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Modeling of the coupling of microstructure and macrosegregation in a direct chill cast Al-Cu billet

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Abstract

The macroscopic multiphase flow and the growth of the solidification microstructures in the mushy zone of a direct chill (DC) casting are closely coupled. These couplings are the key to the understanding of the formation of the macrosegregation and of the nonuniform microstructure of the casting. In the present paper we use a multiphase and multiscale model to provide a fully coupled picture of the links between macrosegregation and microstructure in a DC cast billet. The model describes nucleation from inoculant particles and growth of dendritic and globular equiaxed crystal grains, fully coupled with macroscopic transport phenomena: fluid flow induced by natural convection and solidification shrinkage, heat, mass, and solute mass transport, motion of free-floating equiaxed grains and of grain-refiner particles. We compare our simulations to experiments on grain-refined and non grain-refined industrial size billets from literature. We show that a transition between dendritic and globular grain morphology triggered by the grain refinement is the key to the explanation of the differences between the macrosegregation patterns in the two billets. We further show that the grain size and morphology are strongly affected by the macroscopic transport of free-floating equiaxed grains and of grain-refiner particles.

1. Introduction

Direct chill (DC) casting is the principal process for the production of wrought aluminum alloys. It is used to produce cylindrical billets for extrusion of profiles and rectangular ingots for rolling of plates, strip, and foil. Macrosegregation is a common defect in DC casting and can lead to nonuniform material properties. Since it is generally aggravated by higher casting speeds and casting size, it also limits productivity. Macrosegregation is characterized by variations of the chemical composition on a scale larger than the crystal grains, which cannot be mitigated during the further downstream processing. The redistribution of the chemical species (solutes) at the product scale during solidification is caused by the relative motion of liquid and solid with different compositions. It is generally admitted that the macrosegregation in DC casting is a consequence of an intricate interplay of the melt flow, induced by thermosolutal natural convection, shrinkage, and pouring, and of the transport of solute-lean free-floating grains [1–3]. Melt flow due to stress-induced deformation can also play a role [4]. The importance of the individual mechanisms varies depending on the alloy, casting shape and size and process parameters [3]. It is however difficult to construct a simplified general image of the macrosegregation formation in DC casting, because all these strongly coupled transport phenomena have a significant impact. A thorough understanding of the involved mechanisms and of their coupling is required to be able to develop robust solutions that would

45 mitigate the macrosegregation [5].

46 The interplay of the transport mechanisms that cause macrosegregation is closely linked to the
47 microstructure of the solidifying grains. The grain size and the grain morphology (dendritic or
48 globular) determine the dynamics of settling of the free-floating equiaxed grains, the compactness of
49 the packed grain layer, and its hydrodynamic permeability. They thus affect the macrosegregation
50 induced by the settling of solute-lean grains and the expulsion of solute-rich liquid – the prime cause
51 of the negative centerline segregation. Furthermore, the grain size and morphology influence the
52 macrosegregation formed by permeation of solute-rich liquid through the packed layer. These effects
53 have been clearly demonstrated in steel ingot casting, using a combination of industrial scale
54 experiments and multiscale numerical modeling [6–10]. It was shown that globular free-floating
55 grains lead to much more pronounced settling-induced macrosegregation than dendritic grains. Such
56 detailed modeling has not yet been done for DC casting.

57 Most experimental investigations of the influence of grain motion on macrosegregation in DC casting
58 used varying levels of grain refinement as a control process parameter, in order to stimulate or to
59 stifle grain motion. However, the influence of grain refinement on macrosegregation is complex. On
60 the one hand, grain refinement results into smaller grains that are more easily entrained by the liquid
61 and have smaller slip velocities, thus reducing settling-induced macrosegregation. On the other hand
62 it also promotes more compact globular grain morphologies that settle faster than dendritic grains of
63 the same size and result into a larger fraction of solute-lean solid in the packed grain layer than
64 dendritic grains. Moreover, a modification of grain size and morphology can alter the structure of the
65 multiphase flow in the mushy zone. Due to this complexity, the experimental investigations, which
66 were done with different alloys, casting formats and casting parameters, led to disparate conclusions
67 and interpretations, sparking some controversy. In small 2000 series billets, no significant influence
68 of grain refinement on macrosegregation was observed [11,12]. In large Al-Cu billets Finn et al. [13]
69 observed that the addition of grain refiner led to a reversal from negative to positive centerline
70 segregation, while Vreeman et al. [14] showed an increase of negative segregation. In large 5000-
71 series sheet ingots, grain refinement clearly led to more pronounced negative centerline segregation
72 [15–19]. The grain morphology, more globular in grain-refined castings and more dendritic in non
73 grain-refined castings, was seen as a key factor [17,20]. An impact of the morphology on both the
74 motion of free-floating equiaxed grains and on the permeability of the packed grain layer was argued
75 to be the reason for the modification of the macrosegregation patterns.

