Postprint of Nucleation Model Development and Numerical Simulation of Dendritic Growth During Solidification of Al-7Si-Mg Alloy

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Abstract

Aiming at the characteristics of lower cooling rates in aluminum alloy sand casting, a nucleation function was established through experimental measurement and analysis of cooling curves under different solidification conditions, which describes the exponential variation of nucleation density with maximum nucleation undercooling in aluminum alloys. By coupling with the thermodynamic, kinetic, and equilibrium phase diagram databases of Pandat software and employing algorithms such as spatial coordinate transformation, a cellular automaton (CA) model applicable to two-dimensional and three-dimensional dendritic growth in ternary aluminum alloys was developed. In this model, important factors including solute diffusion, constitutional undercooling, curvature undercooling, crystallographic orientation, and interactions between different components were simultaneously considered. Utilizing the established nucleation and growth models, the two-dimensional dendritic evolution and morphological characteristics of Al-7Si-0.36Mg alloy under various solidification conditions were simulated, the distribution characteristics of solute elements were described, and the variation of secondary dendrite arm spacing was quantitatively predicted, with results compared against experimental data. The three-dimensional dendritic simulation results effectively reflected the complexity and diversity of spatial structures and showed good agreement with experimental results.

Full Text

Nucleation Model and Dendrite Growth Simulation in Solidification Process of Al-7Si-Mg Alloy

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Abstract

Due to the extensive applications of Al-7Si-Mg casting alloys in automotive and aerospace industries, understanding dendritic microstructural formation is crucial for controlling desirable microstructures and thereby modifying casting performance. In this work, through analyzing measured cooling curves under different solidification conditions during sand casting of Al-7Si-0.36Mg ternary alloy, a theoretical nucleation model correlating maximum nucleation undercooling with nucleation density is proposed. Additionally, a 2D and 3D cellular automaton (CA) model is presented for quantitatively predicting dendrite growth in ternary alloys. This model introduces a new neighboring tracking rule algorithm to eliminate mesh dependency effects on dendrite growth. The thermodynamic and kinetic data required for simulations is obtained by coupling with the Pandat software package in combination with thermodynamic/kinetic/equilibrium phase diagram calculation databases. The model accounts for multi-component diffusion, constitutional undercooling, curvature undercooling, dendrite preferential growth angles, and interactions between alloying elements. The model is applied to quantitatively simulate dendrite growth with various crystallographic orientations of Al-7Si-0.36Mg ternary alloy in 2D and 3D during polycrystalline solidification. The predicted secondary dendrite arm spacing (SDAS) shows reasonable agreement with experimental results. The complicated and diverse dendritic microstructures observed experimentally during solidification can be well reproduced by this 3D-CA model, which considers the effects of various preferred growth orientations, interactions between adjacent dendrites, and solid/liquid interface anisotropies. The simulated results effectively demonstrate the model's capability for predicting dendritic microstructure in ternary alloys.

Key words: ternary aluminum alloy, nucleation model, cellular automaton, dendrite growth, secondary dendrite arm spacing

Introduction

Aluminum alloys are widely used as functional structural materials in aerospace, automotive, appliance, food, and other industries due to their advantages of light weight, high strength, corrosion resistance, and easy formability [1]. Al-7Si-Mg alloys (mass fraction, %, hereinafter the same) such as ZL114, A356, and A357 represent typical Al-Si casting alloys. Their high Si content provides excellent fluidity for casting large, thin-walled complex components, while the addition of trace Mg (0.3%~0.7%) enables age-hardening through formation of Mg_2Si precipitates [2], making them typical materials for critical automotive components

like engine blocks and train gearboxes. With continuous development of the automotive industry, higher performance requirements for Al-7Si-Mg alloy castings have drawn persistent attention to the solidification behavior, microstructural evolution, and properties of this alloy under various processes [3-6].

