A hybrid stochastic-deterministic model for the multiphase solidification of alloys

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Abstract

A new mathematical model to predict the formation of grain structure during two-dimensional solidification of binary alloys is proposed. This model, which is called CADE (cellular automaton-deterministic), combines the cellular automaton technique with a deterministic multiphase model that is based on macroscopic conservation equations. The cellular automaton technique is used to track the growth and predict the impingement of each individual grain envelope, enabling accurate calculations of the grain fraction and the interfacial area concentration between grain envelopes and the surrounding liquid. The parameters calculated with the cellular automaton are transferred to the deterministic model, in which a system of differential equations is solved to determine several field variables, such as the temperature, the average liquid concentration, and the fraction solid. The complete CADE model has the advantage of grain structure prediction derived from the cellular automaton technique and the strong coupling between the equations from the deterministic model. The CADE model is applied to the isothermal solidification of an Al-Si alloy. The results are compared with those from a deterministic model published in the literature, showing reasonable agreement when the Avrami correction for grain impingement is used in the deterministic model.

Keywords: Cellular automaton, deterministic modelling, macrostructure.

1. Introduction

Mathematical models to predict the as-cast grain structure formed during solidification have been identified as stochastic (probabilistic) or deterministic. Stochastic models employ the definition of a random variable in some part of their calculations, whereas deterministic models rely only on deterministic equations. One of the first deterministic models of grain structure formation in solidification was proposed by Oldfield [1]. In this model, the microscopic growth of eutectic cells was coupled to the macroscopic heat transfer, originating a micro-macroscopic model. Important simplifying assumptions were adopted: considerations of only equiaxed, spherical (non-dendritic) grains, and a system of uniform temperature (isothermal). During the evolution of deterministic micro-macroscopic models, many of the simplifying assumptions were eliminated and other phenomena important to the formation of as-cast grain structure were included. Maxwell and Hellawell [2] and Greer et al. [3] have incorporated different nucleation models for spherical grains, predicting their final size. Rappaz and Thévoz [4] considered the solidification of spherical dendritic grains, aiming to predict the whole recalescence in the cooling curves of isothermal systems. Thévoz et al. [5] extended this model to the unidirectional solidification of equiaxed dendritic grains, considering a Gaussian distribution of critical undercoolings for nucleation. Predictions of the equiaxed grain radius along a unidirectionally solidified sample showed good agreement with measurements.

To predict the columnar-to-equiaxed transition (CET) in unidirectional steady-state solidification, Hunt [6] included the modelling of columnar dendritic growth in deterministic models. His model was later extended to unidirectional transient solidification [7]. Wang and Beckermann [8,9] proposed a one-dimensional deterministic multiphase model of equiaxed and columnar dendritic solidification to predict the CET, taking into account the interaction between solute fields surrounding equiaxed grains. This model was extended by Martorano et al. [10], who considered the interactions between the solute field surrounding equiaxed and also columnar grains. Although the extension of these deterministic models to predict the CET in two dimensions is complex, there have been a few attempts, by the implementation of special algorithms, to track the position of the two-dimensional columnar front [9,11–14]. Despite the difficulties experienced by deterministic models in two or three-dimensional problems, they have some important advantages: numerous phenomena have already been considered in their governing equations; reliable numerical methods are available to solve their strongly coupled system of differential equations; and relatively coarse numerical meshes are employed to solve their equations.

Stochastic models of solidification have been used to calculate a picture of the as-cast grain structure that can be directly compared with the actual macrostructure obtained experimentally. This is an important attractive feature of stochastic models and a fundamental difference from deterministic models, allowing stochastic models to easily predict not only the grain size in two and three dimensions, but also the CET. Gandin and Rappaz [15] developed a stochastic model, named CAFE, that combined the cellular automaton technique and the finite element method to successfully
predict grain structure formation and the temperature field during solidification. One major disadvantage of stochastic models, however, is the refined mesh necessary to simulate the growth of each grain. This problem becomes even worse in later extensions of stochastic models, which were used to predict phenomena occurring at the length scale of the dendrite arms, originating the so-called modified cellular automaton models [16,17]. In the original model proposed by Gandin and Rappaz [15], the smallest simulated length scale was that of the dendritic grains, i.e., dendrite arms were not considered. There have not been many attempts to incorporate different phenomena into this model, although it has been extensively used.

