Cellular Automaton Simulation of Solidification Microstructure in Ductile Iron (Postprint)

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Abstract

This work establishes a multi-phase cellular automaton (MCA) model for simulating the microstructure evolution of ductile iron solidifying in a divorced eutectic manner. The model employs the local solute equilibrium method to calculate the growth kinetics of graphite and austenite, and considers the density ratio of graphite to iron in the graphite growth model. The model can simulate microstructural morphologies consistent with experimental observations. Using this model, the interaction and competitive growth mechanisms between graphite and austenite are simulated and analyzed, the influence of cooling rate on the size and size distribution of graphite nodules at the end of solidification is discussed, and the simulation results are compared with experimental results. The results indicate that: the precipitation of austenite promotes the growth of adjacent graphite; the growth of both austenite and graphite phases is controlled by carbon diffusion; when graphite is enveloped by austenite, its growth rate decreases. Furthermore, with increasing cooling rate, the solidification time shortens, the average radius of graphite nodules decreases, and the variation trend of graphite nodule size distribution under different cooling rate conditions agrees well with experimental results.

Full Text

Preamble

Simulation of Solidification Microstructure of Spheroidal Graphite Cast Iron Using a Cellular Automaton Method

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Abstract

A multi-phase cellular automaton (MCA) model was developed to simulate the microstructure evolution of spheroidal graphite (SG) cast iron during solidification via a divorced eutectic mechanism. The model employs a local solutal equilibrium approach to calculate the growth kinetics of both graphite and austenite, incorporating the density ratio between graphite and iron. The simulated microstructure morphologies show good agreement with experimental observations. The model was applied to analyze the interactive and competitive growth mechanisms between graphite and austenite, and to investigate the effect of cooling rate on the final size and size distribution of graphite nodules. The simulation results were compared with experimental data, revealing that austenite precipitation promotes the growth of neighboring graphite in the liquid phase, that growth of both phases is controlled by carbon diffusion, and that graphite growth slows significantly after being enveloped by austenite. Additionally, as the cooling rate increases, solidification time decreases and the average graphite nodule radius becomes smaller. The evolution of graphite nodule size distribution under different cooling rates matches well with experimental results.

Keywords: Spheroidal graphite cast iron, solidification, divorced eutectic, cellular automaton, microstructure modeling

Introduction

In spheroidal graphite cast iron, the graphite exists as spherical nodules that create minimal stress concentration. For a given graphite volume, spherical graphite has the smallest surface area, thereby minimizing its detrimental effect on the metallic matrix. Since its inception, spheroidal graphite cast iron has found widespread application in machinery, metallurgy, automotive, mining, and other industrial sectors due to its advantageous mechanical properties, castability, and cost-effectiveness. The macroscopic mechanical properties of castings are intimately related to the microstructures formed during solidification, including graphite nodule shape, size, size distribution, and matrix morphology. Industrially, hypereutectic spheroidal graphite cast iron is most commonly used. During solidification of hypereutectic compositions, primary graphite nucleates and grows in the liquid when the temperature drops below the graphite liquidus temperature. Austenite begins to nucleate at the eutectic temperature and grows dendritically in the liquid until contacting graphite nodules. Upon contact, graphite nodules become rapidly enveloped by austenite shells and continue to grow within the austenite. Because carbon diffusion in austenite is much slower than in liquid, graphite growth decelerates markedly after being surrounded. Simultaneously, the austenite shells continue to grow into the liquid until they impinge, completing the solidification process.

Computational simulation has become an effective method for investigating microstructure evolution during solidification, with the cellular automaton (CA) method emerging as a particularly active research area. Scholars worldwide have developed various two-dimensional and three-dimensional CA models to study dendritic growth, microsegregation, microporosity formation, and other solidification phenomena under diffusive and convective conditions. Charbon and Rappaz first applied the CA method to spheroidal graphite cast iron solidification, visualizing microstructure evolution based on carbon diffusion-controlled growth using classical models. Their simulations showed only one graphite nodule per eutectic grain, contradicting experimental observations. Roxana et al. subsequently incorporated primary austenite effects, assuming graphite growth only after austenite contact, and simulated hypoeutectic spheroidal graphite cast iron solidification, obtaining multiple graphite nodules per austenite grain. However, their model neglected graphite growth in the liquid and determined interface concentrations from equilibrium phase diagrams without coupling to the real-time concentration field.

