A cellular automaton approach for the prediction of grain size in grain refined alloys

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Abstract. Grain refinement by inoculation is widely practised in metal casting, in particular for aluminium alloys. During the last decades several modelling techniques have emerged to quantitatively predict solidification microstructures. The prediction of grain size remains however a challenge due to the complex competition between nucleation and growth, and physical phenomena taking place at very different length scales. Models often address the thermal recalescence and sometimes the suppression of nucleation by the solute boundary layers. The possible clustering tendency of the nucleant particles is however usually not considered, although it can play an important influence on the final grain size. This effect is particularly difficult to quantify because it requires a small-scale approach to describe the clusters, while a large computational domain is needed for grain size statistics. A cellular automaton (CA) model tracking the grain envelopes was developed to address this challenge. The spatial distribution of the nucleant particles and the solute diffusion field in the intergranular liquid were represented directly on the CA grid. Predicted grain sizes were first compared successfully with published experimental and modelling data. An investigation of the role of clustering of nucleant particles was then carried out, which demonstrated the potential of the model to address this topic.

1. Introduction
Grain refinement by inoculation is widely practised in metal casting, in particular for aluminium alloys. The goal is to obtain small equiaxed grains which bring several benefits such as reduced defects, easier processing and improved mechanical properties. The added particles favour the nucleation of many equiaxed grains at low undercooling, avoiding also the formation of undesired columnar structures. During the last decades there has been great interest to quantitatively predict solidification microstructures. Several modelling techniques have emerged such as multiphase-models \cite{1-3} and cellular automata \cite{4} which can provide fields of various microstructural quantities on the scale of an entire cast component. On a smaller scale, the phase-field method can now address the growth of columnar or equiaxed grains with a very high level of details about the solid morphology and the distribution of the solute elements in the microstructure including the effect of fluid flow \cite{5,6}.

In spite of this progress, the prediction of grain size remains a modelling challenge. The reason is that the average grain size in a microstructure is the result of a complex competition between nucleation and growth and is governed by phenomena taking place at very different length scales. A major progress in the understanding of grain refinement and quantification of the grain size has been the free growth theory of Greer \cite{7}. One of the outcomes of the theory is that the particle size distribution of the inoculant can be translated into a distribution of nucleation undercoolings. Combining this with a proper growth model, the grain size can be predicted \cite{8}. Models predicting the grain size normally also include a calculation of a thermal recalescence, which will stop nucleation, leaving the less potent particles...
unused. Recently, Du et al. developed a mean-field model that also applies to continuous cooling conditions, where no recalescence occurs and nucleation can only be stopped by site capture or solutal effects [9]. The problem was addressed by incorporating the effect of the solute boundary layers of the growing grains, which can stifle nucleation on less potent particles in their vicinity. This phenomenon, known also as the solute suppressed nucleation zone (SSNZ), was well described already earlier by Easton and StJohn [10].

Another factor that can influence the grain size is the spatial distribution of the nucleant particles. If they tend to agglomerate, a large proportion of them will be inefficient due to the SSNZ effect or simple capture. This situation is particularly difficult to quantify in a model. It requires on one hand a small-scale approach to describe the geometrical arrangement of the clusters and to address the SSNZ formed by the overlapping diffusion fields inside the clusters and in their vicinity. On the other hand, the computational domain needs to be sufficiently large to allow for sufficient statistics on grain sizes. In addition, to correctly calculate the recalescence, the approach should address heat transfer, either by a direct resolution of the heat conservation equation or through a micro-macro coupling scheme. The model presented here addresses these challenges by incorporating all the important mechanisms that determine the final grain size: the size and spatial distribution of nucleant particles, the solute diffusion field around growing grains, thermal recalescence and site capture. Although it is not fully coupled yet, the model has been designed to be compatible with a resolution of heat flow on the process scale.

2. Model description

Predicting the grain size using a direct representation of the individual grains requires a relatively large computation domain to have enough grains to carry out meaningful statistics. For grain refined alloys, computation domains must typically be in the order of 1 mm to correctly predict the grain size in a given region of the casting. While state-of-the art implementations of the phase-field method [5] would probably allow to address such volumes, the computation cost of the technique remains very high, and prevents any implementation in numerical schemes where such local microstructure calculations would be coupled with heat transfer on the process scale. Another choice would be mean-field approaches, which have been applied with success [9]. A direct description of the grains is however desired when non-random spatial distributions of nucleant particles are analysed, including the complex multiple interactions through their diffusion fields. For these reasons, an envelope model was considered. In this approach, only the grain envelope is directly represented whilst the internal solid morphology is simplified.