The main objective of the present work is to propose a hybrid stochastic-deterministic model, named CADE (cellular automaton-deterministic), which combines the strongly coupled differential equations of the deterministic models with the capabilities of predicting the geometry of solidifying grains presented by the cellular automaton technique. The differential equations have some variables, such as the concentration of interfacial area of grain envelopes and the volume fraction of grains, that depend on the geometry of solidifying grains. To calculate these parameters in the deterministic models, grain envelopes were usually assumed to be spherical or cylindrical by previous authors. In the present work, however, these parameters are obtained from the grain structure predicted by the cellular automaton. Moreover, the solute concentration in the liquid surrounding grain envelopes is calculated by the deterministic model and is used in the cellular automaton model to predict the interaction between solute fields surrounding all grains during growth.

2. Mathematical model

The CADE model implemented in the present work is a combination of governing equations used by deterministic models and the cellular automaton technique employed in stochastic models to predict grain growth during solidification. The details of the model are described below.

2.1 Deterministic model

Governing equations analogous to those proposed by Wang and Beckermann [9] and used by Martorano et al. [10] were adopted in the present work. These equations were based on the volume averaging of the equations of mass, energy, and species conservation within each pseudo-phase, namely, solid (s), interdendritic liquid (d), and extradendritic liquid (l). These pseudo-phases were defined according to a grain envelope concept in which an imaginary envelope encloses each grain. The liquid within the envelope is the interdendritic liquid, while the liquid outside, surrounding the envelope, is the extradendritic liquid. The following assumptions were used to derive the final macroscopic equations for a binary alloy; (a) the system is isothermal, i.e., the temperature is uniform; (b) melt flow, movement of solid, and solute diffusion in the solid are negligible; (c) all grains nucleate immediately below the liquidus temperature (instantaneous nucleation), i.e., the critical undercooling for nucleation is assumed to be zero; (d) the specific heats, \( c_p \), and the densities, \( \rho \), of the pseudo-phases are equal and constant; and (e) the solute concentration in the interdendritic liquid, \( C_{i} \), is uniform and is related to the temperature \( T \) by the liquidus line of the phase diagram, i.e., \( T = T_f + m_i C_i \), where \( T_f \) is temperature, \( T_f \) is the melting point of the pure metal, and \( m_i \) is the slope of the liquidus line from the phase diagram.

The final governing equations of the model are as follows

\[
\frac{dT}{dt} = R + \frac{L_s}{c_s} \frac{dt}{dt}
\]

\[
(t-k)C_i \frac{dC_i}{dt} = \epsilon_i \frac{dC_i}{dt} + \frac{S_{l}}{D_l} (C_i - C_l)
\]

\[
\frac{d}{dt}(e_i, C_i) = C_i \frac{dE_i}{dt} + \frac{S_{l}}{D_l} (C_i - C_l)
\]

\[
\epsilon_s + \epsilon_i + \epsilon_l = 1
\]

where \( R \) is the cooling rate of the isothermal system before the beginning of solidification; \( C \) is the solute concentration in the phase indicated by the subscript; \( t \) is time; \( L_s \) is the latent heat of fusion; \( k \) is the solute partition coefficient; \( D_l \) is the diffusion coefficient of solute in the extradendritic liquid; \( \delta_l \) is an effective diffusion length for solute transport between dendrite envelopes and the extradendritic liquid; \( S_{l} \) is the surface area of grain envelopes per unit volume; and \( \epsilon \) represents the volume fraction, where its subscript indicates the pseudo-phase. The volume fraction of grain envelopes, \( \epsilon_i \), is defined as \( \epsilon_i = \epsilon_s + \epsilon_l \). Note that, in the present work, \( \epsilon_i, S_s, \) and \( \delta_l \) depend on the grain structure, which is obtained from the stochastic part of the present model. In the model used by Wang and Beckermann [9] and Martorano et al. [10], these parameters were completely calculated with deterministic equations based on the assumption of spherical and cylindrical envelopes. Equations 1 to 4 were solved numerically by the finite volume method, explicit formulation [18].