Burbelko and colleagues applied continuous nucleation models using kinetic undercooling as the growth driving force to simulate austenite and graphite nucleation and growth in spheroidal graphite cast iron. Their results reproduced experimentally observed irregular circular graphite morphologies and cooling curves, but they did not investigate the influence of processing parameters such as cooling rate on solidification microstructure, nor did they consider the important density ratio between graphite and iron phases.

Our previous work developed a multi-phase CA model for nucleation and growth of spheroidal graphite and austenite dendrites during spheroidal graphite cast iron solidification, based on the local solutal equilibrium model proposed by Zhu and Stefanescu, with carbon diffusion as the growth driving force for both phases. However, because the model artificially averaged carbon composition at graphite interfaces, simulated graphite appeared as perfect spheres. Additionally, neglecting the graphite-to-iron density ratio resulted in simulated graphite volume fractions and average radii lower than experimental measurements.

Building upon this foundation, the present work proposes a multi-phase cellular automaton (MCA) model that calculates graphite growth kinetics using actual local carbon concentrations at graphite interfaces and incorporates the graphite-to-iron density ratio. This model simulates microstructure morphology and local concentration field evolution during hypereutectic spheroidal graphite cast iron solidification, analyzing the interactive and competitive growth mechanisms between graphite and austenite, and investigating the effect of cooling rate on graphite nodule size distribution and average radius.

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1. Model Description

1.1 Model Framework

This work focuses on the interaction and competitive growth mechanisms between spheroidal graphite and austenite phases, and the evolution of concentration fields during divorced eutectic solidification of hypereutectic spheroidal graphite cast iron. The two-dimensional computational domain is discretized into uniform square cells. In the three-phase system comprising liquid, austenite, and graphite, each cell is assigned variables including concentration, temperature, volume fractions of the three phases (liquid, graphite, and austenite), preferred orientation of austenite dendrites, graphite nodule identification numbers, and cell states: liquid ($fL = 1$), graphite ($fGr = 1$), austenite ($f\gamma = 1$), graphite/liquid interface ($0 < fGr < 1$, $f\gamma = 0$), graphite/austenite interface ($0 < fGr < 1$, $fL = 0$), austenite/liquid interface ($0 < f\gamma < 1$, $fGr = 0$), and graphite/austenite/liquid interface ($fGr > 0$, $f\gamma > 0$, $fGr + f\gamma < 1$), where fL, fGr, and fγ represent the phase fractions of liquid, graphite, and austenite, respectively.

At the beginning of simulation, all cells are initialized with uniform composition and temperature, starting from the liquidus temperature corresponding to the initial composition. For hypereutectic spheroidal graphite cast iron, when the domain temperature falls below the graphite liquidus temperature, graphite precipitates from the liquid and grows approximately spherically. At the eutectic temperature, austenite precipitates and grows together with graphite in a divorced eutectic manner. Nucleation densities for graphite and austenite are determined from literature nucleation models or experimental data. After nucleation, graphite and austenite positions remain fixed, neglecting flotation effects from fluid flow. During solidification, when graphite contacts austenite, the graphite becomes rapidly enveloped by an austenite shell. Subsequently, graphite continues to grow by absorbing carbon solute through the surrounding austenite shell until simulation ends at the eutectoid temperature. During austenite growth, carbon solute is continuously rejected into the surrounding liquid until austenite shells impinge, completing solidification. This model neglects the effect of fluid flow on solute transport; carbon transport occurs only by diffusion. The computational domain is assumed to have uniform temperature and cools at a constant rate. The cooling rate during solidification is influenced by both heat extraction rate and latent heat release from graphite and austenite growth.

The present model employs the local solutal equilibrium model to calculate graphite and austenite growth, with the growth driving force being a function of the difference between interface equilibrium composition and actual composition. Local interface equilibrium composition is calculated based on interface temperature and curvature, while actual composition is obtained from the concentration field. Additionally, the graphite-to-iron density ratio is considered in calculating growth of both phases.

1.2 Concentration and Temperature Field Calculations

Solute equilibrium is assumed at austenite/liquid, graphite/liquid, and graphite/austenite interfaces. On one hand, austenite rejects solute into the surrounding liquid during growth, and solute redistribution at the solid/liquid interface is calculated based on the partition coefficient kγ, where Cγ* and CL* are the compositions of austenite and liquid at the austenite/liquid interface, respectively.