2.1. Nucleation and growth

The model is based on the CAFE model of ProCAST, originally developed at EPFL [4]. The model was recently renovated to support parallel computations (and renamed CAFE2G), before being extended for the current study. The model uses a grid of cells to track the nucleation and growth of grains nucleating either at the mould surface or in the bulk. Nucleation is assumed to be heterogeneous and athermal. The number of nucleated grains is obtained by integration of a distribution of nucleation undercooling, using either a Gaussian distribution function, as in the original model of [4], or a Log-normal function:

\[ D(\Delta T) = \frac{dn}{d(\Delta T)} = \frac{n_0}{\sigma \Delta T_0 2\pi} \exp \left[ -\frac{1}{2} \left( \frac{\Delta T}{\sigma \Delta T_0} \right)^2 \right] \]  

(1)

where \( \Delta T \) is the undercooling, \( \Delta T_0 \) is the median of the distribution, \( \sigma \) is a dimensionless parameter and \( n_0 \) is the total number of sites per unit volume.

During the resolution the local undercooling is calculated based on the local concentration \( c_l \):

\[ \Delta T = T_m + m_l c_l - T \]  

(2)

where \( T_m \) is the melting temperature of the pure metal and \( m_l \) is the liquidus slope.

Grains can be either dendritic or globular. For dendritic grains, only the grain envelopes are tracked using a dendrite tip kinetic model based on the classical LGK theory [11].
Intergranular liquid is obtained by solving numerically the solute\(\phi\) \((\text{eq. } 4)\):
\[\nabla \cdot D_c \nabla c_f = \nabla (\nabla \cdot D_c \nabla c_f)\]
where \(c_f\) is the far-field concentration obtained by averaging \(c_t\) over the liquid cells situated in a region centred on the growing cell. The size of that region is a parameter of the model. For isothermal calculations and small domains, \(c_f\), is simply obtained by averaging \(c_t\) over all the liquid cells. \(\text{Iv}^{-1}\) is the inverse Ivantsov function, which is approximated by the following expression [12]:
\[\text{Iv}^{-1}(\Omega) = 0.4567 \left(\frac{\Omega}{1+\Omega}\right)^{1.195}\]
The envelopes are tracked on the CA grid following the same procedure as in the original model of [4]. The growth rate of globular grains is obtained with a Zener type model, \(v = D\Omega/R\), where \(R\) is the radius of the globular grain, as proposed by Wu et al. [13].

### 2.2. Solute distribution

In the original CAFE model, the far-field concentrations are assumed to stay at the nominal concentration, and the grain envelopes can only be stopped by mechanical contact with other grains or the border. The model was extended to include the influence of the solute diffusion field in the intergranular liquid associated with solid growth. The solute boundary layers can affect the undercooling at the remaining nucleation sites and the growth rate of neighbouring grains (so-called soft impingement).

The distribution of solute in the intergranular liquid is obtained by solving numerically the solute conservation equation:
\[
\frac{\partial c_f}{\partial t} = \nabla \cdot D_c \nabla c_f
\]

The resolution is performed on the CA grid using a finite volume method and a forward Euler time-discretization scheme. The resolution is restricted to the liquid cells of the CA grid, imposing \(c_f^\ast\) as a boundary conditions at the grain envelopes. Numerically this is achieved by prescribing \(c_f^\ast\) on the cells containing a section of a grain envelope at each resolution step before solving the diffusion problem.

### 2.3. Thermal coupling

To correctly predict the nucleation arrest, the model must be coupled with a resolution of heat transfer and predict a possible recalescence. The model can be coupled with a finite element resolution of the heat transfer on the process scales as described in the original article [4]. However, owing to the relatively small domain sizes considered in the present study \((1-10\ \text{mm}^3)\), a simplified version of thermal coupling was used, which considers isothermal solidification conditions (no thermal gradients). Two options were implemented regarding the calculation of the temperature. The first one is to consider the temperature as an input, which is a suitable assumption for Bridgman-type conditions. The second option is to obtain the temperature from a global heat balance using the solid fraction calculated with the CA method and a microsegregation model. More specifically, the cooling rate is obtained from:
\[
\frac{dT}{dt} = \frac{1}{\rho C_p} \left[ L \frac{dg_s}{dt} - \dot{H} \right]
\]
where \(\dot{H}\) is a prescribed heat extraction rate per unit volume, \(\rho C_p\) is the volumetric specific heat and \(L\) is the volumetric latent heat.

