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Modeling of Two-Stage Solidification: Part I Model Development

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Abstract

The paper presents a new numerical model of solidification processes in hypoeutectic alloys. The model combines stochastic elements, such as e.g. random nucleation sites and orientation of dendritic grains, as well as deterministic methods e.g. to compute velocity of dendritic tips and eutectic grains. The model can be used to determine the temperature and the size of structure constituents (of both, the primary solid phase and eutectics) and the arrangement of individual dendritic and eutectic grains in the consecutive stages of solidification. Two eutectic transformation modes, typical to modified and unmodified hypoeutectic alloys, have been included in the model. To achieve this, cellular automata and Voronoi diagrams have been utilized.

Keywords: Cellular automata, Solidification, Microstructure modeling, Dendritic structure, Eutectic structure

1. Introduction

Modeling of hypoeutectic alloys solidification is a complex problem, because increase of solid phase fraction is triggered by constitution of two structures: In the first stage, between eutectic and liquidus temperature, dendritic grains nucleate and grow. In the second stage, below eutectic temperature, eutectic structure forms. In the latter stage eutectic grains nucleate and grow from the liquid phase that did not solidify in the first stage. As has been reported recently [1-4], various modes of eutectic transformation can be observed, depending on the alloy modifications. In unmodified alloys, eutectic grains nucleate in adjacency of dendritic grains. Whereas, in modified alloys (e.g. by adding strontium modifier) eutectic grains nucleate with no relation to dendritic structure, and the number of eutectic grains is smaller compared to the unmodified alloys.

The eutectic formation is the final stage of the hypoeutectic alloys solidification process, and hence the mode of eutectic transformation can modulate occurrence of defects as has been shown in [4]. Consequently, model that would accurately capture various modes of eutectic transformation can contribute to better prediction of defects (e.g. porosity or hot-tearing) in castings. The above observation is the main motivation for current work.

Although to predict dendritic and eutectic structures numerous approaches, including those based on cellular automata technique, have been developed [5-13], there are no models that would allow to capture simultaneously microstructure, temperature and, size of the structure constituents in the two-stage solidification, and that would include different modes of the eutectic transformation.

In this paper a novel model of two-stage solidification, typical to hypoeutectic alloys, is presented. The model has been proposed for two dimensional cases, where multi grain structure is modeled. Two phenomena, nucleation and growth, contribute to the process of evolution of dendritic and eutectic structures. In the paper, details of the nucleation and growth models are given. To describe microstructure evolution cellular automata have been utilized, because of their flexibility and extensibility. In this approach every grain is taken into account individually, thus interactions between grains and structures become inherent to the

model, and there is no need to apply any correction factors, as it is done in the case of models that concern representative elementary volume [6-7, 9-12]. Moreover, the approach captures both temporal and spatial evolution of polistructure. Finally, using proposed model numerical maps of transition between different stages in structure development can be obtained.

2. Nucleation and growth of dendritic grains

The first stage of the process involves nucleation and growth of the primary phase dendrites. The growth process has been divided into two subprocesses, i.e. growth of dendritic envelopes and growth of the primary solid phase crystals. Here, we introduce a notion of the dendrite envelope to separate phenomena that occur on different length scales, such as e.g. solute diffusion process. The envelope is defined as a smooth surface that encloses the primary and the secondary dendrite arms, and connects the outer-most points of the crystal. In this way, the envelope allows to approximate complex shapes of dendritic grains by the virtual surface, and it simplifies analysis of the problem.

The growth process of the dendritic grains is based on the following assumption. Constitutional undercooling is the driving force for the dendritic envelope growth. Increase of solid phase is computed on the basis of heat and solute balances. Within liquid phase, outside the grain envelope, the solute diffusion equation is solved, and the full solute mixing within liquid phase inside the grain envelope is assumed. In turn, within the solid phase, there is no solute diffusion. Note that although our assumptions are similar to those proposed in [12], our problem is significantly harder because we are considering all grains individually. Moreover, the process we analyze involves two types of structure that need to be traced separately. Keeping this in mind, to simulate the process of grain growth we employ cellular automata (CA), due to their flexibility and extensibility. To introduce a new type of structure it is sufficient to add its corresponding state to the set of CA feasible states, and to define resulting transition rules.

The main scheme of computations, and the assumptions regarding solute and heat processes, are analogous to that proposed in [12], and hence we limit further considerations only to those parts of the model that need to be reformulated. Nevertheless, for the sake of clarity, we give the general idea of the model below.