On the other hand, graphite absorbs carbon solute from the surrounding liquid or austenite phase during growth. The combined effect creates concentration gradients ahead of the growth interface, driving carbon diffusion. The solute transport governing equation incorporating solute redistribution is:

$$\frac{\partial C_i}{\partial t} = \nabla \cdot (D_i \nabla C_i) + \text{source terms}$$

where t is time, Ci is composition, Di is the diffusion coefficient, and subscript i denotes austenite (γ) or liquid (L). Cint* represents the composition of austenite at the graphite/austenite interface, or the liquid composition at the graphite/liquid and graphite/austenite/liquid interfaces. CGr is the carbon content in graphite (CGr = 100%). The second term on the right-hand side represents solute redistribution generated at the austenite/liquid or austenite/graphite/liquid interface during austenite growth, implemented by adding the rejected solute to the remaining liquid in the same interface cell. If that interface cell is fully solidified in the current time step, the rejected solute is added to adjacent liquid cells. The third term represents solute redistribution at the graphite/liquid, graphite/austenite, or graphite/austenite/liquid interface during graphite growth, implemented by subtracting the absorbed solute from the remaining liquid or austenite in the same interface cell. If that interface cell has fully transformed to graphite, the absorbed solute is subtracted from adjacent liquid or austenite cells.

The simulation system includes three distinct phases (liquid, graphite, and austenite) and four different interfaces (austenite/liquid, graphite/liquid, graphite/austenite, and graphite/austenite/liquid). Carbon composition and diffusion coefficients vary significantly across different phases and interfaces. To effectively handle these discontinuities, equivalent composition Ce and equivalent diffusion coefficient De are employed to solve the governing equation. For example, equivalent compositions for liquid and austenite are Ce = CL and Ce = Cγ/kγ, respectively; equivalent diffusion coefficients are De = DL for liquid and De = Dγ for austenite. The equivalent diffusion coefficient between graphite and its neighboring phases is De = 0 because the equilibrium composition of graphite is 100% C. Although the concentration difference between graphite and its surrounding liquid or austenite is large, their chemical potentials are equal according to phase equilibrium principles, preventing carbon diffusion from graphite to liquid or austenite. Detailed determination methods for cell types and equilibrium concentrations in different cell types are described in reference [19].

The explicit finite difference method with no-flux boundary conditions is used to discretize and solve the governing equation. The stable time step Δt is determined by the carbon diffusion coefficient in liquid and mesh size Δx:

$$\Delta t = \frac{\Delta x^2}{4D_L}$$

During solidification, heat exchange with the mold causes temperature decrease, providing the driving force for solidification, while latent heat release from phase transformation causes temperature increase. The combined effect makes the system temperature vary with solidification time. Although temperature distribution and state vary throughout the casting at different temperatures, the simulation domain is relatively small, and the thermal diffusivity of Fe-C alloys is approximately three orders of magnitude larger than the carbon diffusion coefficient in liquid. Therefore, this model assumes uniform temperature within a microscale region, with temperature evolution governed by both heat extraction rate and latent heat release:

$$T = T_0 + \dot{T}t - \frac{L_{V,Gr} + L_{V,\gamma}}{\rho_{Fe} C_p}$$

where $\dot{T}$ is the heat extraction rate (cooling rate before and after phase transformation, determined by fitting cooling curves), Fe is the density of iron, Cp is the specific heat capacity of iron, and LV,Gr and LV,γ are the volume latent heats of graphite and austenite, respectively.

1.3 Graphite Growth Model

During spheroidal graphite cast iron solidification, graphite growth occurs in two stages: growth in liquid and growth within the austenite shell after being enveloped. As described previously, the local solutal equilibrium model is employed to calculate graphite growth, with the driving force being the difference between local interface equilibrium composition and actual composition. When graphite grows in liquid, interface curvature effects must be considered. The equilibrium composition at the graphite/liquid interface, CeqGr/L, is calculated as:

$$C_{eq}^{Gr/L} = C_0 + \frac{T^* - T_{eq}^{Gr/L}}{m_{L,Gr}} + \frac{\Gamma_{Gr}}{m_{L,Gr}} K_{Gr/L}$$

where T* is the interface temperature, TeqGr/L is the equilibrium temperature corresponding to the initial composition C0, mL,Gr is the liquidus slope of graphite, KGr/L is the curvature of the graphite/liquid interface, and ΓGr is the Gibbs-Thomson coefficient of graphite. In this 2D model, the average interface curvature is calculated by:

$$K_{Gr/L} = \frac{2\pi - \sum \theta}{V_C}$$

where VC is the area of each cell and ΣfGr is the total graphite volume fraction belonging to the same graphite nodule.