The solid fraction, \(g_s\), is the volume fraction of solid in the calculation domain. It is obtained from the volume fraction occupied by the grains, \(g_g\), and a model for the internal solid fraction. Whilst the latter is simply the unity for globular grains, a microsegregation model is required for dendritic grains.
Lever rule and Scheil approximations were implemented. A set of non-linear equations is finally obtained, and iterations should in principle be carried out to determine the temperature to be used for the CA resolution. As this procedure would be too computationally expensive a linearization technique is used to avoid iterating on the CA resolution.

2.4. Initialization and algorithms
During initialization, the nucleant particles are randomly distributed in space and receive random nucleation undercoolings with a probability determined by the Gaussian or log-normal distribution (equation (1)). All cells are initialized at the liquid state and the temperature is usually set at the liquidus temperature of the alloy. The liquid concentration is initialized at the nominal concentration of the alloy, $c_i = c_{i_0}$ in all cells. The time-step is taken as half of the maximum value given by the Fourier stability criterion for explicit time discretization schemes.

Nucleation arrest usually occurs at a very low solid volume fraction (typically between 0.01% and 0.1%). Hence, the calculation of the grain and solid fractions can be quite inaccurate if only based on the number of captured CA cells, because at this stage grain radii are still very small in comparison with the cell size. For a better accuracy, the contribution of the small grains to the average grain fraction is obtained analytically from the size of their growing sphere/octahedron, until they can be discretized with sufficient accuracy on the CA grid. The grain envelope is then no longer described with a single octahedron or sphere centered on the nucleation sites, but by a series of off-centered shapes. A cell grain fraction is calculated in the cells that contain a section of the grain envelope. The calculation is based on the position of the grain envelope with respect to the cell centre. This method offers a better accuracy than simply considering a binary state (captured/not captured) as it is often done in CA methods.

3. Results and discussion

3.1. Model validation
A potential difficulty with CA methods is to make them quantitative and guarantee a correct accuracy of the resolution. To address this difficulty, the model was first validated against the well-established models of Rappaz-Thévoz [14] and Wang-Beckermann [1]. As in [14] and [1], the model was applied to the solidification of an Al-5wt%Si alloy, considering a single dendritic grain in a spherical domain and a cooling rate at liquidus of 45 K/s. The model was applied to three different final grain radii: 0.1, 1 and 10 mm, using the physical properties listed in [1]. As can be seen in figure 1, a very good agreement is obtained between the present model and the results of [1] and [14]. The results are also compared with the original CAFE model where solute diffusion is not addressed and grain growth is only stopped by the domain border or hard impingement. The better agreement obtained with the present model than the original one already shows the benefits of including solute diffusion into CA solidification models.

3.2. Comparison with reference X-ray radiography measurements and mean-field models
A second series of comparisons was made with the excellent investigation done at Sintef, where data have been collected by in-situ microfocus X-radiography during near-isothermal melt solidification experiments [9]. Constant cooling rates and suppressed melt convection conditions were almost achieved, which provides ideal conditions for comparisons. Data were obtained for an Al-10%Cu alloy solidified at different cooling rates without recalescence.

The present CA model was applied to the same conditions and same nucleation parameters as the mean-field model presented in [9] for 0.1 wt% Al-5Ti-1B: $d_0 = 0.77 \, \mu m$, $\sigma = 0.5$, $\eta_0 = 5 \times 10^{11} \, m^3$. The calculations were carried out in a cubic domain of 1.5 mm in size, assuming a globular grain morphology.

Figure 2 shows the concentration field in the liquid at the time of nucleation arrest for different cooling rates. The concentration at the interface is higher when the cooling rate is high, which is associated with a higher undercooling at nucleation arrest.