CA operate on the mesh of cells for which Moore neighborhood is assumed. At the beginning of the simulation all cells are in liquid state with the initial solute concentration assigned. When the temperature drops below liquidus temperature, nucleation sites are chosen randomly among all liquid cells. These cells are changed into growing cells and become the origin of dendritic grains. The number of cells that are changed into growing cells is determined by means of a nucleation model, here instantaneous nucleation model.

In every time step, the algorithm of envelope growth is applied. Then, in the same time step, for every cell that does not belong to the growing dendritic grains, and that is in liquid state, solute diffusion equation is solved, and solute concentration associated with it is updated. Moreover, the solute content within this cell is computed, and is next used to achieve solute balance. In our approach the solute balance is obtained on the basis of solute content in CA-cells, depending on their state. The envelope growth algorithm allows to compute a grain fraction, whereas solute content in extragranular area is a result of solute content computations for every CA-cell in every time step. By combining the above quantities with the solute and heat balances, in every time step we can compute and update the current temperature and the current solid phase (see [12] for detailed explanation).

2.1 Growth of dendritic grain envelopes

As we already pointed out, to consider multigrain growth several problems have to be solved. In Figure 1 several growing grains with their envelopes have been presented. A growing grain rejects solute into surrounding liquid, and as a result a solute diffusion layer builds around it (as shown in Figure). When the layers associated with neighboring grains overlap, rejected solute is accumulated in the place of contact. This has additional effect, it leads to the decrease in the growth velocities of some parts of the envelopes in this area. This effect is due to decrease in solutal undrecooling, which is the driving force of growth. Because dendritic grains have random locations, their diffusion layers will start interacting in different time steps. Note, that all above characteristics should be reflected in the model.

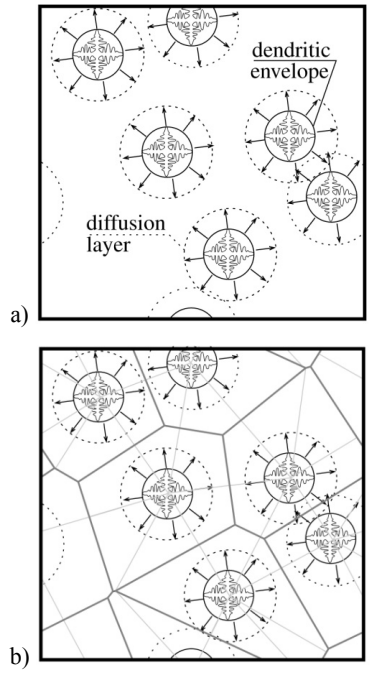

Fig. 1. Schematic presentation of dendritic grains growth (a) together with corresponding Voronoi Diagram (b): Contact of the diffusion layers takes place in the hyperplane of VD. In the model periodical boundary conditions are applied

Consequently, the main challenge is to find, unambiguously, the point from which the solute concentration far from the dendrite envelope will be taken. Additionally, when the solute diffusion layers of neighboring grains overlap with each other, the growth of their envelopes in this area should be slowed down. At the same time in the area where the diffusion layers do not overlap, the growth of grain envelopes should be unrestrained. To address the above issues we use Voronoi Diagrams (VD). This however requires the following conditions to be satisfied:

- the dendrite envelope must be approximated by a sphere,
- conditions for the grain growth, including temperature, must be homogeneous within the considered area,
- instantaneous nucleation model must be used.

The first and the last condition are common assumptions and can be found in other grain growth models [9-11]. In turn, the size of the considered area can be tuned to satisfy the second condition.

If the above requirements are satisfied, contact of the solute layer of neighboring grains will take place in the hyperplane of VD. Thus, the solute concentration far from the envelope can be determined uniquely, which property is exploited in the envelope growth algorithm.

Growth algorithm of dendritic grains envelopes

Once the nucleus sites have been randomly chosen, their corresponding Voronoi Diagram is determined. Parameters of VD do not change in the consecutive time steps, thus VD generation is one time effort. Each grain is assigned one cell of the VD. A cell of the VD is characterized by its vertices and hyperplanes. The first cell of grain, which is its nucleus, is attached as so called growing cell. It is defined to be the center of the entire grain.

Each growing cell is characterized by the local envelope radius (we define radius to be distance from the center of a grain). In every time step, for every growing cell, the local envelope radius is updated and its neighborhood is built. Then, we check if any cell from the neighborhood has been engulfed by the envelope. If the distance from the grain center to the center of neighboring cell is smaller than the local envelope radius, then this cell is engulfed by the envelope and becomes a growing cell. In the next time step the growth algorithm is called for such engulfed cell. The cell inherits the local envelope radius from its parent (i.e. engulfing) cell. When neighborhood of a cell consists of growing cells only, the cell itself is no longer considered growing.