Based on interface solute conservation and considering the density difference between graphite and iron, the increment of graphite volume fraction ΔfGr in a graphite/liquid interface cell during one time step is:

$$\Delta f_{Gr} = \frac{D_L(C_{eq}^{Gr/L} - C_L^{Gr/L})G_{Gr}}{\rho_{Gr}(C_{Gr} - C_{eq}^{Gr/L})}$$

where Gr is the density of graphite, CGr/L is the composition at the graphite/liquid interface obtained from the concentration field calculation, and GGr is a geometry factor related to neighboring cell states:

$$G_{Gr} = \sum_I \min(1, S_I) + \frac{1}{\sqrt{2}} \sum_{II} \min(1, S_{II})$$

where SI and SII are state parameters of the four nearest-neighbor and four second-nearest-neighbor cells, respectively. The geometry factor GGr accounts for the influence of neighboring graphite cells on the growth of the current cell, considering that nearest neighbors have greater influence than second-nearest neighbors.

When graphite grows within austenite, the equilibrium temperature TeqGr/γ and solidus slope ms,Gr for the graphite/austenite interface replace TeqGr/L and mL,Gr in equation (5), respectively, while interface curvature effects are neglected. The equilibrium composition at the graphite/austenite interface, CeqGr/γ, is:

$$C_{eq}^{Gr/\gamma} = C_0 + \frac{T^* - T_{eq}^{Gr/\gamma}}{m_{s,Gr}}$$

Similar to equation (7), the increment of graphite volume fraction ΔfGr at the graphite/austenite interface during one time step is:

$$\Delta f_{Gr} = \frac{D_\gamma (C_{eq}^{Gr/\gamma} - C_\gamma^{Gr/\gamma}) G_{Gr}}{\rho_{Gr}(C_{Gr} - C_{eq}^{Gr/\gamma})}$$

where CγGr/γ is the local average composition of austenite at the graphite/austenite interface. For an interface cell I, the local average composition is calculated as:

$$C_\gamma^{Gr/\gamma} = \frac{C_\gamma^I + \sum_J C_\gamma^J}{N+1}$$

where CγI is the austenite composition in interface cell I, ΣCγJ is the sum of austenite compositions in neighboring interface cells, and N is the number of neighboring interface cells. The local austenite composition is obtained from the concentration field calculation.

When the graphite volume fraction in a graphite/liquid or graphite/austenite interface cell reaches 1, the cell state transforms from interface to graphite, and new neighboring graphite/liquid or graphite/austenite interface cells are captured, allowing continued graphite growth.

1.4 Austenite Growth Model

The local solutal equilibrium method is also employed to calculate austenite dendrite growth kinetics, considering preferred growth direction, interface curvature, and anisotropy of interfacial energy and growth kinetics. The driving force for dendrite growth is the difference between actual and equilibrium compositions at the dendrite/liquid interface. The equilibrium composition at the austenite/liquid interface, Ceqγ/L, is calculated as:

$$C_{eq}^{\gamma/L} = C_0 + \frac{T^* - T_{eq}^{\gamma/L}}{m_{L,\gamma}} + \frac{\Gamma_\gamma}{m_{L,\gamma}} K_\gamma f(\theta)$$

where mL,γ is the liquidus slope of austenite, Γγ is the Gibbs-Thomson coefficient of austenite, Kγ is the austenite/liquid interface curvature calculated by the counting cell method, and f( ) is the interfacial energy anisotropy function:

$$f(\theta) = 1 + \delta_t \cos[4(\theta - \theta_0)]$$

where δt is the interfacial energy anisotropy strength and 0 is the preferred dendrite growth direction.

Based on interface solute conservation, the increment of austenite volume fraction Δfγ in an austenite/liquid interface cell during one time step is:

$$\Delta f_\gamma = \frac{D_L(C_L^{\gamma/L} - C_{eq}^{\gamma/L})G_\gamma f_k(\theta)}{\rho_{Fe}(C_{eq}^{\gamma/L} - C_\gamma^{\gamma/L})}$$

where Ceqγ/L is the equilibrium composition calculated from equation (12), Cγ/L* is the actual composition at the austenite/liquid interface obtained from the concentration field, Gγ is the geometry factor calculated similarly to equation (8) but using austenite volume fraction instead of graphite, and fk( ) is the interface kinetic anisotropy function:

$$f_k(\theta) = 1 + \delta_k \cos[4(\theta - \theta_0)]$$

where δk is the interface kinetic anisotropy strength.

