Prediction of as-cast grain size of inoculated aluminum alloys melt solidified under non-isothermal conditions

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Abstract. In this paper, a multi-scale as-cast grain size prediction model is proposed to predict as-cast grain size of inoculated aluminum alloys melt solidified under non-isothermal condition, i.e., the existence of temperature gradient. Given melt composition, inoculation and heat extraction boundary conditions, the model is able to predict maximum nucleation undercooling, cooling curve, primary phase solidification path and final as-cast grain size of binary alloys. The proposed model has been applied to two Al-Mg alloys, and comparison with laboratory and industrial solidification experimental results have been carried out. The preliminary conclusion is that the proposed model is a promising suitable microscopic model used within the multi-scale casting simulation modelling framework.

1. Introduction
Prediction of as-cast grain size of inoculated aluminum alloys have been an active research topic in the past 15 years since the development of the "Free Growth" nucleation model by Greer et al [1-7]. Most of the recent numerical grain size prediction models have followed the approach pioneered by Maxwell and Hellawell (hereafter referred to as MHN model) [8], in which the continuous solid grain size distribution is described by discrete size classes. The influences of solute content, the type of grain refiners, inoculant particle size distribution and cooling rate have been taken into account. While it is clear that with given alloy composition, cooling and inoculation conditions quantitative predictions of the final as-cast grain size could be made even for multicomponent alloys, a gap still exists to apply the MHN-type model to predict grain structure at the scale of a typical casting ingot.

The prediction of grain structure at the scale of casting component would require a multi-scale model to address both of macroscopic heat and solute transportation, melt flow, solid grain floating and microscopic grain nucleation and growth. Cellular Automaton Finite Element (CAFE) method, originally proposed by Gandin and Rappaz in [9] with the most recent development reported in [10], is a popular approach for this purpose. Indeed CAFE had been applied by Greer and his co-workers [11, 12] to predict as-cast grain size of inoculated aluminium alloys. Although these application successfully illustrated the trends in the observed variation of grain diameter with refiner addition level and cooling conditions [11, 12], the limitations of CAFE modelling in predicting the

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² It should be noted that the size class subdivision concept in the MHN model is identical to the Kampmann-Wagner Numerical (KWN) model used in precipitation kinetics modelling.
constitutional undercooling was also revealed. As stated by Quested in [12], the CAFE model's limitations originate from its assumption that dendritic growth occurs under steady state and dendrite growth rates are equal to those in isolated dendritic arrays. This assumption means that the interactions among the growing grains (i.e., soft impingement) have not been taken into account. To overcome the limitations, it requires the determinations of both solute and temperature fields and their interdependence in the melt. It should be noted that the temperature field vary at the scale of the casting component (~0.1-1 m) while the solute at the scale of grain size (~10-100 µm) due to the huge disparity between thermal and solutal diffusion coefficients.

In this work, we are aiming to develop such a multi-scale model by integrating a MHN-type microscopic model (solute field) into macroscopic heat diffusion equation (temperature field). To do so the most recent CALPHAD-coupled multi-component as-cast grain size prediction model [1] is extended to cover the full primary phase solidification range (instead of the initial solidification stage). Then this microscopic model is applied to compute the release of the latent heat in all the finite volume elements used in solving the macroscopic heat transfer equation. Meanwhile the numerical solution of the heat diffusion equation gives a thermal history of each volume element, which is a required input to the microscopic model to compute the volume element's solidification path. To achieve the cross-scale coupling numerically the splitting technique, borrowed from combustion modelling [13] and successfully applied for homogenization heat treatment modelling [14], is adopted.

The paper is organized as follows: Section 2 is devoted to model description and Section 3 to results and discussions. Conclusions are given in Section 4.

2. Model description

The proposed multi-scale grain size prediction model consists of a macroscopic heat transfer equation with a latent heat release source term and a microscopic model describing grain nucleation and growth.

\[
\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (D \nabla T) + L \frac{df_s}{dt} \tag{Eq.1}
\]

T is temperature, L is the volumetric latent heat, \(f_s\) is solid volume fraction, \(\rho, C_p\) and \(D\) are the weighted density, specific heat capacity and diffusivity, which are expressed by

\[
\rho = f_s \rho^L + f_s \rho^S
\]

\[
C_p = f_s C_p^L + f_s C_p^S
\]

\[
D_T = f_s D_T^L + f_s D_T^S
\]

\(\rho^L, \rho^S\) are the density of liquid and solid phase. \(C_p^L, C_p^S\) are the specific heat of liquid and solid phase. \(D_T^L, D_T^S\) are the thermal conductivity of liquid and solid phase.

An explicit Finite Volume Method is employed to solve the heat transfer equation at the scale of the whole ingot. The calculation of the changing rate of solid fraction is done by the microscopic MHN-type model. This model is applied to each volume element, and able to track grain nucleation and growth. It is an extension of the one reported in [1] to the full solidification range. The next subsection is devoted to the microscopic model.