To update the local envelope radius R_g for the growing cell, the growth law [14] is utilized:

$$\frac{dR_g}{dt} = \frac{Dm}{\pi^2 \Gamma c_0 (k-1)} \left(c^{*\prime} - c_\infty\right)^2 \tag{1}$$

where D is the solute diffusion coefficient, m is the slope of the liquidus line, Γ is the Gibbs-Thomson coefficient and k is the solute partitioning coefficient. The solutal undercooling is assumed to be the driving force for the envelope growth. Solutal undercooling is computed on the basis of a difference between solute concentration on the envelope $c^{*\prime}$, and far form it c_∞. The solute concentration far from the envelope is taken from the cell that is adjacent to its corresponding hyperplane (as shown in Figure 2).

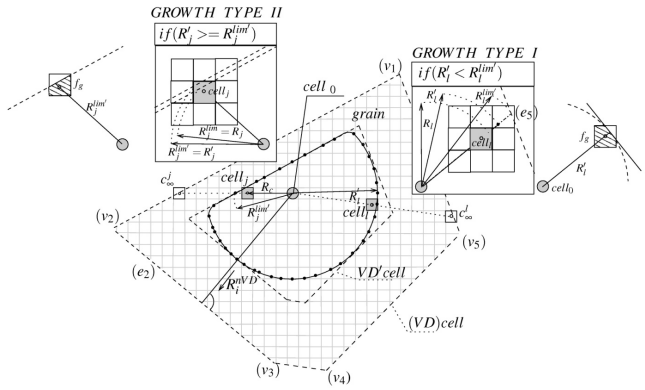

Fig. 2. Schematic presentation of the growth algorithm of the dendritic grains' envelopes. The f_g-function have been introduced into the model in order to make smooth changes between states while keeping relatively large size of CA-cells without sacrificing accuracy

To ensure that the growing envelope is convex, we restrain the local envelope radius by a limiting radius associated with the limiting lines constructed for each hyperplane of VD cell. Every limiting line is defined by two parameters: a slope that is identical to the slope of the corresponding hyperplanes, and by a radius defined as a distance from the grain center, measured along the line segment bounded by the two centers of two neighboring grains. The slope is constant during the growth process, however the radius is updated in every time step, in the same manner as the envelope radius, and according to the growth law (Eq. 1).

The relation between values of the two envelope radii: the local envelope radius and the limiting envelope radius, allows to differentiate between two types of growth (as shown in Figure 2). When the local envelope radius is smaller than the limiting radius then the Type-I growth is applied, otherwise the Type-II growth is used. The type-II growth restrains the local envelope radius by the value of limiting radius, which guarantees convexity of the dendritic grain envelope as describe previously. In turn, the Type-I growth is typical for unrestrained growth. At the beginning of the growth process, the the Type-I growth is dominant. However, when the solute layers of the neighboring grains overlap, the Type-II growth becomes more important.

We observed that the model is sensitive to the size of the cell. To solve this problem we introduced additional parameter which is grain fraction of the cell. It is defined as a cell area fraction occupied by the grain (see Figure 2). Such parameter allows for smooth changes between states, while keeping relatively large size of CA-cell without sacrificing the accuracy.

2.2 Approximation of the primary dendrite shape

We come now to the second subprocess of the dendritic grain growth that corresponds to the growth of the primary solid phase crystals within envelopes. As it was mentioned before, the

increase of solid phase in every time step is known, and it is computed on the basis of solute and heat balances. However, the challenge is to propose the algorithm to approximate the spread of the solid phase within the determined envelope. The task is hard because the shape of dendritic crystal is usually very complex. First of all, the dendritic crystal can be characterized by different order arms. Secondly, in the presented model a random sites of nucleation are assumed, and consequently resulting grains have irregular shapes, i.e. the main arms can have different length. Lastly, the grain can have random orientation.

To address the above requirements we utilized the analytical model of dendrite shape given by Equation 2 (see [15] for more details). In the proposed model only main arms are taken into account, and each l-th main arm has length L_l. Thanks to this, irregular shapes, with various main arms lengths, can be described. The second parameter that describes dendrite is shape parameter A_l. It can vary from 0 to 1, which translates into shapes from spherical to shapes with highly distinctive main arms, respectively. The model is described by the following formula:

$$R(\Theta)=\sum_{l=0}^{3} L_l \cdot Z \cdot Q \tag{2}$$

$$Z=\left(1-A_l \cdot \left|\sin(2\Theta)\right|\right)$$

$$Q=\left(H\left(\Theta-\left(\frac{\pi}{4}(2l-1)\right)\right)-H\left(\Theta-\left(\frac{\pi}{4}(2l+1)\right)\right)\right)$$

Where R and θ are radial and angular coordinates, respectively and function H is the Heaviside function. In Figure 3, example shape is given. One important feature of this model is that random orientation of the grain can be introduced by rotating the dendritic shape by the angle corresponding to the random orientation. Usually, random orientation is defined as a angle between main arm and the x-axis.