When the austenite volume fraction in an austenite/liquid interface cell reaches 1, the cell state transforms from interface to austenite, and neighboring liquid cells are captured as new austenite/liquid interface cells, enabling continued austenite growth. When graphite grows within austenite, the increment of graphite volume fraction in a graphite/austenite interface cell equals the decrement of austenite volume fraction.

1.5 Coupling of Graphite and Austenite Growth Models

The coupling method integrating nucleation and growth of graphite and austenite with concentration and temperature field calculations proceeds as follows: At simulation start, all cells are initialized with uniform initial concentration and temperature. The domain cools at a rate determined by heat extraction. Based on the instantaneous temperature, nucleation criteria for graphite and austenite are evaluated. When satisfied, nucleation occurs and state values are assigned to nucleated cells. Multiple interface types coexist during solidification. During austenite growth, the austenite fraction increment is calculated using equations (12)-(15). Graphite growth in liquid and austenite is calculated using equations (5)-(8) and (9)-(11), respectively. Carbon diffusion and solute redistribution due to phase transformation are computed using the governing transport equation. As mentioned, the graphite-to-iron density ratio is considered. To ensure solute conservation throughout the domain, an equivalent graphite carbon concentration is used during solute redistribution after graphite growth. Due to local solute redistribution errors inherent in the CA method, total solute content is monitored and compensated each time step before calculating carbon diffusion. Heat extraction causes temperature decrease, while latent heat release from graphite and austenite precipitation causes temperature increase. Their combined effect on domain temperature is calculated using equation (4). Changes in temperature and concentration fields subsequently affect graphite and austenite nucleation and growth, initiating a new cycle. Thus, the model captures the physical mechanism of divorced eutectic solidification microstructure evolution in spheroidal graphite cast iron, coupling concentration and temperature field calculations with nucleation, growth, and interaction processes. Physical properties of the Fe-C alloy used in calculations are listed in .

2. Results and Discussion

2.1 Microstructure Evolution in Hypereutectic Spheroidal Graphite Cast Iron

In this work, the effect of silicon is considered using carbon equivalent, calculated according to reference [21]. To compare some simulation results with experimental data, carbon equivalent is used to account for Si. [Figure 1: see original paper] shows the simulated microstructure and concentration field evolution during solidification of a spheroidal graphite cast iron with carbon equivalent C0 = 4.71 wt% (initial composition). The computational domain consists of 200 × 200 cells with a cell size of 2 m. For better comparison with experimental micrographs, nucleation positions for some graphite nodules and austenite were artificially set in the simulation shown in [Figure 1: see original paper]. The initial temperature is the graphite liquidus temperature corresponding to C0 (1357°C). As the Fe-C melt cools below the graphite liquidus temperature, graphite continuously precipitates from the liquid. When the temperature reaches the eutectic temperature Teut, austenite begins to form. The heat extraction rate above (Teut - 10°C) is -4.6exp(-0.0031t), and below this temperature is 1°C/s. The temperature field is influenced by both heat extraction and latent heat release from graphite and austenite solidification.

The numbers in [Figure 1: see original paper] indicate local carbon concentration. At solidification onset, graphite nucleates and grows as the primary phase in the liquid. Graphite growth absorbs carbon solute, reducing the carbon concentration in the domain below the initial composition C0 = 4.71 wt%, with slightly lower concentration near graphite compared to regions far from it ([FIGURE:1(a)]). When the temperature drops to the eutectic temperature, austenite nucleates. At this stage, the domain concentration approaches the eutectic composition Ceut = 4.28 wt%, with slightly higher carbon content in liquid near austenite and slightly lower near graphite ([FIGURE:1(b)]). Upon graphite-austenite contact, graphite becomes rapidly enveloped by austenite ([FIGURE:1(c)]). Subsequently, austenite gradually transitions from dendritic to globular growth, and carbon solute transports from liquid through the austenite shell to the graphite, enabling continued graphite growth within austenite. After most graphite nodules are enveloped, carbon concentration in the liquid gradually increases ([FIGURE:1(d)-(e)]) because austenite rejects solute during growth, and carbon diffusivity in austenite is much lower than in liquid, causing most rejected solute to enter the liquid phase. At solidification end, austenite shells impinge, completing solidification with multiple graphite nodules encapsulated within each austenite grain. As temperature continues to decrease, graphite continues to grow by absorbing carbon from the non-uniform austenite, with carbon concentration at graphite/austenite interfaces lower than in regions far from graphite ([FIGURE:1(g)]). Comparison between the simulated result at 740°C ([FIGURE:1(g)]) and an experimental micrograph ([FIGURE:1(h)]) shows good agreement.