Casting mechanical properties are primarily determined by solidification microstructure, which in turn depends on alloy composition and casting process parameters. To enhance strengthening effects, practical alloys generally contain multiple alloying elements, making it theoretically and engineeringly significant to investigate how different solidification conditions affect the solidification microstructure of multi-component alloys. For Al-7Si-Mg alloys, the primary phase solidification temperature range is relatively narrow, leaving insufficient time for adequate dendrite arm growth before eutectic formation. Consequently, dendrite arm structures in this alloy are generally well-developed. Primary and secondary dendrite arm spacings, particularly the latter, are important parameters characterizing solidification microstructure features, directly influencing the distribution of composition, second phases, and shrinkage porosity, thereby affecting casting service performance [7]. In recent years, the relationship between secondary dendrite arm spacing and casting mechanical properties has attracted widespread attention [1,7-10].

With rapid development of computer technology, numerical simulation has become an effective approach for predicting solidification microstructural evolution. It is now possible to quantitatively simulate microstructural evolution under different solidification conditions and predict local mechanical properties. Current microstructural simulation methods mainly include the cellular automaton (CA) method and phase field (PF) method. The CA method offers high computational efficiency and can establish complex mathematical equations describing solid/liquid interface morphology and solute distribution by coupling heat diffusion, solute diffusion, and dendrite growth kinetics, thereby simulating dendritic morphology evolution at larger scales closer to practical engineering applications. Consequently, it has developed rapidly and been widely applied in recent years [11-14]. Li et al. [11] established an instantaneous nucleation model and corresponding CA method for ZL114A aluminum alloy in low-pressure casting based on extensive experimental statistical analysis, achieving good agreement with step casting experiments. Huo et al. [12] and Wu et al. [13] developed CA models for dendrite growth in magnesium alloys with hcp structures by defining different neighbor cell types and capture rules, simulating solidification microstructures under different cooling rates and dendrite morphologies under different die-casting processes, with results matching experimental observations. Shi et al. [14] developed an improved CA model based on directional solidification experiments of transparent NH_4Cl-H_2O alloy, simulating and analyzing the effects of temperature gradient, dendrite preferential orientation, and pulling velocity on columnar grain growth morphology. However, most practical alloys are ternary or multi-component, while the above models are mainly limited to binary alloy dendrite simulation. Therefore, establishing multi-component alloy dendrite growth models is essential from both theoretical

analysis and engineering application perspectives. Zhu et al. [15] coupled a CA model with thermodynamic phase equilibrium calculation database (PanEngine) to establish a mathematical model describing multi-component aluminum alloy dendrite growth and microsegregation, comparing results with Scheil model predictions. However, the kinetic growth coefficient introduced in this model is an empirical parameter whose value affects calculation results. References [16-18] primarily solved interface growth velocity based on local interface equilibrium to calculate solid fraction changes when simulating multi-component alloy dendrite growth. This method requires solving the interface normal direction, which is complicated, particularly for three-dimensional dendrite calculations, and generally requires iterative solutions. Therefore, existing CA models for multi-component alloy dendrite growth need further improvement and refinement. Actual solidification involves complex three-dimensional multi-dendrite spatial structures, making it a major challenge to establish CA models describing three-dimensional multi-dendrite growth processes that yield results more consistent with actual solidification microstructures. Additionally, combining simulation research with industrial production practices for actual alloy systems to enhance model practicality represents a future development direction [19].

Nucleation is the first step in solidification, determining phase precipitation sequence and distribution, and is a prerequisite for accurately predicting solidification microstructure features. Therefore, establishing an appropriate nucleation model is particularly important. Previous solidification microstructure simulations mostly employed continuous nucleation models based on Gaussian distribution [20], Oldfield's [21] continuous nucleation model for simulating chilled gray iron, or Stefanescu's [22] instantaneous nucleation model. The grain density $n(\Delta T)$ in the Gaussian continuous nucleation model can be expressed as a function of undercooling ΔT, but determining grain density or final nucleation number requires measuring three parameters: average nucleation undercooling ΔT , undercooling standard deviation ΔT , and maximum nucleation site density n . These parameters are difficult to measure, requiring fitting from nucleation density data at different undercoolings. Moreover, nucleation density calculations are sensitive to deviations in these three parameters, often resulting in significant discrepancies between calculated final nucleation numbers and reality within small dendrite calculation regions (mm^2 scale). Oldfield's nucleation model, based on experimental statistics, provides relatively accurate predictions, but its nucleation parameters lack clear physical meaning.

