demonstrating the validity of the numerical model.

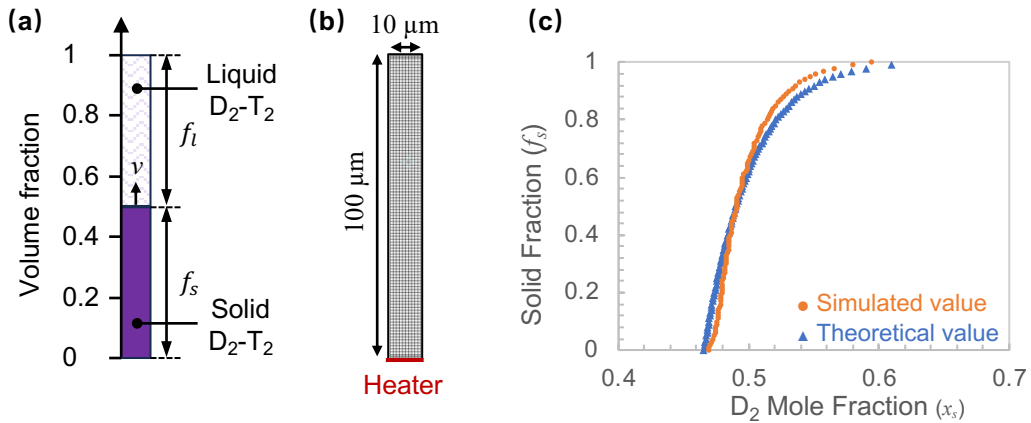


FIG. 2. (a) Schematic of unidirectional solidification process in rectangular D₂-T₂ bar. (b) Geometry and mesh of rectangular. (c) Comparison of simulated and theoretical values for the D₂ mole fraction distribution.

3.2. Compositional distribution during radial solidification

The solid-liquid interface advances in a parallel manner during unidirectional solidification. Mass transport occurs primarily along the same direction. The compositional distribution exhibits a linear concentration gradient, consistent with theoretical predictions. In cylindrical or spherical geometries, solidification proceeds radially from

the outside inward. The solid-liquid interface remains concentric and oriented toward the geometric center, consequently altering the direction and spatial distribution of solute transport.

To investigate the impact of geometry variation on macrosegregation, radial solidification of D₂-T₂ annuli with a fixed thickness of 100 μm and varying inner-to-outer radius ratios (r/R) was simulated, as shown in Fig. 3(a). An annular sector was extracted as a representative unit for analysis. Fig. 3(b) shows that when the r/R approaches 1, the annular sector approximates a rectangular shape. As r/R reduces to zero, the annular sector becomes a circular sector. This geometric variation further extends to 3D spherical shells. When the r/R equals zero, the spherical shell degenerates into a solid sphere, and the analysis unit becomes a spherical pyramid.

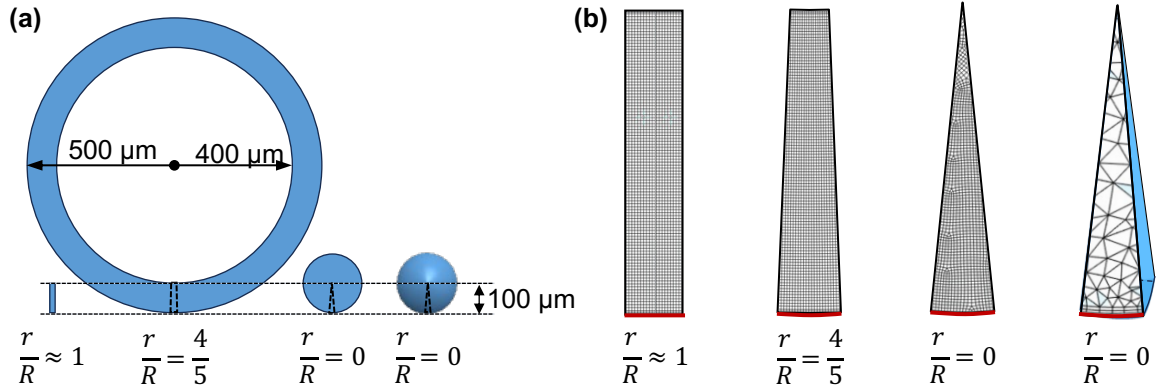


FIG. 3. (a) Geometric models of radial solidification. (b) Meshes of the models.