In figure 3, the grain size and maximum nucleation undercooling are compared with the experimental and modelling data from [9]. The average grain size calculated with the CA model shows very good
agreement with the experimental and mean-field results in figure 3(a). Interestingly, the CA model is slightly closer to the experimental data than the mean-field approach. This improvement is more visible in figure 3(b) where a lower maximum nucleation undercooling in substantially better experimental agreement is obtained with the CA method than mean-field. The difference can be explained in terms of overlapping diffusion fields. The CA model can account for the cumulative effect of several neighbouring grains which can stifle less potent nucleant particles in their vicinity. This effect cannot be considered in mean-field approaches where grains and nucleants are not localized.

Figure 1. Comparison of the CA model with reference results for a single dendritic grain.

Figure 2. Concentration in the intergranular liquid at nucleation arrest during globular solidification in Al-10wt%Cu at (a) 0.05, (b) 0.2 and (c) 1 K/s. Solid has the same colour as liquid at the interface.

Figure 3. Predicted (a) grain sizes and (b) maximum nucleation undercoolings compared with [9].

3.3. Role of the solute suppressed nucleation zone
The influence of the solute diffusion field on nucleation arrest was analysed. Figure 4 shows the proportion of nucleated particles for different size classes or nucleation potencies. The grey bars simply correspond to the log normal size distribution of nucleation undercoolings introduced in the simulation. As expected, a large proportion of the particles in the most potent classes initiate grains whilst none of the less potent particles are activated due to nucleation arrest occurring at a lower undercooling. The red bars indicate the proportion of particles that had reached their nominal nucleation temperature before
they were physically captured by the growing solid but were prevented from initiating a grain due to the local liquid concentration. This data shows the role of the solute suppressed nucleation zone, which is quite substantial under these conditions.

Similar calculations were carried out using a constant heat extraction rate rather than constant cooling, solving equation (7) to calculate the cooling curve. As can be seen in figure 5, a recalescence is obtained, which is deeper as the heat extraction rate (or cooling rate at liquidus indicated in the diagram) increases.

Figure 6 shows that the proportion of sites affected by the solute boundary layers is much lower than in the case of constant cooling. This is obviously due to the recalescence which is now the dominant mechanism for the nucleation arrest. The solute suppressed nucleation mechanism seems to be the most prominent at intermediate cooling rates. This can be understood by considering that slow cooling leads to distant grains and recalescence at low undercooling, whilst fast cooling leads to thin boundary layers.
3.4. Introducing clustering
The clustering tendency of the nucleant particles can be an important factor affecting the effectiveness of grain refinement. This phenomenon was investigated with the CA model by introducing nucleant particles according to a non-uniform probability in space. This was achieved by considering clusters of nucleant particles characterized by two parameters: an average number of particles per cluster, $N_{cl}$, and a spreading parameter, $K$. The latter determines the average cluster radius, $R_{cl}$, obtained from the following expression: $4\pi n_0 R_{cl}^3/(3 N_{cl}) = K$. The nucleant particles were then attributed randomly to the clusters with a random position within the average cluster radius.

Calculations were carried out with $K=0.1$, 0.03, 0.01, 0.003, which corresponds to an increasing clustering tendency. An example of spatial distribution of nucleation sites is shown in figure 7. Figure 8 shows the cooling curves for the different conditions. As anticipated, recalescence occurs at higher undercooling as the clustering tendency increases. This is explained by an increasingly important effect of the solute suppressed nucleation zone. This was confirmed by the quantification of the proportion of nucleant particles in the SSNZ, which is shown in figure 9.

4. Conclusion
A cellular automaton model based on the new CAFE2G module of ProCAST has been developed to address the challenge of predicting the grain size in grain refined alloys without using adjustable nucleation parameters. The model incorporates all the important mechanisms affecting the final grain size: potency of the nucleant particles, competition between nucleation and growth, recalescence, the solute suppressed nucleation zone and the spatial distribution of the nucleant particles. The model has been validated against well-established mean-field models and shows very good agreement with experimental data. A first investigation of the role of clustering of the nucleant particles was carried out to demonstrate the potential of the model to address this topic in a quantitative manner.
Figure 7. Spatial distribution of nucleant particles considering a clustering tendency ($K=0.01$).

Figure 8. Cooling curves for different clustering tendencies.

Figure 9. Proportion of nucleant particles in the SSNZ for different clustering tendencies.

Acknowledgments
The author wishes to thank Prof. Z. Fan from BCAST for the inspiring discussions on grain refinement and the useful suggestions.

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