This work first establishes a nucleation model suitable for Al-7Si-0.36Mg alloy solidification under low cooling rate conditions (sand casting) based on experimental data statistics, and couples it with a CA dendrite growth model to make the simulation more consistent with solidification physics. By coupling with Pandat software thermodynamic, kinetic, and equilibrium phase diagram databases, and using spatial coordinate transformation algorithms, 2D and 3D CA models suitable for simulating ternary aluminum alloy dendrite growth are established. The models simultaneously consider important factors including solute diffusion, constitutional undercooling, curvature undercooling, crystal preferential

orientation, and interactions between different components. Using the established nucleation and growth models, 2D dendrite evolution and morphology characteristics of Al-7Si-0.36Mg alloy under different solidification conditions are simulated, solute component distribution features are described, secondary dendrite arm spacing changes are quantitatively predicted, and results are compared with experimental data. Three-dimensional dendrite simulation results effectively reflect the complexity and diversity of dendritic spatial structures, showing good agreement with experimental results.

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1. Nucleation Model Establishment

Oldfield [21] established a function for predicting eutectic cell or graphite spheroid nucleation density during solidification of cast iron and nodular iron based on experimental statistics. This model expresses nucleation density N as a function of maximum nucleation undercooling ΔT, where y and k are nucleation constants obtained through experimental statistics, and $\Delta T = T_q - T$ (T_q is the equilibrium liquidus temperature, T is the minimum nucleation temperature corresponding to the lowest point on the cooling curve). Due to its experimental data fitting basis, simple calculation, relatively accurate results, and strong engineering applicability, this model is widely used in various simulation software, though its parameters lack clear physical meaning. This work borrows from this model's concept to establish a nucleation model suitable for equiaxed grain solidification in aluminum alloys, where model parameters have clear physical meaning and can be conveniently coupled with dendrite growth models.

To establish a nucleation model for solidification of Al-Ti-B refined aluminum alloy melts, the complex nucleation essence is simplified through the following assumptions: (1) Nucleus dissolution and coalescence are neglected, meaning the original nucleation number equals the final grain number, while ignoring fading effects of refiners; (2) According to references [23,24], the initial nucleation undercooling of α-Al in Al-Ti-B inoculated aluminum alloy melts is very small, generally less than 0.5 K and can even reach 0.01 K, therefore nucleation can be considered to begin once temperature drops below the liquidus temperature and ends when temperature reaches T ; (3) TiB_2 morphology observed in experiments shows hexagonal platelets, and TiB_2 is assumed to be the only nucleation substrate [25,26], with its {0001} basal plane shown in [Figure 1: see original paper].

Many researchers [25,26] have conducted experimental studies on α-Al nucleation processes in Al-Ti-B inoculated aluminum alloy melts, demonstrating that α-Al nucleates exclusively on TiB_2 {0001} basal planes. Initially, the nucleus grows along the TiB_2 substrate until covering the entire {0001} basal plane, after which growth proceeds primarily by reducing the solid/liquid interface curvature radius r , meaning the height h in [Figure 1: see original paper]b con-

tinuously increases. As h increases, r decreases, but according to free growth theory [25], r cannot be smaller than the critical nucleus radius r* at a fixed undercooling ΔT, i.e., r ≥ r. *The critical radius r* relates to ΔT as:

$$r^* = \frac{2\gamma}{\Delta S_V \Delta T}$$

where γ represents solid/liquid interfacial energy and ΔS is the volumetric entropy of fusion. Equation (2) shows that α-Al can only grow further by increasing ΔT to reduce r*. When α-Al reaches a hemispherical shape on the TiB_2 substrate as shown in [Figure 1: see original paper]b, r reaches its minimum value of r = 0.5d_N (where d_N is the nucleation substrate diameter shown in [Figure 1: see original paper]b), after which the crystal can grow freely. According to assumption (2), during the nucleation stage when temperature drops to T , nucleation undercooling reaches its maximum ΔT , corresponding to the minimum substrate diameter capable of nucleation, expressed as:

$$d^* = \frac{4\gamma}{\Delta S_V \Delta T_m}$$

From equation (3), if the size distribution of TiB_2 nucleation substrates is known, the functional relationship between nucleation density and ΔT can be established.

