An Efficient 3D Stochastic Model for Predicting the Columnar-to-Equiaxed Transition in Alloy 718

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Abstract. A three-dimensional (3D) stochastic model for simulating the evolution of dendritic crystals during the solidification of alloys was developed. The model includes time-dependent computations for temperature distribution, solute redistribution in the liquid and solid phases, curvature, and growth anisotropy. The 3D model can run on PCs with reasonable amount of RAM and CPU time. 3D stochastic mesoscopic simulations at the dendrite tip length scale were performed to simulate the evolution of the columnar-to-equiaxed transition in alloy 718. Comparisons between simulated microstructures and segregation patterns obtained with 2D and 3D stochastic models are also presented.

1. Introduction
A two-dimensional (2D) stochastic model has been proposed for the modelling of the evolution of dendritic morphologies during solidification [1]. The significance of the stochastic modeling approach is that the evolution of simulated microstructures can be directly visualized and compared with the actual microstructures from experiments at two different scales: dendrite grain characteristics such as grain size and location and size of the columnar-to-equiaxed transition can be visualized at the meso-scale, while dendrite morphology (including dendrite tip, various dendrite arm spacings, microsegregation patterns) can be viewed at the microscale.

One of the challenges associated with stochastic modeling is reduction in mesh anisotropy. Basically, because of mesh anisotropy, dendrites will grow aligned with the axis of the mesh or at 45 degrees independently of the initial crystallographic orientation, which can be called anisotropy in growth direction. Furthermore, dendrites aligned with the axis would have narrower tips than predicted by classic theories, which can be termed anisotropy in growth kinetics. To reduce the mesh anisotropy, a formulation to correct the curvature in 2D was proposed and successfully validated [2]. A detailed discussion about correcting the grid anisotropy using the phase field approach is presented in [3].

Detailed comparison of predictions with measured microstructures requires the 2D stochastic model to be extended to 3D. This is why several 3D models for simulating dendritic growth have been proposed in the literature including phase field approach [3, 4] and Lattice Boltzmann and Cellular Automaton approach [5]. To be efficient, these models require parallel computations and can be used only for relatively small domains. A 3D Cellular Automaton approach for modeling the evolution of dendritic grains has also been developed [6]. This 3D dendritic grain model is relatively fast but it cannot simulate the evolution of dendrites and of microsegregation patterns at the dendrite tip level.
The main goal of the current contribution is to utilize an efficient 3D stochastic mesoscale dendritic nucleation and growth model, which is based on the recently developed 3D stochastic mesoscale model presented in [12], to predict the columnar-to-equiaxed transition in alloy 718. The 3D model includes computations for temperature, solute, curvature and anisotropy at the dendrite tip level. Another goal of the paper is to compare the 2D and the 3D predictions in terms of microstructure evolution (including the columnar-to-equiaxed transition) and segregation patterns during the solidification of alloy 718.

2. Description of the Stochastic Dendritic Solidification Model

The mathematical representation of the dendritic solidification process of a binary alloy is considered in a restricted 3-D domain ($\Omega$). A cross section of this 3D domain is shown in Fig. 1. Here, $\mathbf{n}$ is the interface normal vector, $K$ is the mean curvature of the interface, and the curve $\Gamma$ represents the solid/liquid (S/L) interface which evolves in time and has to be found as part of the solution.

The solidification of binary alloys is governed by the evolution of the temperature ($T(x, y, z, t)$) and concentration ($C(x, y, z, t)$) fields that have to satisfy several boundary conditions at the moving S/L interface as well as the imposed initial and boundary conditions on the computational domain. The equations that describe the physics of the solidification process in 3-D Cartesian coordinates are as follows (see the 2D equations in [1, 6-8]).

- **Temperature** ($T$) in $\Omega$ (heat transfer equation):

  $$\rho c_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( K \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( K \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( K \frac{\partial T}{\partial z} \right) - \rho L \frac{\partial f_L}{\partial t} \quad \text{with} \quad f_L = 1 - f_S$$  

  (1)

  where $t$ is time, $\rho$ is the density, $K$ is the thermal conductivity, $c_p$ is the specific heat, $L$ is the latent heat of solidification, $f_L$ is the liquid fraction, $f_S$ is the solid fraction, and $x$, $y$ and $z$ are the domain coordinates.