[Figure 2: see original paper] shows the evolution of solid fraction, austenite fraction, and graphite fraction with temperature. Before the eutectic temperature, graphite grows in the liquid, increasing the graphite volume fraction. At the eutectic temperature, austenite forms and all three fractions increase rapidly, indicating that solidification proceeds mainly during the eutectic stage. After solid fraction reaches 100%, graphite fraction continues to increase slowly while austenite fraction decreases, demonstrating austenite-to-graphite transformation after solidification completion.

compares simulated graphite volume fraction and average radius at 740°C ([FIGURE:1(g)]) with experimental results ([FIGURE:1(h)]) and lever rule calculations based on the equilibrium phase diagram. Simulated graphite fraction and average radius were measured by both program calculation and Imagetool software, while experimental values were measured by Imagetool. The program-calculated and Imagetool-measured simulation values are very close. The simulated graphite volume fraction is slightly lower than the lever rule prediction because the lever rule assumes complete equilibrium, whereas CA simulation incorporates non-equilibrium kinetic factors. Conversely, the simulated value is slightly higher than the experimental result because experiments are three-dimensional, and the graphite cross-sections in [FIGURE:1(h)] may not represent the maximum diameters, while simulation is two-dimensional. Considering these factors, the simulation results are reasonable.

2.2 Interaction Mechanism Between Graphite and Austenite

To investigate the interaction and competitive growth mechanisms between graphite and austenite during solidification, the solidification process of a hypereutectic spheroidal graphite cast iron with C0 = 4.65 wt% was simulated. Four graphite nodules and one austenite dendrite were positioned in a 200 × 200 cell domain with 2 m cell size. The initial temperature was the graphite liquidus temperature for this composition (1329°C), cooling at 3°C/s until reaching (Teut - 10°C), then held constant. [Figure 3: see original paper] shows the evolution of graphite and austenite morphologies and concentration fields. During initial solidification, when temperature drops below the graphite liquidus, graphite nucleates in the liquid. As graphite grows by absorbing carbon solute, the carbon concentration in liquid near graphite becomes lower than in regions far from graphite. The concentration near Graphites II and III is slightly lower than near Graphite I ([FIGURE:3(a)]) because Graphites II and III jointly consume solute in their vicinity. As temperature decreases to the eutectic temperature, austenite nucleates ([FIGURE:3(b)]). During austenite growth, it first envelops the nearest Graphite I, then Graphites II and III, forming austenite shells that continue to grow. Because austenite growth rejects carbon solute while graphite growth absorbs it, carbon concentration near austenite becomes higher than near graphite ([FIGURE:3(b)-(g)]).

[Figure 4: see original paper] shows graphite growth velocity and radius evolution during solidification, with growth velocity calculated from the rate of increase in average graphite radius. Initially, all three graphite nodules grow rapidly. As they absorb carbon solute, liquid carbon concentration decreases quickly, creating concentration gradients. According to equation (7), decreased interface carbon concentration reduces graphite growth velocity, which subsequently reaches a relatively stable value ([FIGURE:4(a)]) because the surrounding concentration remains essentially constant ([FIGURE:3(b)-(d)]). From solidification start until 11 s, carbon concentration near Graphite I is slightly higher than near Graphites II and III ([FIGURE:3(a)-(c)]), resulting in slightly faster growth. Then austenite begins to envelop Graphite I ([FIGURE:3(d)]). Solute rejected during austenite growth promotes Graphite I growth, significantly increasing its growth velocity. After complete envelopment by austenite, Graphite I growth velocity rapidly decreases below that of Graphite II and stabilizes at a lower value. This occurs because graphite must absorb solute through the austenite shell, where carbon diffusivity is much slower than in liquid. Meanwhile, Graphite II continues growing in liquid, maintaining higher growth velocity. As solidification proceeds and austenite approaches Graphites II and III, their growth velocity variations during contact and interaction follow similar patterns: velocity increases significantly when approached and enveloped by austenite, then rapidly decreases to a steady value after complete envelopment. [FIGURE:4(b)] shows that initial graphite radii are similar, but Graphite I becomes larger during solidification. When each graphite is enveloped by austenite, radius evolution follows similar patterns to growth velocity. Throughout solidification, Graphite III exhibits the lowest growth velocity and radius due to competition from neighboring graphites, resulting in the smallest final size and elliptical morphology.