Greer et al. [25] performed extensive statistical analysis of TiB_2 particle size distributions in Al-5Ti-1B (mass fraction, %) inoculants using scanning electron microscopy (SEM) image analysis, fitting the data with an exponential function to obtain the following distribution function:

$$N(d_N) = \frac{N_0}{d_0} \exp\left(-\frac{d_N}{d_0}\right)$$

where d_0 represents the characteristic diameter of the size distribution, N_0 is the total number of TiB_2 particles per unit melt volume, and N(d_N) is the density of TiB_2 particles with disc diameters between d_N and d_N + dd_N. The relationship between N(d_N) and d_N is shown in [Figure 2: see original paper]a. The area under curve N(d_N) between particle diameters d_N, to d_N,∞ (where d_N, and d_N,∞ are minimum and maximum nucleation substrate diameters, respectively) represents the total number of nucleation substrates per unit melt volume N_0, while the gray area between d* and d_N,∞ represents the number of effective substrates capable of nucleation. According to assumption (2), when undercooling in the melt reaches its maximum ΔT , nucleation ceases as shown in [Figure 2: see original paper]b. At this point, TiB_2 particles with size d_N ≥ d* can all meet nucleation conditions, while particles with size d_N < d* cannot achieve the required undercooling. Therefore, only a small fraction of TiB_2 particles in the melt can serve as effective nucleation substrates. Under

fixed inoculation conditions, reducing T to increase ΔT increases the number of effective substrates capable of nucleation, enhancing alloy refinement.

Through integration, the number of effective nucleation substrates can be obtained, yielding the nucleation number per unit melt volume N , calculated as:

$$N_V = \int_{d^*}^{\infty} N(d_N)\, dd_N$$

Combining equations (3)-(5), N can be expressed as:

$$N_V = N_0 \exp\left(-\frac{4\gamma}{\Delta S_V d_0 \Delta T_m}\right) = N_0 \exp\left(-\frac{a}{\Delta T_m}\right)$$

As seen in equation (6), both parameters N_0 and a have clear physical meaning, with parameter a determined by γ, ΔS , and d_0. Due to lack of data for N_0, γ, ΔS , and d_0, specific parameter values in equation (6) cannot be obtained. Therefore, this work primarily uses experimental data for N and ΔT to determine N_0 and a through data fitting.

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2. Dendrite Growth Process Description

The degree of microstructural refinement in aluminum alloy solidification manifests in two aspects: grain refinement and secondary dendrite arm spacing refinement. The former is mainly affected by nucleation conditions, while the latter depends on the growth process. Therefore, based on the nucleation model, this work employs a dendrite growth CA model to describe dendrite growth under different solidification conditions and predict final dendrite morphology. The model describes dendrite growth based on the following reasonable assumptions: (1) Since the microscale calculation domain is very small (mm scale), solute diffusion can be considered to satisfy solute conservation within the microscale domain using closed boundary conditions; the temperature field is assumed uniform throughout the calculation domain with temperature values obtained from actual cooling curves; (2) Due to small density differences between Al and Si elements, natural convection has minimal effect on dendrite growth, so liquid flow effects are neglected; (3) Kinetic undercooling is ignored, making the model suitable only for simulating dendrite growth under low Péclet number conditions.

2.1 Multi-component Solute Diffusion Equation Microscale CA cells have three states: liquid, solid, and solid/liquid interface. As solid fraction increases in interface cells, each solute element is rejected, creating solute gradients ahead of the interface that drive solute diffusion. Therefore, solute diffusion equations must be solved separately for each element:

$$\frac{\partial w_i^\phi}{\partial t} = \nabla \cdot \left(\sum_j D_{ij}^\phi \nabla w_j^\phi \right) + \frac{\partial f_S}{\partial t}(w_i^L - w_i^S)$$

where w represents the solute content of component i (Si or Mg) in phase ϕ (liquid or solid), D is the diffusion matrix in phase ϕ, f is the solid fraction, t is time, and k is the equilibrium solute partition coefficient, which is no longer constant in multi-component cases but depends on composition and temperature, requiring coupling with equilibrium phase diagram databases.