- **Concentration** ($C$) in $\Omega$ (solute diffusion equation):

  In the liquid phase ($C_L$):

  $$\frac{\partial C_L}{\partial t} = \frac{\partial}{\partial x} \left( D_L \frac{\partial C_L}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_L \frac{\partial C_L}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_L \frac{\partial C_L}{\partial z} \right)$$

  (2)

  In the solid phase ($C_S$):

  $$\frac{\partial C_S}{\partial t} = \frac{\partial}{\partial x} \left( D_S \frac{\partial C_S}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_S \frac{\partial C_S}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_S \frac{\partial C_S}{\partial z} \right)$$

  (3)

  where $D_L$ and $D_S$ are the interdiffusion coefficients in the liquid and solid, respectively.

  Local equilibrium at the S/L interface on $\Gamma(t)$ (here, "*" means at interface):

  $$C_{S^*}^* = k \; C_{L^*}^*$$

  (4)
Solute conservation at the S/L interface:

\[
V_n^* C_L^* (k - l) = \left( -D_L \left( \frac{\partial C_L}{\partial x} + \frac{\partial C_L}{\partial y} + \frac{\partial C_L}{\partial z} \right) + D_s \left( \frac{\partial C_s}{\partial x} + \frac{\partial C_s}{\partial y} + \frac{\partial C_s}{\partial z} \right) \right) \cdot \vec{n}
\]

(5)

where \( V_n^* \) is the normal velocity of the interface and \( \vec{n} \) denotes the normal to the S/L interface that is pointing into the liquid (see Fig. 1).

The interface temperature \( T^* \) is defined as (assuming local equilibrium with both phases):

\[
T^* = T_{LE} + \left( C_L^* - C_o \right) m_L - \Gamma \vec{K} f(\phi, \theta)
\]

(6)

where \( T_{LE} \) is the equilibrium liquidus temperature of the alloy, \( m_L \) is the liquidus slope, \( \vec{K} \) is the mean curvature of the S/L interface, \( \Gamma \) is the Gibbs-Thomson coefficient, and \( f(\phi, \theta) \) is a coefficient that accounts for growth anisotropy, where \( \phi \) is the growth angle (i.e., the angle between the normal and the growth axis) and \( \theta \) is the crystallographic orientation angle.

**Figure 1.** A 3D domain for dendritic solidification

In Eq. (6), second term in the right side is the constitutional undercooling and the last term in the right side is the curvature undercooling. The interface temperature is also affected by the kinetic undercooling. The kinetic undercooling is not accounted for in this model since its effect becomes significant only at very high solidification velocities (i.e., in the rapid solidification regime). Also, the coefficient \( f(\phi, \theta) \) in Equation (6) assumes an axisymmetric approximation where the anisotropy and the interface shape are independent of the polar angle \( \phi \) in the \( x-y \) plane perpendicular to the growth axis [2].
The solidification process is governed by Eqs. (1) to (6) and a stochastic model for nucleation and growth. The numerical procedures for calculating the nucleation and growth, temperature and concentration fields as well as the growth velocity of the S/L interface are described in details in [9]. It consists of a regular network of cells that resembles the geometry of interest. The model is characterized by (a) geometry of the cell; (b) state of the cell; (c) neighborhood configuration; and (d) several transition rules that determine the state of the cell. In this work, the geometry of the cell is a cube. Each cell has three possible states: “liquid”, “interface”, or “solid”. The selected neighborhood configuration is based on the cubic von Neumann’s definition of neighborhood, that is the first order configuration and it contains the first six nearest neighbors. Solidification behavior depends to a great extent on the transition rules. In this model, the change of state of the cells from “liquid” to “interface” to “solid” is initiated either by nucleation or by growth of the dendrites.

An explicit finite difference scheme is used for calculating the concentration fields in the liquid and solid phases. Zero-flux boundary conditions were used for cells located at the surface of the geometry. The solution algorithm includes the “interface” cells by multiplying the concentration in the liquid by the liquid fraction and the concentration in the solid by the solid fraction of the particular interface cell. Also, during each time-step calculation and for each “interface” cell, the previous values of the liquid and solid concentrations are updated to the current values of the interface liquid and solid concentrations calculated with Eqs. (4), (5) and (7). The calculation of the interface liquid concentration, \( C'_L \) can be obtained from Eq. (6) as

\[
C'_L = C_L + \frac{(T^* - T_L^{eq}) + \Gamma K f(\varphi, \theta))}{m_L}
\]  

(7)

The procedures for calculating \( K \) and \( f(\varphi, \theta) \) are described below.

The average interface curvature for a cell with the solid fraction \( f_s \) is calculated with the following expression:

\[
K = \frac{1}{a} \left( \frac{f_s + \sum_{k=1}^{N} f_s(k)}{N + 1} \right)
\]  

(8)

where \( N \) is the number of neighboring cells and \( a \) is the mesh size. In the present 3D calculations, \( N = 26 \), that contains all the first order neighboring cells (including the diagonal cells). Equation (8) is a simple counting-cell technique that approximates the mean geometrical curvature (and not the local geometrical curvature). An improved curvature model is presented in [8]. A comparison of 2D simulations obtained with these curvature models was also provided in [8], based on the work presented in [10, 11].