The post-solidification distribution of D₂ mole fraction is shown in Fig. 4(a). When r/R approaches 1, the direction of mass diffusion at the solid-liquid interface is almost perpendicular. As r/R reduces to zero, diffusion converges toward the geometric center during the final stage of solidification. In 3D solid sphere ($r/R = 0$), the liquid phase space undergoes rapid contraction. In radial solidification, as the solid-liquid interface advances, the available space for liquid-phase diffusion is more rapidly compressed. Solute rejected into the liquid phase becomes increasingly enriched, thereby intensifying macrosegregation in the final solid. Fig. 4(b) demonstrates that smaller values of r/R lead to more severe macrosegregation. Conversely, a higher r/R enhances the compositional homogeneity of the solid D-T layer.

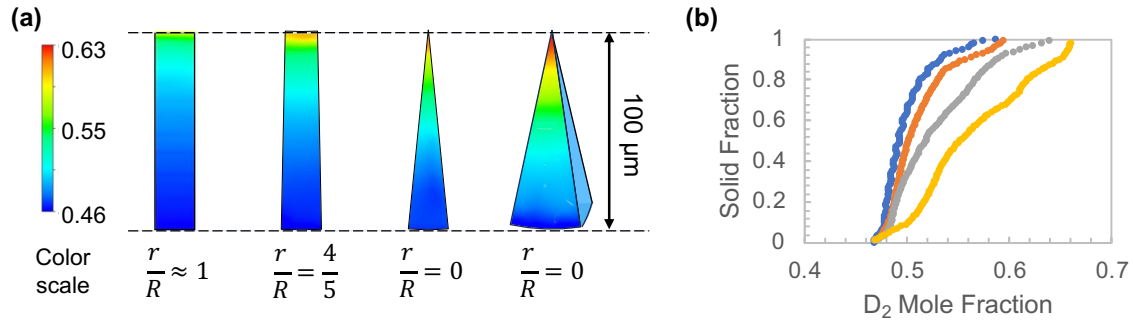


FIG. 4. (a) D₂ mole fraction distribution. (b) Radial distribution of mole fraction from the outer surface inward.

3.3. Compositional distribution in D-T target

Solidification in target proceeds radially toward the center. A 3D spherical model was used to accurately capture the influence of layer thickness on composition distribution. The target model was represented as a double layer concentric sphere. The inner layer consisted of D-T gas with a radius of 0.4 mm, and the outer layer consisted of liquid D-T with a radius of 0.5 mm [16], as shown in Fig. 5(a). The model was discretized using an unstructured

mesh into 429,277 tetrahedral elements, with an average mesh size of about $20\ \mu\text{m}$ (Fig. 5(b)). To resolve steep gradients near the component boundaries, the inflation tool was applied to improve mesh quality in these regions.

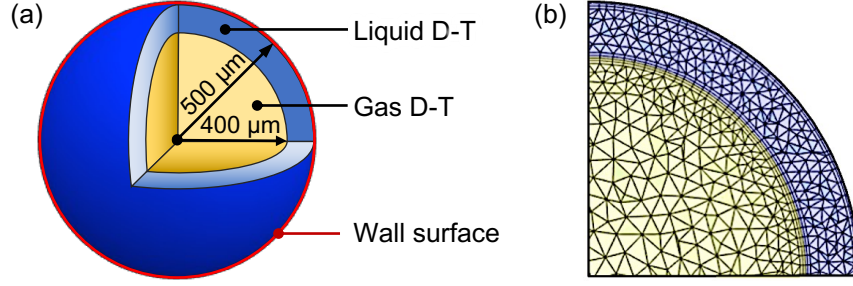


FIG. 5. (a) Geometry of 3D fuel sphere (b) Mesh of 3D fuel sphere.

3.3.1. Homogeneity of solid D-T fuel layer

The half-time of the isotope exchange reaction has been estimated to be approximately 18 hours in an equimolar mixture of D_2 and T_2 liquid at 21 K [17], which is longer than the 5–14 h required for β -layering. Isotopic exchange may therefore be incomplete in the solid D-T layer. The actual D_2 : DT: T_2 composition could range from 5:0:5 (no exchange) to 3:4:3 (complete equilibrium) [18]. To evaluate the effect of isotopic exchange on macrosegregation, simulations were performed for a D_2 : $\text{T}_2 = 1:1$ mixture and D_2 : DT: $\text{T}_2 = 3:4:3$ mixture.

A fixed time-stepping method was employed with a time step size of 0.01 s and a maximum of 20 iterations per step. The wall temperature was gradually reduced by 0.05 K every 20 steps. During phase transition, the liquid-solid interface remained constant at the freezing temperature. A radial temperature gradient was established from the cooled wall to the liquid-solid interface, driving the inward advance of solidification. Solidification was essentially complete once the wall temperature decreased from about 19.55 K to 19.2 K. Fig. 6(a) and (b) present the spatial compositional distribution in D_2 - T_2 and D_2 -DT- T_2 layers. T_2 solidified preferentially and accumulated rapidly at the wall boundary; D_2 increased in the subsequently solidified regions, culminating in a radial concentration gradient.