2.2 Growth Kinetics and Solid Fraction Increment Calculation For interface cells, the combined effects of constitutional undercooling and interface curvature enable local thermodynamic equilibrium at the interface, so the interface equilibrium temperature can be expressed as:

$$T(t) = T_{liq}(w_{Si}^L, w_{Mg}^L) - \Delta T_C - \Delta T_R$$

where T q(w , w) is the equilibrium liquidus temperature read from the phase diagram based on Si and Mg composition values w in the liquid phase. ΔT_C and ΔT_R correspond to constitutional undercooling and curvature undercooling, respectively, calculated as:

$$\Delta T_C = \sum_{i=Si,Mg} m_{Li}(w_i^L - w_{0i})$$

$$\Delta T_R = \frac{2\Gamma}{R} g(\mathbf{n})$$

where m is the liquidus slope when only component i is present, w_0 is the initial composition of component i, w is the local equilibrium solute composition at the interface, is the local interface mean curvature (calculation methods can be found in references [27,28]), and g(n) is the anisotropy function given by [16]:

$$g(\mathbf{n}) = 1 - 3\varepsilon \left[\cos^4\theta_1 + \sin^4\theta_1 \left(1 - 2\sin^2\theta_2 + \frac{4}{3}\sin^4\theta_2 \right) \right]$$

where Γ is the Gibbs-Thomson coefficient, is the interface energy anisotropy coefficient, $_1$ = arccos(n_z), and $_2$ = arccos(n_x / sin $_1$).

Interface cell solid fraction calculation mainly employs two methods: one solves interface growth velocity based on solute conservation during solid/liquid interface advancement to obtain solid fraction increment; the other considers local interface equilibrium, using the difference between interface equilibrium solute composition and actual interface solute composition as the driving force for dendrite growth, then directly obtains solid fraction increment using local lever

law, avoiding solution of dendrite growth velocity and improving computational efficiency. This model adopts the latter method. If an interface cell rejects portions of Si and Mg elements due to solid fraction increase during a time step, the following equations are satisfied:

$$w_{Si}^{L}(1-f_S-\Delta f_S)=w_{Si}^{L}(1-f_S)-\Delta f_S w_{Si}^{S}$$

$$w_{Mg}^{L}(1-f_S-\Delta f_S)=w_{Mg}^{L}(1-f_S)-\Delta f_S w_{Mg}^{S}$$

where Δf is the solid fraction increment of the interface cell during each time step. Combining equations (9)-(14) uniquely solves for w , w , and Δf .

2.3 Arbitrary Angle Dendrite Growth Description During solidification, dendrite preferential growth directions are random, as shown in [Figure 3: see original paper]a. However, CA grid anisotropy causes dendrite growth directions to deviate from their inherent preferential growth directions. To eliminate inherent effects of grid anisotropy on dendrite growth, this work employs spatial coordinate transformation and eccentric algorithms to simulate arbitrary angle dendrite growth.

As shown in [Figure 3: see original paper]a, each dendrite grain is assigned its own local coordinate system (x_0, y_0, z_0) with axes aligned along respective [001] directions, while the world coordinate system (x, y, z) aligns with grid division directions. The positional relationship between the two coordinate systems can be realized through spatial coordinate rotation matrices Rotate_1, Rotate_2, Rotate_3:

$$\mathrm{Rotate}_1=\begin{pmatrix}\cos\alpha & \sin\alpha & 0\\ -\sin\alpha & \cos\alpha & 0\\ 0 & 0 & 1\end{pmatrix},\quad \mathrm{Rotate}_2=\begin{pmatrix}\cos\beta & 0 & -\sin\beta\\ 0 & 1 & 0\\ \sin\beta & 0 & \cos\beta\end{pmatrix},\quad \mathrm{Rotate}_3=\begin{pmatrix}1 & 0 & 0\\ 0 & \cos\gamma & \sin\gamma\\ 0 & -\sin\gamma & \cos\gamma\end{pmatrix}$$

$$\mathrm{Rotate}=\mathrm{Rotate}_1\cdot\mathrm{Rotate}_2\cdot\mathrm{Rotate}_3$$

where α, β, γ represent Euler angles between the two coordinate systems, as shown in [Figure 3: see original paper]b. Under 2D conditions with $\alpha=0$ and $\beta=0$, the coordinate transformation simplifies to:

$$\begin{pmatrix}x\\ y\end{pmatrix}=\begin{pmatrix}\cos\gamma & -\sin\gamma\\ \sin\gamma & \cos\gamma\end{pmatrix}\begin{pmatrix}x_0\\ y_0\end{pmatrix}$$