2.2 Stochastic model

The cellular automaton (CA) technique proposed by Gandin and Rappaz [15] was adopted in the present model to calculate the grain structure of a two-dimensional melt of square shape that solidified isothermally. The...
two-dimensional melt is subdivided by a mesh of square CA cells containing a CA site at the centre. Substrates for heterogeneous nucleation are distributed randomly among the CA cells, with no more than one substrate per cell. When the temperature of the isothermal system, \(T\), is below the liquids temperature, all substrates are activated, i.e., a square representing a grain envelope, centred at the CA site and of orientation chosen randomly between 48 classes in the range of \(-45^\circ\) and \(45^\circ\), is created at the cells with a substrate. To simulate the growth of grain envelopes, these squares begin to grow with time and their half-diagonals increase with velocity \(V\) given as [19]:

\[
V = \frac{D \cdot m \cdot (k - 1) \cdot C_l}{\pi \cdot \Gamma} \cdot \left[ 0.4567 - \frac{\Omega}{(1 - \Omega)} \right]^{1/4} \tag{5}
\]

where \(\Gamma\) is the Gibbs-Thomson coefficient and \(\Omega\) is a dimensionless undercooling, defined as \(\Omega = (C_d - C_l) / (C_d(1 - k))\), in which \(C_d\) and \(C_l\) are given by the deterministic part of the model. The dependence of \(\Omega\) on \(C_l\), the solute concentration in the extradendritic liquid, simulates the interaction between the solute fields surrounding the grain envelopes, which was not taken into account in the original CA technique [15]. When the growing square reaches the position of a neighbour site that does not have a substrate for nucleation, this neighbour site is activated by associating with it another square with centre and size defined as proposed by Gandin and Rappaz [15]. The diagonals of this square also begin to grow with the velocity given by Equation 5. Further details of this algorithm can be found elsewhere [15].

The governing equations of the deterministic part of the model depend on some variables, namely, \(S_e\) and \(\varepsilon_g\), that are related to the geometry of the three-dimensional microstructure. As opposed to the deterministic models proposed previously, in the present work these variables are obtained from the two-dimensional macrostructure calculated by the stochastic part of the model. First, the area fraction of envelopes, \(A\), and the perimeter of envelopes (interface with the extradendritic liquid) per unit area, \(L\), are obtained from the two-dimensional cellular automaton macrostructure. Next, stereological relations are used to calculate \(S_e\) and \(\varepsilon_g\), which are transferred to the deterministic model equations. Two sets of stereological relations were used to simulate two different solidification systems. In the first system, all grain envelopes were assumed as cubes equally spaced of equal size and orientation. The CA calculation plane was considered to be parallel to one face of these cubic envelopes. Therefore, the image of any envelope at the calculation plane was always a square whose sides had the same size as those of the cubic envelope. For this system, the simulation domain was a square unit cell containing only one grain envelope. The stereological relations used were \(\varepsilon_g = A^{1/2}\), \(S_e = 3L^{2}/8A^{1/2}\), and \(n_v = n^{1/2}\), where the

![Figure 2: CADE model results for equidistant and equally oriented cubic envelopes are compared with those from an implementation of the deterministic model (Det) of Wang and Beckermann [8] adjusted for cubic envelopes: (a) cooling curves; (b) solid (\(\varepsilon_s\)) and grain (\(\varepsilon_g\)) volume fractions; (c) solute concentration in extradendritic liquid (\(C_l\)); (d) concentration of interface area between envelope and extradendritic liquid (\(S_e\)).](image-url)
number of substrates for heterogeneous nucleation per unit volume is \( n_a \), and per unit area of the CA calculation plane is \( n_v \). In the second system, the cellular automaton calculation plane was assumed to be a random section through a three-dimensional set of randomly positioned and oriented grain envelopes. In this case, \( \varepsilon_g = \frac{4}{3} \pi L_A^3 \) [20]. When the mean diameter of the envelopes is calculated by assuming they consist of equivalent spheres of equal size, \( n_v = \frac{\sqrt{\pi}}{\sqrt{6}} n_a^{3/2} \) [15]. The effective diffusion length, \( \delta_e \), was calculated by assuming that all grain envelopes are equal sized spheres, as described by Martorano et al. [10].

3. Results and Discussion

The numerical procedures to obtain \( A_A \) and \( L_A \) from the CA macrostructure were verified by simulating the growth of one square envelope centred in a square domain (1.5 x 1.5 mm) and with an orientation of 45° in relation to one side of this domain. The CA calculation results for both a coarse (100 x 100) and a refined (1000 x 1000) two-dimensional mesh show very good agreement with those obtained with an exact analytical expression (Figure 1). The values of \( A_A \) calculated for the coarser mesh, however, change in small steps. After the corners of the square reached the square domain boundary, the value of \( L_A \) began to decrease, because the envelope perimeter that was in contact with the outside liquid decreased. An analogous effect is observed when two envelopes impinge during solidification.