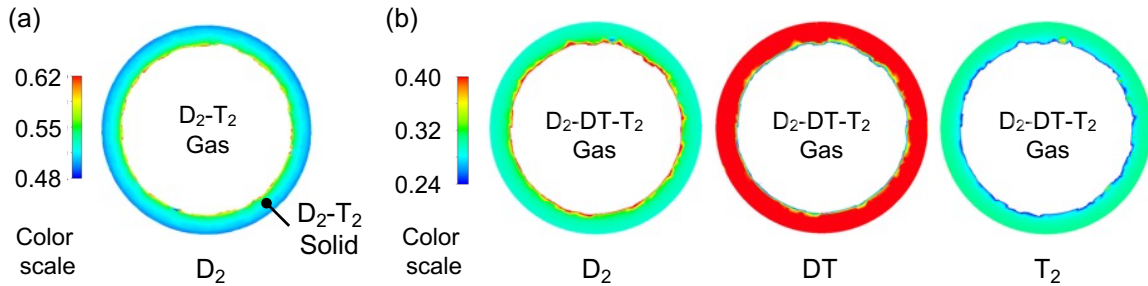


FIG. 6. Compositional distribution D-T layer in cross-section (a) D_2 mole fraction in solid D_2 - T_2 layer (b) mole fraction of each molecule in solid D_2 -DT- T_2 layer

In the binary D_2 - T_2 layer, the mole fraction of D_2 (or D atom) radially distributed inward from the outer surface of the solid D-T layer ranged from 0.480 to 0.600, while in the ternary D_2 -DT- T_2 layer, the mole fractions across the solid layer varied from 0.293 to 0.397 for D_2 , 0.400 to 0.348 for DT, and 0.307 to 0.254 for T_2 , respectively. Correspondingly, the radial mole fraction of D atom ranged from 0.493 to 0.571. Fig. 7 compares the radial distribution of D atoms for the D_2 - T_2 and D_2 -DT- T_2 cases. The actual distribution of D atoms is expected to lie between these two bounds.

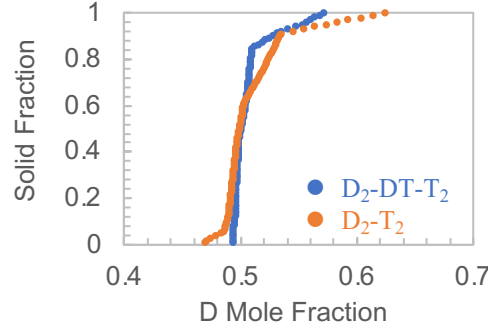


FIG. 7. Radial profile of the D atom mole fraction from the outer surface of the solid D-T layer inward.

3.3.2. Composition of the central D-T gas

The central D-T gas is in equilibrium with the surrounding solid D-T layer [19], sustaining saturated vapor pressure. Although gas was not involved in mass transfer and phase change in the simulation, isotopologue fractionation in practice causes its composition to deviate from the initial 1:1 ratio. According to Raoult's law, the partial pressure of each component in an ideal mixture equals the vapor pressure of the pure component multiplied by its mole fraction in the solid phase:

$$P_i^G = P_i^S X_i^S \quad (2)$$

Where P_i^G is the partial pressure of the component i ($i = D_2, DT, \text{ or } T_2$) in the gas phase, P_i^S is the saturated vapor pressure of the pure i . The gas-solid interface remains the freezing temperature (19.650 K). The empirical formulas for P_i^S (Pa) as a function of temperature T (K) are provided in Ref. [20]. X_i^S is the mole fraction in the solid phase. Once equilibrium is reached, Raoult's law combined with Dalton's law yields the total vapor pressure. The mole fraction of each gas component can be expressed as:

$$X_i^G = \frac{P_i^G}{\sum_i P_i^G} \quad (3)$$

After solidification, the equilibrium mole fractions of gas components were evaluated based on the composition of the adjacent solid layer. The D_2 (or D atom) proportion in D_2-T_2 gas can reach 75.6% in the gas. The compositions of D_2-DT-T_2 gas were approximately 52.1% D_2 , 31.6% DT, and 16.3% T_2 , corresponding to a D atom proportion of 67.9%. The concentration of D atoms in the gas is substantially higher than the initial 50%. If isotope exchange is incomplete, the D content will increase further.