76 The support of modeling is vital in the investigation of the physical mechanisms that link the
77 microstructure and the macrosegregation in a complex casting process. However, concurrent coupled
78 simulation of microstructure and the transport phenomena in DC casting is still lacking. Past models
79 were not able to capture these couplings because they used various simplifications. Most models were
80 based on simple equilibrium models of phase change and lacked a description of the kinetics of the
81 microstructure nucleation and growth [21–25]. Models that included grain growth kinetics did not
82 account for grain motion [20] or assumed a globular grain morphology [1,26]. Detailed modeling
83 focusing on grain structure and morphology did not consider grain motion and macrosegregation [27].
84 The success in predicting macrosegregation was limited to globular structures and could not correctly
85 capture the impact of grain morphology and of grain refinement.

86 Recently we presented a more advanced model of DC casting, which accounts for macrosegregation
87 with grain motion, fully coupled with nucleation on grain-refiner particles and with the kinetics of
88 grain growth and morphology development [28,29]. This model was successful in predicting the
89 influence of the flow on the distribution of grain size and the influence of grain refinement on the
90 grain morphology in a large sheet ingot [10,29]. In the present paper we use this model to provide a

91 fully coupled picture of the links between macrosegregation and microstructure in a DC cast billet.
92 We use the experimental results presented by Vreeman et al. on Al-Cu billets [14] as a reference to
93 which we compare our model predictions. The experiments were made on two grain-refined and non
94 grain-refined billets cast under the same conditions. We perform a detailed comparison of the
95 simulations to the experiments and a critical analysis of the coupled phenomena of macroscopic
96 transport and microstructure formation. We show that a morphology transition triggered by the grain
97 refinement is the key to the explanation of the differences of the macrosegregation patterns in the two
98 billets and we show that the microstructure is strongly affected by the macroscopic transport of free-
99 floating equiaxed grains and of grain-refiner particles.

100

101 2. Model

102 The multiscale multiphase model of solidification is based on a volume averaging method with three
103 hydrodynamic phases [30,31], as schematically shown in Fig. 1. The model is an extension of the
104 earlier two-phase model [1,6,28,32] and uses the same constitutive relations and numerical solution
105 methods. The two crucial extensions are the more detailed three-phase description of the grain growth
106 kinetics and the inclusion of inertial and of solid-solid interaction forces in the momentum balance
107 equations for the solid phase. Both extensions are detailed below and all model equations are
108 summarized in Table 1. A detailed description of the model derivation is given in the aforementioned
109 references.

110

111 The model couples phenomena at the microscopic and the macroscopic scales. The conservation
112 equations for mass, solute, momentum, and the population of inoculant particles and of solid grains
113 include terms accounting for macroscopic transport and for interphase exchange at the microscopic
114 scale. On a microscopic scale, the dendritic grain structures are described in a volume-averaged sense
115 by the volume fractions of the solid phase, of the grain envelopes, of the extradendritic liquid (liquid
116 outside the envelopes), and of the interdendritic liquid (liquid inside the envelopes). The modeling of
117 the growth kinetics of these structures is presented in Section 2.1. The motion of the liquid melt and
118 of the solid grains at the macroscopic scale are modeled by taking into account: buoyancy-driven
119 flow induced by liquid density variations (thermosolutal convection modeled with the Boussinesq
120 approximation) and by the density difference between solid and liquid phases, inertial forces, drag
121 forces at the solid-liquid interfaces, momentum transfer by collisions between grains, and fluid flow
122 induced by solidification shrinkage. The solid grains are assumed to nucleate exclusively by athermal
123 heterogeneous nucleation [33] on inoculant particles. A size distribution of the inoculant particles is
124 considered and the nucleation undercooling is inversely proportional to the particle size. The
125 distribution is discretized into classes (or bins) and population balances are used to describe the
126 advection and the vanishing of the individual size classes [1,28]. The inoculant particles are supposed
127 to move at the liquid velocity. This description of nucleation is suited for modeling nucleation on
128 grain refiner particles, used in DC casting. After nucleating, the solid grains are considered as a
129 dispersed solid phase and remain such until the local grain envelope volume fraction exceeds the
130 packing fraction g_{ep} . We call this the *slurry* flow regime. At envelope volume fractions above
131 packing, $g_e > g_{ep}$, we consider the solid phase to be stationary and the drag at the solid-liquid
132 interfaces is described by a Darcy term modeled by the Kozeny-Carman law. We call this the *porous*
133 flow regime. Note that the grain morphology plays a major role in packing. Dendritic grains, whose
134 envelopes occupy more space than their solid skeleton, pack sooner in the solidification process (i.e.
135 at lower solid fraction) than globular grains.