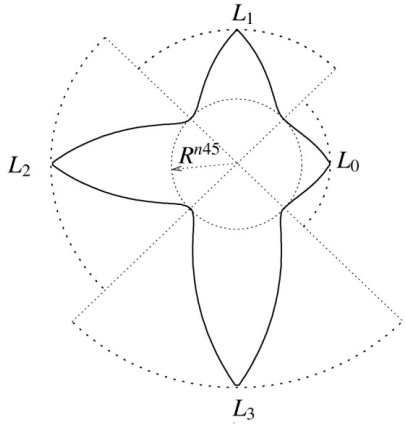

Fig. 3. Analytical shape model employed to approximate the shape of primary dendrites. In Figure the additional parameter R^{n45} is shown. This auxiliary parameter binds shape parameters A_l such that for each l, $L_l \cdot (1 - A_l) = R^{n45}$

In our problem the length of the main arms depends on local conditions, and it is known in every time step. The shape parameters depend on the internal solid fractions ($f_i = f_s/f_g$), and are known in every time step as well. Furthermore, we assume that there is no diffusion in the solid phase. It means that the distribution of the solid phase does not influence intermediate results and the procedure to determine the solid phase distribution can be called only once when the eutectic temperature is reached.

The procedure to determine the solid phase distribution consists of a few steps and is applied to each dendritic grain. Firstly, we choose grain orientation among 49 preferential directions [8] and find four cells that correspond to four main tips of the dendrite. The envelope radii that are assigned to those cells become the length of the main arms L_l. Then, we compute the area of the grain S_g by summing up grain fractions in the cells attached to this grain (as a result of engulfing by the growing envelope). Given grain fraction and internal fraction of the considered grain we can compute the area of solid phase, S_s. We call this value an exact value of the solid phase area. At this point the only missing parameter of the shape model is the shape parameter A_l. Because we utilize the analytical shape model, for every given shape parameter the area of such solid phase can be computed. Thus, applying the reverse procedure, given the area of the solid phase, the corresponding shape parameter corresponding to it can be found easily. Given all parameters of the shape model we can proceed to the last step. For every cell added to the grain we check if it belongs to the primary solid phase area. From equation (2) the corresponding radius and tangential to the dendrite are computed. Thus we can check if given cell is occupied by the solid phase entirely. This can be done by computing the solid fraction of the cell f_s. If the fraction is greater than zero the cell remains in the dendrite grain, otherwise its state is changed into liquid. Solid fractions of all cells are accumulated and the sum is compared with corresponding area of the solid phase S_s. When the difference is greater than 1% of the exact solid phase area assigned to the grain, an iteration procedure to determine the solid phase distribution is applied in order to decrease resulting error (in practice few iterations are sufficient).

3. Nucleation and growth of eutectic grains

Let us now proceed to the second stage of the hypoeutectic alloys solidification. One important element of the presented two-stage solidification model is its ability to capture two different eutectic transformation modes. The main difference between those modes is in nucleation process, while the grain growth is identical. Similarly to the first stage, we consider two phenomena that affect the structure formation, i.e. the nucleation and the growth.

We begin with the nucleation model. In our approach we use instantaneous nucleation. Similarly to the dendritic nucleation, all eutectic grains nucleate in the same time step. However, depending on which eutectic transformation mode is applied, we utilize different nucleation densities, and we define differently the set of cells that can become the nucleus of the eutectic grains. In case of modified alloys, nuclei of eutectic grains are randomly

chosen among all cells that are in liquid state. In case of unmodified alloys, nuclei of eutectic grains are chosen among liquid cells from the set of cells that are in the neighborhood of the cells that belong to the dendritic grain.

On the other hand, to model the growth of eutectic grains we use the approach based on the cellular automata presented in [7]. In the model the eutectic grain is approximated by a sphere. The radius of eutectic envelope R_e is computed according to the formula:

$$\frac{dR_g}{dt} = \mu \cdot (\Delta T)^2 \tag{3}$$

where μ is coefficient of growth, and ΔT is undercooling, computed with the respect to the eutectic temperature. In every time step, temperature is computed on the basis of heat balance. More details can be found in [7], here we limit further consideration only to the parts that we extended.