3.3.3. Negative effects of inhomogeneity in D-T target

Inhomogeneity in the solid D-T fuel layer can introduce several adverse effects during ICF process:

(1) Rayleigh-Taylor instability. The implosion extremely compresses the D-T fuel, generating radial inward acceleration. Even solid D-T fuel exhibits a fluidic state. Rayleigh-Taylor instability develops at the interface between fluids of different densities. The simulation results show that a radial density gradient forms within the solid D-T layer. In addition, microsegregation and dendritic structures may generate additional irregularities along isodensity surfaces, as shown in Fig. 8(a). These inhomogeneities act as seeds for Rayleigh-Taylor instability, allowing initially minor perturbations to grow exponentially, threatening the implosion uniformity [21].

(2) Deviation of D/T ratio. Since the reaction rates of D-D and T-T fusion are several orders of magnitude lower than that of D-T fusion, deviations from the optimal 1:1 D/T ratio can impair fusion reaction efficiency. In ignition phase, hot-spot forms in central D-T gas [22]. Simulations indicate that isotopic fractionation leads to an excess of D atoms in gas. Fig. 8(b) shows that the hot-spot forms in D-T gas with a distorted D/T ratio, reducing the theoretical upper limit of the fusion reaction efficiency. Excess D atoms that do not participate in the reaction as impurities, potentially further interfering ignition. Incomplete isotope exchange amplifies this effect. Increasing the T_2 content in the injected fuel could raise the T concentration in the D-T gas and partially compensate for this imbalance [23].

The energy release fusion reactions in the hot spot region ignites the surrounding solid D-T main fuel, triggering a chain fusion reaction [24]. Similarly, fusion reactions are difficult to occur in regions enriched with D or T. In solid D-T layer with radial concentration gradient, deviations from the 1:1 D/T ratio can hinder the propagation of the burn wave, as illustrated in Fig. 8(c). The present fractionation model is based on the liquid-to-solid phase transition. In practical β -layering, phase transitions involving direct gas-to-solid deposition may also occur, which could potentially increase the fractionation level.

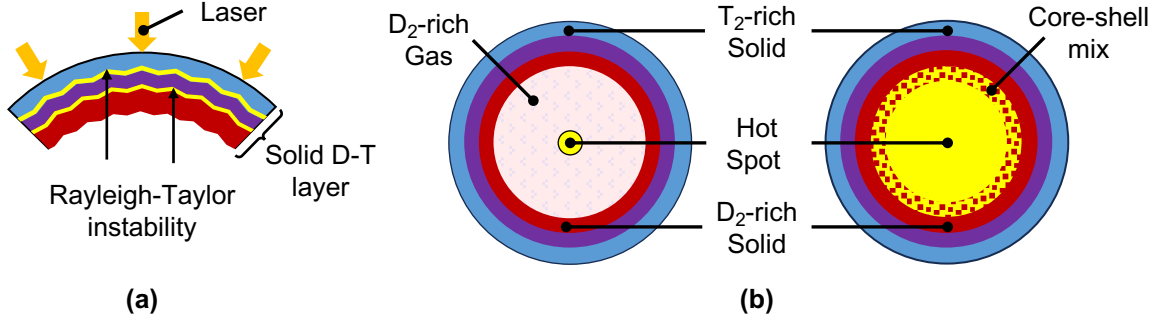


FIG. 8. Detrimental effects of composition inhomogeneity on ICF performance. (a) Rayleigh-Taylor instability. (b) Deviation of D/T ratio

4. CONCLUSIONS

This study conducted a numerical investigation of compositional inhomogeneity arising from isotopologue fractionation in the solid D-T fuel layer of cryogenic ICF targets. A comprehensive physical framework coupling heat transfer, species transport and phase transition, was employed to simulate the dynamic compositional distributions during solidification in D-T mixture. The current limitations of the model are its inability to account for gravitational effects and to capture complex phase-change processes induced by decay heat.

Simulations quantified the maximum macrosegregation occurring in D₂-T₂ mixtures during unidirectional solidification; the influence of the r/R ratio on macrosegregation in radial solidification; and the composition distribution in D-T target during solidification. The results demonstrate that a radial compositional gradient produced in the solid fuel layer, as well as a significant deviation of the central gas D/T ratio from the initial 1:1 value. Such compositional inhomogeneity is expected to degrade ICF performance: nonuniform density profiles may seed Rayleigh–Taylor instability growth during implosion, while deviation from the optimal 1:1 D/T ratio reduce the theoretical fusion yield and hinder both hot-spot ignition and burn propagation.

This numerical approach overcomes the limitations of experimental characterization by enabling direct evaluation and prediction of the compositional distributions in D-T targets of varying sizes. The findings indicate that neglecting fuel inhomogeneity may lead to overestimation of target performance. Achieving compositional uniformity should therefore be considered a critical design criterion for future ICF targets. This study advances understanding of the isotopologue fractionation mechanisms, and provides a theoretical basis and practical guidance for predicting and improving compositional homogeneity in target design.

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