2.3 Effect of Cooling Rate on Solidification

In actual solidification processes, for a given spheroidal graphite cast iron composition, different cooling rates affect graphite nodule size and size distribution, thereby influencing casting properties. This work investigates cooling rate effects on graphite nodule size in a cast iron with C0 = 4.31 wt%. The simulation domain and cell size match those in [Figure 3: see original paper]. Graphite nodule numbers are determined from experimental measurements in reference [24] and converted from 3D to 2D using NA = (NV/2.37)^(2/3), where NA and NV are nodule numbers per unit area and volume, respectively. Reference [23] suggests particles smaller than 5 m are mostly inclusions and micropores, so these were excluded when calculating nodule numbers from reference [23], yielding nucleation densities of approximately 700 and 1400 nodules/mm^2 for 4 mm and 1.5 mm section thicknesses, respectively. Austenite nodule numbers are calculated using the nucleation model from reference [18]: NγA = 4.81\$×10^{7}\$ + 5.33\$×10$^{\{-6\}(dT/dt)+8.7\times10}${4}$(dT/dt)$^{2}\$. Since reference [23] does not provide cooling rates for different section thicknesses, heat extraction rates from reference [24] are used: 16°C/s and 21°C/s for 4 mm and 1.5 mm plates, respectively. Domain cooling rate is influenced by both heat extraction and latent heat release. Based on experimental observations that undercooling for graphite and austenite nucleation increases with cooling rate, nucleation temperatures for the 4 mm plate are set as: primary graphite at Tliqu, austenite at Teut - 15°C, and eutectic graphite at Teut - 35°C. For the 1.5 mm plate, nucleation temperatures are Tliqu - 20°C, Teut - 25°C, and Teut - 45°C for primary graphite, austenite, and eutectic graphite, respectively. Nucleation positions are randomly determined.

[Figure 5: see original paper] and [Figure 6: see original paper] show simulated graphite size distributions and microstructures at 740°C for different heat extraction rates. As cooling rate increases, graphite nodule number increases, size distribution peaks become more pronounced and shift toward smaller radii, and maximum nodule diameter decreases. This indicates that higher cooling rates produce smaller graphite nodules. At low cooling rates, the size distribution exhibits two peaks due to the two-step nucleation mechanism of primary and eutectic graphite. As cooling rate increases, the time interval between the two nucleation steps shortens, limiting primary graphite growth before eutectic graphite formation, resulting in a single-peak distribution. Comparison of [Figure 5: see original paper] and [Figure 6: see original paper] with experimental data from reference [23] shows good agreement in the variation of graphite size distribution and morphology with cooling rate.

To further investigate cooling rate effects on average graphite nodule radius, additional simulations for 3 mm and 2 mm section thicknesses were performed. Nodule numbers were determined using the same method: nucleation densities of 880 and 1160 nodules/mm^2 for 3 mm and 2 mm plates, respectively, with heat extraction rates of 18°C/s and 20°C/s. For the 3 mm plate, nucleation temperatures are Tliqu - 8°C, Teut - 19°C, and Teut - 39°C for primary graphite, austenite, and eutectic graphite, respectively. For the 2 mm plate, they are Tliqu - 16°C, Teut - 23°C, and Teut - 43°C. Other simulation conditions match those in [Figure 5: see original paper]. [Figure 7: see original paper] shows the relationship between average graphite radius and solidification time. As solidification time shortens (higher cooling rate), average graphite radius decreases because higher cooling rates produce more graphite nodules while the final graphite volume fraction remains essentially constant for a given composition. The simulation results are consistent with experimental trends.