2.1. The microscopic as-cast grain size prediction model
The microscopic model adopted in this multi-scale framework is based on the as-cast grain size prediction model reported in [1]. The main assumptions adopted in the extended model include:

I. Solid grains are of spherical shape and their growth/dissolution is solely controlled by diffusion in the melt.

II. Local equilibrium modified by the Gibbs-Thomson effect prevails at the solid-liquid interface; thermal and kinetic undercoolings are neglected.

III. The diffusion field surrounding each growing solid grain is at quasi-steady state.

The model consists of a nucleation model and a growth model. Below a brief description of the nucleation model is given, which is followed by the growth model with a focus on their modification to cover the full solidification range.

Greer’s free growth model [1] is adopted to describe nucleation on inoculant particles. According to this model, the number of growing grains is determined by a free-growth condition. For an undercooling of $\Delta T$, the critical diameter, $d_c$, of refiner particle to initiate a free growth grain is given by:

$$d_c = \frac{4\sigma}{\Delta S_V \Delta T}$$  \hspace{1cm} (Eq. 3)

where $\sigma$ is the interfacial energy, $\Delta S_V$ entropy of fusion. All the particles with diameter above this critical diameter are active. One of the key input parameters to the free growth model is inoculation particle size distribution density function, which has been obtained by fitting to the experimentally measured distribution [15] as:

$$PSD(d) = N_V \exp\left(-\frac{d}{d_0}\right)$$  \hspace{1cm} (Eq. 4)

where $d$ is the diameter of disc-shaped inoculant particle, $d_0$ is the characteristic width of the distribution ( $d_0 = 0.72 \mu m$ ) and $N_V$ is the total number of TiB$_2$ particles per unit volume of aluminum melt when Al-5Ti-1B grain refiners are added[1]. $N_V$ could be calculated according to the following empirical rule [15]: for 1 Part Per Thousand (PPT) by weight Al-5Ti-1B addition, $N_V$ is equal to $1.15 \times 10^{12} m^{-3}$. Via Eq. (3) and Eq.(4) nucleation rate could be calculated for a given undercooling.

The growth model consists of four group of equations as described in [1]. The first set of equations is derived from solute conservation law accounting for the melt enrichment/depletion resulted from the growth of solid grains. The second equation is derived from heat balance calculating the local temperature change induced by heat extraction and latent heat release. The third and fourth sets of equations are applicable to each discrete size class; the former comes from the interfacial local equilibrium assumption (modified by the Gibbs-Thomson effect) and the latter are multi-component growth rate equations. The growth rate equations still follow the ones used in [1], i.e.:

$$\dot{R}_j = \frac{D_i \Omega_i}{R_j}$$  \hspace{1cm} (Eq. 5)

$D_i$ and $\Omega_i$ are the diffusivity and dimensionless super-saturation of alloying component $i$, respectively. However to cover the full solidification range, the concept in the standard Avrami analysis that the fraction of the real volume increment during each time increment is proportional to the volume fraction of untransformed is employed. Therefore

$$d f_s^{Real} = d f_s (1 - f_s)$$  \hspace{1cm} (Eq. 6)

The treatment of secondary phase formation is still in progress.

2.2. The direct coupling of the microscopic microstructure evolution with macroscopic heat transfer
While the FVM-based algorithm is used to solve the heat transfer equation (Eq.1) across the casting component scale, the microscopic model introduced in Section 2.1 is applied to each volume element of the 1D domain to capture the solidification kinetics resulting from the variation in local thermal history. These two models are tightly coupled and involve two length scales. This coupling is mathematically analogous to the one in homogenization heat treatment modeling reported in [16]. The splitting method adopted in [16] is employed in this paper to deal with the coupling. The interested readers are referred to Ref. [16] and the references therein for the complete details.

3. Results and discussions

In this section, the proposed model will be applied to both of laboratory scale and industrial casting experiments. The model predictions are going to be examined in the light of the experimental results. The model's input parameters are listed in Table 1, and they are taken from Ref. [12]. The simulation domain is in 1D with adiabatic heat boundary condition on one side. On the other side, the two-step heat-transfer coefficients identified in [12], i.e. 707 W m\(^{-2}\) K\(^{-1}\) for the first 25 seconds and 9000 W m\(^{-2}\) K\(^{-1}\) for the rest, is employed to mimic DC casting heat extraction condition. Grain refiner addition level, alloy composition and the simulated domain geometry are illustrated within in the text below.