Capture rules employ a 3D eccentric regular octahedron algorithm or 2D eccentric square algorithm [27]. The basic principle states: if a grain with preferential

growth direction (α, β, γ) nucleates, the distance L(t) from the eccentric regular octahedron vertex to its center is calculated based on its f :

$$L(t) = \frac{\Delta f_S \Delta x}{\sqrt{2}} + \sqrt{2}\Delta x$$

where Δx is the grid size. After obtaining L(t), the coordinates of the six vertices of the eccentric regular octahedron in the local coordinate system (x_0, y_0, z_0) are determined, then transformed to world coordinates (x, y, z) using equation (16). When an octahedron vertex touches a neighboring liquid cell at any moment, that liquid cell is captured as an interface cell and assigned the preferential growth angle values (α, β, γ). During subsequent growth, this cell calculates its own regular octahedron vertex coordinates based on its f to capture surrounding liquid cells.

3.1 Experimental Method

This work used a cast aluminum alloy with main composition Al-7Si-0.36Mg (mass fraction, %). The melting temperature was 720-740 °C, with Ar degassing, Al-5Ti-1B refinement, and Al-10Sr modification treatments. To obtain different solidification conditions, the treated melt was poured into a quartz sand mold cavity with a step shape. To record solidification processes at different steps, K-type thermocouples were placed at the center of each step and connected to temperature recorders to capture cooling curves. The step casting geometry and thermocouple positions are shown in [Figure 4: see original paper], where solid dots indicate thermocouple locations labeled No.1-No.7 according to step thickness from thick to thin.

To analyze microstructural morphology and grain size under different solidification conditions, circular cross-section samples with 8 mm diameter were sectioned at each thermocouple position (6 mm diameter at No.7). Standard sample preparation procedures were used to obtain metallographic specimens. Anodizing and polarized light metallography were employed to distinguish different grains. The anodizing solution consisted of 38 mL H_2SO_4 + 43 mL H_3PO_4 + 19 mL H_2O, applied at 15 V with current controlled at 0.05-0.10 A for 40-60 s. Anodized specimens were observed using an Axio Scope.A1 optical microscope (OM) with compensator. The area method was used to determine surface grain density N :

$$N_S = \frac{N_1 + 0.5N_2}{A}$$

where N_1 is the number of complete grains within region A, and N_2 is the number of grains on region A's edges. Volume grain density N was then calculated using the Poisson-Voronoi model [29]:

$$N_V = 1.5\frac{N_S^{3/2}}{\sqrt{\pi}}$$

Secondary dendrite arm spacing was measured using the intercept method.

3.2 Experimental Results and Analysis

[Figure 5: see original paper] shows cooling curves at different thermocouple positions (No.1-No.7). The alloy solidification process mainly comprises two stages: first, α-Al nucleation and growth, with a noticeable temperature recalescence after reaching T due to latent heat release; second, when temperature reaches the eutectic temperature, (Al+Si) eutectic reaction occurs, significantly slowing the cooling rate due to massive latent heat release. [Figure 5: see original paper] clearly shows that as step thickness decreases from 40 mm at No.1 to 4 mm at No.7, T decreases from 611.3 °C to 595 °C, enlarging the nucleation interval and creating more effective nuclei in the melt, thereby increasing nucleation density.

[Figure 6: see original paper] presents microstructure images at thermocouple positions No.1-No.7. Due to the complexity of three-dimensional dendritic structures, dendrites observed on 2D planes are not very regular but basically reflect the planar fourfold symmetry structure of α-Al dendrites. Additionally, under low cooling rate conditions, fewer nucleation events and non-uniform nucleation positions cause large variations in final dendrite sizes within the same sample, showing smaller dendrites where nucleation density is high and larger, unrestricted dendrites where nucleation density is low. As cooling rate increases, nucleation density increases and nucleation core distribution becomes more uniform, resulting in more consistent final dendrite sizes.