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3. Conclusions

1. A multi-phase cellular automaton (MCA) model was developed to simulate microstructure evolution in spheroidal graphite cast iron solidifying via a divorced eutectic mechanism. The model employs a local solutal equilibrium approach to calculate graphite and austenite growth, using the difference between interface equilibrium composition and actual composition as the growth driving force. The graphite-to-iron density ratio is incorporated in graphite growth kinetics calculations. The model successfully reproduces experimentally observed divorced eutectic solidification microstructures. Simulated graphite volume fraction and average radius are slightly higher than experimental measurements but slightly lower than lever rule predictions, demonstrating the kinetic effects captured by the simulation.

2. The interactive and competitive growth mechanisms between graphite and austenite were analyzed. Results show that austenite growth in liquid promotes neighboring graphite growth, both phases are controlled by carbon diffusion, and graphite growth velocity decreases significantly after being enveloped by austenite due to lower carbon diffusivity in austenite than in liquid. Competitive growth also occurs between adjacent graphite nodules. Austenite grows dendritically before contacting graphite nodules, then transitions to globular growth after enveloping them.

3. Cooling rate effects on final graphite nodule size distribution and average radius were investigated. At low cooling rates, the size distribution shows two peaks from primary and eutectic graphite two-step nucleation. As cooling rate increases, the interval between nucleation steps shortens, limiting primary graphite growth before eutectic graphite formation, resulting in a single peak that shifts toward smaller radii. Higher cooling rates produce shorter solidification times and smaller average graphite nodule radii. Simulation results agree well with experimental data from literature.

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References

[1] Lei F J. Hot Working Technol, 2008; 37: 125

[2] Zhou J Y. Color metallography of cast irons. Beijing: China Machine Press, 2002: 98

[3] Shi W F, Xu Q Y, Gong M, Liu B C. Acta Metall Sin, 2011; 47: 620

[4] Wu M W, Xiong S M. Acta Metall Sin, 2010; 46: 1534

[5] Jiang H X, Zhao J Z. Acta Metall Sin, 2011; 47: 1099

[6] Zhang X F, Zhao J Z. Acta Metall Sin, 2012; 48: 615

[7] Dai T, Zhu M F, Chen S L, Cao W S, Hong C P. Acta Metall Sin, 2008; 44: 1175

[8] Yang Z R, Sun D K, Pan S Y, Dai T, Zhu M F. Acta Metall Sin, 2009; 45: 43

[9] Li Z Y, Zhu M F, Dai T. Acta Metall Sin, 2013; 49: 1032

[10] Charbon Ch, Rappaz M. In: Lesoult G, Lacaze J, editors. Physical Metallurgy of Cast Iron V. Switzerland: Scitec Publications; 1997: 453

[11] Ruxanda R, Beltran-Sanchez L, Massone J, Stefanescu D M. AFS Trans, 2001; 109: 1037

[12] Gurgul D, Burbelko A. Arch Metall Mater, 2010; 55: 53

[13] Burbelko A, Fras E, Gurgul D, Kapturkiewicz W, Sikora J. Key Eng Mater, 2011; 457: 330

[14] Burbelko A A, Gurgul D, Kapturkiewic W, Górny M. Mater Sci Eng, 2012; 33: 012083

[15] Zhao H L, Zhu M F, Stefanescu D M. Key Eng Mater, 2011; 457: 324

[16] Zhu M F, Pan S Y, Sun D K, Zhao H L. ISIJ Int, 2010; 50: 1851

[17] Zhu M F, Stefanescu D M. Acta Mater, 2007; 55: 1741

[18] Stefanescu D M. Science and engineering of casting solidification. 2nd ed. New York: Springer; 2009: 220

[19] Zhu M, Zhang L, Zhao H, Stefanescu D M. Acta Mater, 2015; 84: 413

[20] Brown S G R, Williams T, Spittle J A. Acta Metall Mater, 1994; 42: 2893

[21] Stefanescu D M, Katz S. "Thermodynamics of Iron-Base Alloys" in ASM Handbook vol. 15 -Casting, S. Vishwanatan editor, ASM International, Materials Park, Ohio 2008, pp. 41-45

[22] Zheng H L, Sun Y C, Zhang N, Tian X L. ISIJ Int, 2010; 50: 1981

[23] Pedersen K M, Tiedje N S. Mater Charact, 2008; 59: 1111

[24] Torres J F L. PhD Thesis, Tuscaloosa, America: the University of Alabama, 2003

[25] Loper C R. AFS Trans, 1999; 523

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

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