Although the model allows to simulate the eutectic structure formation in hypoeutectic alloys, we must introduce one extension into the base model, that deals with the contact of eutectic grains with dendritic grains. In the second stage of the solidification process, dendritic grains and other eutectic grains form a natural barrier for eutectic growth. If a cell from the neighborhood of growing cell is not in the liquid state (because it belongs to the dendritic grain or other eutectic grains), it is not considered during the growth process. As a result impingement between different types of grains is naturally included into the model. However, when considering the two stage solidification we must take one additional case into consideration. Growing eutectic grain that encounters the dendritic grain, is blocked initially by it in the contact area. Later, when the eutectic grain continues to grow in all other directions it may happen that the eutectic grain will be larger than the dendritic grain. Consequently the dendritic grain will be no longer a barrier that limits eutectic growth, and eutectic grains regain the ability to overgrow the dendritic grain (see Figure 4). If this happen the curvature radius of the eutectic envelope changes, and two problems must be solved. The first one is how to detect this type of contact, and the second one is how to determine new curvature radius.

The change in curvature radius may occur when the cell is engulfed by the growing eutectic envelope and when the neighborhood of such cell consists of at least one cell in some state corresponding to the dendritic structure. To detect such situation the direction of two vectors, $\vec{n}^{n}_{ngbr}$ and $\vec{n}^{loc}_{ngbr}$, are compared. The first vector, $\vec{n}^{n}_{ngbr}$, is a normal vector to the envelope segment assigned to parent cell, and it has the same slope as the line that connects center of the k-th cell and the center of the circle assigned to its parent cell, C_j (see Figure 4). The second vector, $\vec{n}^{loc}_{ngbr}$, is a normal vector to the envelope but it is computed on the basis of local fraction of the eutectic grain within neighborhood of the $cell_k$ as proposed in [16]. If the angle between two vectors is greater than some limiting value (we assume 5°), then a new radius of curvature must be determined and assigned to the growing cell, that has been engulfed by the eutectic grain. New circle center is determined, as shown in

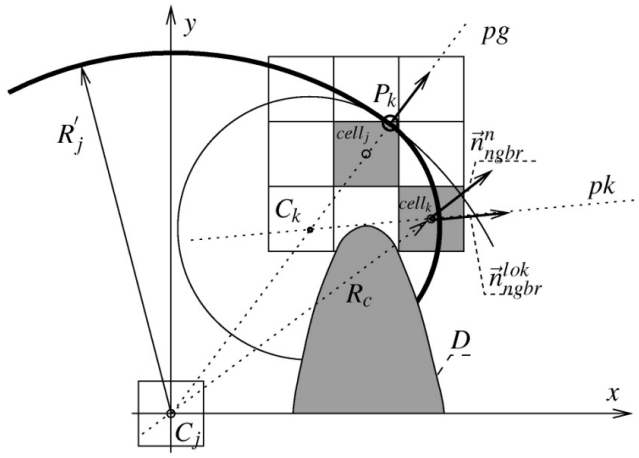

Fig. 4. Contact of growing eutectic grain with center in C_j with dendritic grain *D*

Figure 4, which is intersection of the two lines pg and pk. Line pg is defined by the center of the parent cell (j-th cell) and the slope of a normal to the envelope of the parent cell. Line pk is defined by the slope of vector $\vec{n}^{loc}_{ngbr}$ and the center of the k-th cell. The length of new radius of curvature is the difference between previous radius and the distance of segment $|C_jC_k|$. This procedure guarantees that the two circles are internally tangent and the envelope is continuous in point P_k.

4. Conclusion

In a paper model of two-stage solidification have been proposed. The model exploits flexibility and extensibility of the cellular automata technique. New algorithms to simulate multigrain growth structure in hypoeutectic alloys have been developed, and implemented. Novel solutions include the growth algorithm of dendritic envelopes, approximation of the primary dendrite shape and the growth algorithm of eutectic structure that takes into account contact with dendritic grains. In addition, Voronoi Diagram has been exploited in dendritic envelope grain growth algorithm to tackle the contact of diffusion layers of neighboring grains.

The model has been verified, which we report in Part II of this paper. Computer simulations have been executed, and obtained results are further discussed in Part II of the paper. What is important our model allows to determine the temperature, the size of the structure constituents (of both the primary solid phase and eutectics) and the arrangement of individual dendritic and eutectic grains in the consecutive stages of the solidification process. This result is especially interesting because gradual topological transition of microstructure, mostly in the last stage of solidification, is directly related to the defects formation in castings.

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