The anisotropy of the surface tension (see Eq. (6)) is calculated as [2]:

\[
f(\varphi, \theta) = 1 + \frac{4\delta}{1 - 3\delta} \left( \cos^4(\varphi - \theta) + \frac{3}{4} \sin^4(\varphi - \theta) \right) \text{ with } \varphi = \arccos \left( \frac{V_z}{(V_x)^2 + (V_y)^2 + (V_z)^2} \right)^{1/2}
\]  

(9)

where \( \varphi \) is the angle between the normal direction to the solid-liquid interface and the [100] direction (growth axis), \( V \) is computed with Eq. (12) in [12], \( \theta \) is calculated with Eq. (7) in [12] and \( \delta \) accounts for the degree of anisotropy. For cubic symmetry, \( \delta = 0.047 \) [2].
The algorithm is presented in [12]. The simulation software was written in Visual Fortran 90. The model output consists of screen plotting at any chosen time of $C$, $T$, or color indexes of all cells. The values of $C$, $T$, and color indexes of all cells are also saved at selected times on the computer disk.

### 3. Simulation Results and Discussion

Thermo-physical properties of the alloys used in simulations are presented in Table 1. The domain size is 10mmx30mm in 2D and 10mmx30mmx2mm in 3D. Newton cooling boundary condition was applied only to the bottom of the computational domain. All other boundaries were perfectly insulated.

The surface heat transfer coefficient used in the present simulations is $h = 3.0 \times 10^3 \text{ W m}^{-2} \text{ K}^{-1}$. Zero-flux solute boundary conditions were applied at the boundaries of the computational domain (i.e., a closed system was assumed). The initial melt temperature was 1400 °C. Also, an initial concentration equal to $C_o$ was assumed everywhere on the computational domain.

| Property   | $\mu_N^C$ [m$^{-1}$ K$^{-2}$] | $\mu_E^C$ [m$^{-1}$ K$^{-2}$] | $L$ [J/kg] | $\rho$ [kg/m$^3$] | $K$ [W/m/K] | $c_p$ [J/kg/K] | $T_L$ [°C] |
|------------|-------------------------------|-------------------------------|------------|------------------|-------------|--------------|----------|
| Value      | $1 \times 10^6$               | $5 \times 10^8$               | $2.9 \times 10^5$ | 7620             | 30.1        | 720          | 1336     |

| Property   | $C_o$ [wt.%]  | $C_{eut}$ [wt.%] | $k_o$ [m$^2$ s$^{-1}$] | $D_L$ [m$^2$ s$^{-1}$] | $D_S$ [m$^2$ s$^{-1}$] | $m_L$ [%°C$^{-1}$] | $\Gamma$ [K m] |
|------------|---------------|------------------|------------------------|-----------------------|----------------------|-------------------|---------------|
| Value      | 5.0           | 19.1             | 0.48                   | $3 \times 10^{-9}$     | $1 \times 10^{-12}$   | -10.5             | $3.65 \times 10^{-7}$ |

In Fig. 2, a comparison of 2D and middle cross sections of 3D simulated microstructures during the unidirectional solidification of alloy 718-5 wt. % Nb is presented. The legend in Fig. 2 shows 256 color indexes ($CI$) (shown here in 16 classes, where each class contains 16 different colors), which are used for displaying the preferential crystallographic orientation angle.

The 2D computations were done by using the 2D stochastic dendritic model presented in [1, 7-9]. The competition between nucleation and growth of multiple columnar dendrites is evident in both 2D and 3D cases. The strong growth competition from the sample bottom (>30 dendrites) to 1/4 of the sample height (16 dendrites in 2D and 24 dendrites in 3D) to 1/2 of the sample height (7 dendrites in 2D and 10 dendrites in 3D) to 3/4 of the sample height (5 dendrites in 2D and 7 dendrites in 3D) can be observed. Note also that the columnar morphologies are slightly different in 3D than in 2D.

Figure 2 also shows a comparison between 2D and 3D computations in terms of columnar-to-equiaxed transition (CET). The competition between nucleation and growth of both equiaxed and columnar morphologies can be clearly shown in Fig. 2. It can also be seen from Fig. 2 that the CET starts at an earlier location in 3D than in 2D. The full transition also occurs earlier in 3D than in 2D (about 5/6 of the sample height in 3D and about 11/12 of the sample height in 2D). This is because (i) for a 2D geometry the grain growth is confined in the x-y plane while for the 3D case it is free in the third (z) direction; (ii) more nucleation sites are available in 3D than in 2D and (iii) some dendrites that are visualized in the middle plane of the 3D simulation are cut from a different plane.