To quantitatively characterize solidification parameters and microstructural features at different thermocouple positions, summarizes minimum nucleation temperature T , maximum nucleation undercooling ΔT , eutectic nucleation temperature T , average cooling rate R_c, grain density, and secondary dendrite arm spacing. As step thickness decreases, heat dissipation intensifies, grain density N increases, and secondary dendrite arm spacing becomes finer.

To determine parameters N_0 and a in equation (6), curve fitting was performed using ΔT and N data from . Taking logarithms of both sides of equation (6) simplifies it to a linear function of ln(N) versus ΔT^{-1}:

$$\ln(N_V) = \ln(N_0) - a \cdot \Delta T_m^{-1}$$

[Figure 7: see original paper] shows the functional relationship between ln(N) and ΔT^{-1}. Data points No.1-No.6 show a basically linear relationship between

ln(N) and ΔT^{-1}, but No.7 shows large deviation. The main reasons are: position No.7 is very thin (4 mm), where heterogeneous nuclei introduced by mold walls significantly influence nucleation, violating assumption (3) that TiB_2 is the only nucleation substrate; additionally, heat dissipation is very strong, making accurate temperature measurement difficult (as seen in [Figure 5: see original paper]), potentially causing large errors in T in . Therefore, this work primarily uses data from No.1-No.6 for fitting ln(N) and ΔT^{-1}, yielding the functional relationship shown in [Figure 7: see original paper]. This gives $N_0 = e^{7 \cdot 776} = 2382.7$ cm^{-3} and a = 7.747 °C, resulting in the nucleation model:

$$N_V = 2382.7 \exp\left(-\frac{7.747}{\Delta T_m}\right)$$

A similar method yields the surface nucleation density function:

$$N_S = 260.1 \exp\left(-\frac{5.165}{\Delta T_m}\right)$$

Since data point No.7 was not considered, the obtained nucleation function is not applicable to solidification conditions at No.7 and requires further improvement in future work. Before using this model for nucleation density prediction, actual or simulated cooling curves of castings must first be obtained to measure maximum nucleation undercooling ΔT , which then yields nucleation density at that location.

4.1 Two-Dimensional Dendrite Growth Simulation

Using the established nucleation model and ternary dendrite growth CA algorithm, numerical simulations were performed for microstructures at thermocouple positions No.1-No.7. To demonstrate the nucleation model's applicability to this alloy's nucleation process, nucleation numbers at positions No.1-No.6 were calculated using equation (22), while measured values from were used at No.7. Required temperature values were obtained from measured cooling curves. Thermophysical parameters for Al-7Si-0.36Mg alloy used in simulations are listed in . The simulation employed 1800$\times$1800 $grids with 4 \mu m grid size, and dendrite preferential growth angles were randomly assigned. [Figure 8 : see original paper] shows dendrite evolution and solute field distribution at position No.6. During early solidifica$ 8a). $As temperature decreases, dendrite growth velocity \nu increases (\nu \propto \Delta T\hat{}\{2\}$), intensifying solute enrichment ahead of the interface and increasing constitutional undercooling, causing solid/liquid interface destabilization and prominent branching with well-developed secondary dendrite arms (FIGURE:8b). Moreover, as dendrites grow, neighboring dendrites hinder each other's growth, primarily manifested through solute field changes. When adjacent dendrites approach each other, rejected solute cannot be effectively

removed due to poor diffusion channels, forming highly concentrated liquid zones ahead of interfaces that slow dendrite growth, causing asymmetric dendrite growth (FIGURE:8b). During late solidification, remaining liquid regions become highly solute-enriched, suppressing dendrite tip growth, and further dendrite development proceeds mainly through dendrite arm growth, coarsening, and coalescence, with secondary dendrite arm spacing automatically adjusting to an appropriate range (FIGURE:8c).