The CADE model was then used to simulate an Al-5 wt%Si alloy solidifying from its liquidus temperature (894.5 K) in the form of equal cubic envelopes equidistant from each other and oriented 30° in relation to the side (1.6 mm length) of the square unit cell, as described before. In Figure 2, results from this simulation are compared with those obtained with an implementation of the deterministic model proposed by Wang and Beckermann [8], adjusted for the isothermal solidification of equiaxed cubic envelopes, rather than the original spherical ones. The CA mesh size was 200 x 200 and the alloy properties and simulation conditions were: \( D_l = 3 \times 10^{-9} \text{ m}^2 \text{ s}^{-1} \); \( L_p = 372 \times 10^3 \text{ J kg}^{-1} \); \( c_p = 921.5 \text{ J kg}^{-1} \text{ K}^{-1} \); \( m_s = -7.7 \text{ K (wt pct)}^{-1} \); \( k = 0.117 \); \( \Gamma = 9 \times 10^{-9} \text{ m} \text{s}^{-1} \); \( T_f = 933 \text{ K} \); \( R = 45 \text{ K s}^{-1} \); and \( n_v = 2.4 \times 10^8 \text{ m}^{-3} \). The cooling curve (including the recalescence) and curves for time evolution of \( \varepsilon_s, \varepsilon_g, C_l, C_d, S_e \) calculated with the CADE model coincide with those given by the model of Wang and Beckermann [8] until 0.8 s. After this time, they differ because the square envelope impinges on the unit cell boundary, causing \( S_e \) to decrease with further envelope growth (Figure 2d).

The second solidification system, in which 30 grains were assumed to nucleate with random orientation and position, was simulated with the CADE model using a mesh of 200 x 200 cells in a square domain of length 7.9 mm. In
Figure 3, the model results are compared with those from the adjusted deterministic model of Wang and Beckermann [8]. Some discrepancy is observed for the cooling curves and the time evolution of $\varepsilon_g$, $C_l$, $S_e$. Nevertheless, the evolution of fraction solid ($\varepsilon_s$) agreed very well. The grain structure calculated with the CADE model is embedded in Figure 3a, showing a typical equiaxed structure. In Figure 3d, the concentration of envelope interface area, $S_e$, calculated with the CADE model begins to decrease after approximately 1 s, when the volume fraction of grains ($\varepsilon_g$) reaches about 0.4.

After this moment, the effect of impingement of neighbour envelopes on each other becomes increasingly important. This effect is not observed in the deterministic model, in which $S_e$ increases up to a plateau, because no correction for impingement (Avrami correction) was considered in the original model [8]. To examine this source of discrepancy, the Avrami correction was included in the purely deterministic model equations and the results are also presented in Figure 3. Now, Figure 3d shows that $S_e$ for the deterministic model also begins to decrease at approximately 1 s, showing a trend similar to that of the CADE model. This correction resulted in better agreement between all of the CADE and deterministic model results, especially the cooling curves.

During recalescence, the agreement was improved mainly after the maximum temperature. The agreement for $\varepsilon_g$ and $C_l$ also improved with the Avrami correction, because it decreases the envelope interfacial area, through which the envelope grows and solute is rejected to the extradendritic liquid. Therefore, the rate of increase of $\varepsilon_g$ and $C_l$ with time decreases, approaching the CADE results.

### 4. Concluding remarks

A hybrid stochastic-deterministic model, named CADE (cellular automaton-deterministic), has been proposed to simulate the solidification of binary alloys. This model combines the governing equations from the deterministic model proposed by Wang and Beckermann [8] and Martorano et al. [10] with the cellular automaton technique developed by Gandin and Rappaz [15]. The CADE model was used to simulate two different solidification systems: equidistant cubic envelopes of the same orientation and a set of 30 envelopes randomly oriented and positioned in a two-dimensional square domain. The CADE results (cooling curves, time evolution of solid and grain fraction, solute concentration and envelope interfacial area) showed very good agreement with the results of a purely deterministic model for the equally oriented cubic envelopes until the envelopes impinged on the unit cell boundary. Some discrepancy was observed, however, between the CADE results and those from the deterministic model for the set of 30 random grains. Part of this discrepancy was caused by the impingement of envelopes on each other, which was simulated by the CADE model, but not considered in the original deterministic model.