| Property                          | Value                        |
|----------------------------------|------------------------------|
| Interfacial energy, \(\sigma\)   | 0.158 J/m\(^2\)             |
| Molar volume of liquid and solid phase | 1.0\times10\(^5\) m\(^3\) |
| Entropy of fusion, \(\Delta S_V\) | 1.11\times10\(^6\) J K\(^{-1}\) m\(^{-3}\) |
| Latent heat per unit volume, \(\Delta H_V\) | 9.5\times10\(^6\) J m\(^{-3}\) |
| Heat capacity of melt            | 2.58\times10\(^6\) J K\(^{-1}\) m\(^{-3}\) |
| Heat capacity of solid           | 3.1\times10\(^6\) J K\(^{-1}\) m\(^{-3}\) |
| Thermal conductivity of melt     | 95 W K\(^{-1}\) m\(^{-1}\) |
| Thermal conductivity of solid    | 215 W K\(^{-1}\) m\(^{-1}\) |
| Diffusivity of Mg                | Diffusion constant (m\(^2\)/s) | activation energy (kJ/mole) | Evaluated at 650 °C |
|                                  | 9.90\times10\(^{-5}\)       | 71.6                       | 8.78\times10\(^{-9}\) |
| Linear binary phase diagram data of Mg | Liquidus slope (K/at\%) | Partition coefficient | 0.3 |

3.1. The application of the model to DC casting simulator experiments

The DC casting simulator, described by Quested in [12], is able to reproduce the variation in solidification conditions that occur through the thickness of a full-scale DC ingot. The simulator is composed of a tube furnace containing a slightly tapering steel crucible (with internal diameter of 43.8 mm at top and 40.8 mm at the base) that fits closely to the surrounding insulation of the tube furnace. At the base of the crucible is a removable base plug, which sits above a water jet. During solidification, the top of the crucible is covered with an insulating blanket.
The proposed model has been applied to the casting simulator experiment of Al-3.5Mg-0.2Fe-0.05Si alloy. The 1D simulation domain is 21 cm in length consisting of 105 finite volumes. The multi-component alloy is approximated as binary Al-3.8at%Mg to decrease the computational time. The model’s grain size predictions are plotted together with the measured and predicted (with CAFE model) ones by Quested [12] in Fig. 1. It is clear the proposed model has a better agreement with the measured grain size than the one obtained by CAEF model performed by Quested et al [12].

![Grain size comparison](image)

**Fig. 1** The proposed model predictions for Al-3.5Mg-0.2Fe-0.05Si DC-simulator samples; the predicted and measured grain size by Quested are also reproduced for comparison.

### 3.2. The application of the model to industrial DC casting trial of an AA5182 alloy

In the industrial DC casting trial reported in [17], the melt, with the nominal composition of Al-0.36wt%Fe-4.12wt%Mg-0.22 wt%Mn-0.19wt%Si and grain refiner addition level of 4 PPT, was DC-cast into an ingot of dimensions of 510 mm in thickness and 1880 mm in width. The variation of grain size through the thickness of the ingot was also reported, which is reproduced in Fig. 2. The measurement shows initially grain size increases with increasing distance from the skin of the ingot. At about the distance of 150 mm to the ingot surface (or 105 mm to the ingot center) it reaches a maximum value of 207 µm. Then after it drops down to a local minimum value of 167 µm, it finally rises to 195 µm at the ingot center. It is interesting to note the largest grain sizes do not appear at the center but at a location about 40 mm away from the center. This observation was believed to be due to "the lack of a heat reservoir in the melt" in [17].

The proposed model is able to shed some lights on the mechanisms behind this complex grain size variation with the distance. The 1D simulation domain is 25.4 cm in length consisting of 127 finite volumes. The multi-component alloy is approximated as a binary Al-5.8at%Mg to decrease the computational time. The predicted grain size variation is also plotted in Fig. 2. It is in consistence with the experimental measurement that the largest grains do not appear at the center but at a location about 40 mm away from the center. To explain this observation the predicted cooling curves during the whole nucleation stage for these two positions are plotted in Fig.3. It is surprising to see the
correlation between the predicted grain size and the average cooling rate at the center and the one 40 mm away from the center. Although the former has smaller grain size, its average cooling rate is lower by 0.07K/s. Even the average cooling rates do not correlate with the predicted grain size in the usual manner, the cooling rates at the latter nucleation stage do. Therefore maybe instant cooling rate should be used in grain size-cooling rate correlation relation. Indeed as shown in Fig. 4, at the same temperature, the one with the slower instant cooling rate has a lower grain number density due to the longer time to reach the same temperature, therefore the more solid grain growth solutally stifling more nucleation event.

Fig. 2 The measured and predicted (with binary assumption) variation of grain size through the thickness of the ingot.
Fig. 3  The predicted cooling curves at two selected volume cells in the 1D simulation domain

![Cooling Curves](image)

Fig. 4  The predicted grain number density evolution with temperature at two selected volume cells in the 1D simulation domain

![Grain Number Density](image)

The discrepancy between the experimental measurement and model predictions could be due to the settling of "travelling" crystals, which has not been considered in the performed simulations.

4. Conclusions

A multi-scale modeling framework has been established to predict grain structure at the scale of casting component. The model has been applied to a DC casting simulator experiment and industrial DC casting trials, and the model predictions have been compared with the reported experimental results. The preliminary conclusion is that the MHN-type model is a promising suitable microscopic model used within the multi-scale casting simulation modelling framework. Simulations without adopting the binary assumption are in progress.

5. References

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