Figure 3 shows a comparison of segregation patterns of 2D and cross sections of 3D simulated columnar dendritic morphologies. It can be observed that the segregation values are higher in 2D than in 3D in both liquid and solid phases.
Figure 2. Comparison of 2D ((a) and (c)) and middle cross sections of 3D ((b) and (d)) simulated microstructures during the unidirectional solidification of alloy 718-5 wt. % Nb.

Figure 3. Comparison of 2D ((a) and (c)) and middle cross sections of 3D ((b) and (d)) simulated segregation patterns during the unidirectional solidification of alloy 718-5 wt. % Nb.
The Nb segregation index \( SI \) shown in the legend in Fig. 3 is computed as follows:

\[
SI = \frac{C_{av} - k \times C_o}{C_{eut} - k \times C_o} \times 255 \quad \text{with} \quad C_{av} = C_S f_S + C_L f_L
\]  

Slightly higher segregation values are observed in 2D than in 3D in both liquid and solid phases. This is because the microstructure is finer in 3D than in 2D. In both Figures 2 and 3, the equiaxed grain density was \( 1 \times 10^8 \) nuclei/m\(^2\) in 2D and \( 1 \times 10^{12} \) nuclei/m\(^2\) in 3D. The conversion factor between 2D and 3D is \( f = \pi / 4 \) [14].

A RAM memory size of 100 bytes/cell is needed in the present computations. CPU-time is related to the domain size and the time-step. The 3D simulation problems shown in Figs. 2 and 3 took less than 1 day on a Dell Precision T7500 desktop.

4. Conclusions

An efficient 3D mesoscale stochastic model was applied to study the evolution of the microstructures including CET and the segregation patterns in alloy 718 - 5wt. % Nb.

Based on the current simulations, it was determined the following:

(i) The 3D columnar dendrites are finer than the 2D ones;
(ii) Segregation values are higher in 2D than in 3D in both liquid and solid phases;
(iii) The CET in 3D starts at an earlier location than in 2D and the full transition occurs earlier in 3D than in 2D;
(iv) The CET length is larger in 2D than in 3D.

References

[1] Nastac L 1999 Numerical Modeling of Solidification Morphologies and Segregation Patterns in Cast Dendritic Alloys, Acta Met 47(17) 4253-62.
[2] Karma A, and Rappel WJ 1998 Phys. Rev. E 57(4) 4323-4.
[3] Jeong JH, Goldenfeld N, Dantzig JA 2001 Phys. Rev. E 64(041602) 1-14.
[4] Eshraghi M, Felicelli SD, Jelinek B 2012, Journal of Crystal Growth 354(1) 129-34.
[5] Gandin CA, Rappaz M 1997 Acta Met 45(5) 2187-95.
[6] Nastac L 1999 A New Stochastic Approach for Simulation of Solidification Morphologies and of Segregation Patterns in Cast Dendritic Alloys, In 4th Pacific Rim International Conference on Modeling of Casting and Solidification Processes (MCSP-4), Ed. Hong CP, Yonsei University, Seoul, Korea.
[7] Nastac L 2000 A Stochastic Approach for Simulation of Solidification Morphologies and Segregation Patterns in Cast Alloys. In Proceedings of the Modelling of Casting, Welding, and Advanced Solidification Processes IX. Engineering Foundation. Aachen, Germany.
[8] Nastac L 2004 Modeling and Simulation of Microstructure Evolution in Solidifying Alloys, Springer New York (ISBN 978-1-4020-7831-6).
[9] Nastac L, Stefanescu DM 1997 Modelling and Simulation in Materials Science and Engineering Institute of Physics Publishing 5(4) 391-420.
[10] Kothe DB, Mjolsness RC, Torrey MD 1991 RIPPLE, A Computer Program for Incompressible Flows with free surfaces, Los Alamos National Lab, LA-10612-MS, Los Alamos, NM, USA.
[11] Chen S, Merriman B, Osher S, Smereka P 1997 J. Comp. Physics 135 8-29.
[12] Nastac L 2014 Journal of Metallurgical Research & Technology, EDP Sciences. 111, 311-319 (DOI: 10.1051/metal/2014016).
[13] Nastac L, Stefanescu DM 1997 Metallurgical Transactions 28A 1582-7.
[14] Kong M, Bhattacharyal RN, James C, Basu A 2005 Geological Society of America Bulletin 117(1-2) 244-9.