[Figure 9: see original paper] shows final solidification microstructures simulated under different solidification conditions. As cooling rate increases, nucleation number increases, grain size decreases, and secondary dendrite arms become finer, matching experimental results in [Figure 6: see original paper]b, d, f, g. To quantitatively demonstrate simulation accuracy, [Figure 10: see original paper] compares predicted and experimental secondary dendrite arm spacing at different thermocouple positions. Simulation and experimental results show good agreement with errors generally within 4%. References [7,30] correlated secondary dendrite arm spacing with average cooling rate R_c through power functions $l_2 = aR\_c^{-}$, where a and b are coefficients. This work fitted simulated data using this approach, obtaining $l_2 = 42.8R\_c^{-0 \cdot 302}$, which closely matches the relationship $l_2 = 39.4R\_c^{-0 \cdot 317}$ fitted from extensive experimental results in reference [30]. These comparisons demonstrate the model's accuracy in simulating Al-7Si-0.36Mg alloy dendrite growth and confirm its capability for predicting secondary dendrite arm spacing, providing a prerequisite for subsequent property prediction.

4.2 Three-Dimensional Dendrite Growth Simulation

Since actual dendrite growth involves three-dimensional spatial structures, 2D planar dendrite simulations (FIGURE:9) cannot adequately represent the extremely complex dendrite morphologies observed experimentally (FIGURE:6). Therefore, 3D simulation of dendrite evolution is essential and represents a major development trend in dendrite simulation. Actual solidification involves multi-dendrite growth where neighboring dendrites hinder each other, influenced by nucleation position, nucleation density, and dendrite preferential growth orientation. Researchers [31] have combined X-ray tomography with phase field simulation to investigate 3D dendrite structures, explaining the complexity and diversity of observed 3D dendrite structures. This work uses the established CA model to simulate 3D dendrite growth processes and morphologies in Al-7Si-0.36Mg alloy. The simulation employed 400$×400×$400 grids with 5 m grid size (calculation domain size 2 mm × 2 mm × 2 mm). 3D dendrite growth simulation was performed for position No.7, where nucleation number in the calculation domain was obtained from measured nucleation density data in , each nucleus was randomly assigned a growth orientation, and temperature data was read from measured cooling curves. [Figure 11: see original paper] shows the evolution of 3D multi-equiaxed dendrite microstructure under solidification

conditions at position No.7. During early solidification, small dendrite size and weak mutual influence allow dendrites to grow freely in undercooled melt along preferential growth directions (FIGURE:11a). As temperature decreases and undercooling increases, dendrite growth accelerates, interface stability decreases, primary dendrite trunks develop secondary branches, and dendrites begin showing asymmetric growth due to neighboring dendrites or calculation domain boundaries (FIGURE:11b and c). During late solidification, dendrites fill the entire calculation domain, dendrite arms coarsen and coalesce, resulting in final microstructures with complex structures and diverse morphologies (FIGURE:11d).

[Figure 12: see original paper] compares 2D cross-section dendrite morphologies at the end of primary solidification with experimental results. Secondary dendrite arms appear as intermittent distributions, and 3D simulation results show greater similarity to experimental results than 2D simulations (FIGURE:9). Additionally, using the intercept method, secondary dendrite arm spacing at position No.7 was measured as 28.9 m experimentally and 24.9 m from simulation. These results fully demonstrate the feasibility of this model for 3D dendrite growth simulation.

Conclusions

(1) By analyzing cooling curves at different positions in step castings and based on reasonable nucleation assumptions, a nucleation model suitable for equiaxed grain solidification of Al-7Si-Mg alloys under conventional solidification conditions was established. In this model, nucleation density N varies exponentially with the inverse of maximum undercooling ΔT^{-1}, and model parameters have clear physical meaning.

(2) By coupling with thermodynamic, kinetic, and equilibrium phase diagram databases and using spatial coordinate transformation algorithms, 2D and 3D CA models suitable for ternary aluminum alloy dendrite growth were established. Using the established nucleation and growth models, 2D dendrite growth simulations were performed for Al-7Si-0.36Mg alloy, analyzing dendrite evolution and morphology characteristics under different solidification conditions. Predicted secondary dendrite arm spacing showed good agreement with experiments, demonstrating the model's applicability for simulating ternary aluminum alloy dendrite growth.

(3) Three-dimensional multi-dendrite growth of Al-7Si-0.36Mg alloy was simulated and compared with experimental results on 2D cross-sections, preliminarily analyzing the complexity and diversity of 3D dendrite spatial structures and demonstrating the model's feasibility for 3D dendrite growth simulation.

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