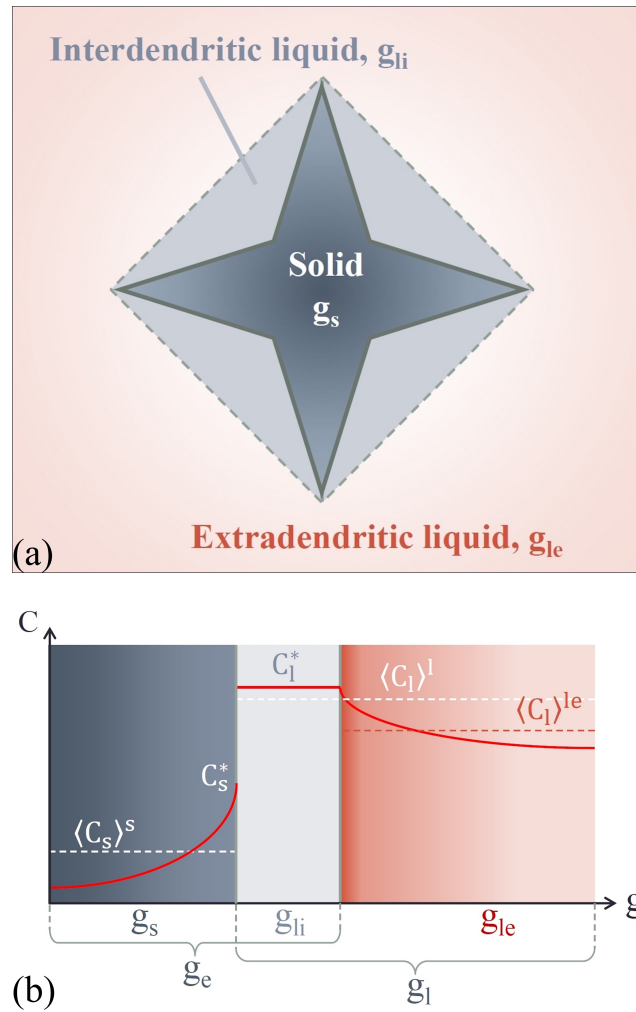


Figure 1: (a) Schematics of the hydrodynamic phases in the three-phase model. (b) Solute profiles in the three phases, solute concentrations and volume fractions.

Table 1: Summary of the model equations

Averaged conservation equations	
Total mass conservation	$\frac{\partial}{\partial t} (g_s \rho_s + g_l \rho_l) + \nabla \cdot (g_s \rho_s \langle \vec{v}_s \rangle^s + g_l \rho_l \langle \vec{v}_l \rangle^l) = 0$
Mass balance, solid phase	$\frac{\partial}{\partial t} (g_s \rho_s) + \nabla \cdot (g_s \rho_s \langle \vec{v}_s \rangle^s) = \Gamma_s + \Phi_s$
Volume balance, dendrite envelopes	$\frac{\partial}{\partial t} (g_e) + \nabla \cdot (g_e \langle \vec{v}_s \rangle^s) = \Gamma_e + \frac{\Phi_s}{\rho_s}$
Solute mass balance, solid phase	$\frac{\partial}{\partial t} (g_s \rho_s \langle C_s \rangle^s) + \nabla \cdot (g_s \rho_s \langle C_s \rangle^s \langle \vec{v}_s \rangle^s) = \Gamma_s C_s^* + \frac{\rho_s S_v^s D_s}{\delta_s} (C_s^* - \langle C_s \rangle^s) + \Phi_s k \langle C_l \rangle^l$
Solute mass balance, liquid phase	$\frac{\partial}{\partial t} (g_l \rho_l \langle C_l \rangle^l) + \nabla \cdot (g_l \rho_l \langle C_l \rangle^l \langle \vec{v}_l \rangle^l) = -\Gamma_s C_l^* + \frac{\rho_l S_v^e D_l}{\delta_{le}} \frac{g_l}{g_{le}} (C_l^* - \langle C_l \rangle^{le}) - \Phi_s k \langle C_l \rangle^l$
Energy balance	$\frac{\partial}{\partial t} (g_s \rho_s h_s + g_l \rho_l h_l) + \nabla \cdot (g_s \rho_s h_s \langle \vec{v}_s \rangle^s + g_l \rho_l h_l \langle \vec{v}_l \rangle^l) = \nabla \cdot (\lambda \nabla T)$ $h_s = c_p T h_l = c_p T + L_f$
Grain density balance	$\frac{\partial N_g}{\partial t} + \nabla \cdot (\langle \vec{v}_s \rangle^s N_g) = \sum_{i=1}^m \phi_i$
Inoculant density balance, for each class i	$\frac{\partial N_{nuc}^i}{\partial t} + \nabla \cdot (\langle \vec{v}_l \rangle^l N_{nuc}^i) = -\phi_i (i = 1 \dots m)$
Momentum conservation, solid phase, only when $g_e < g_{ep}$	$\frac{\partial}{\partial t} (\rho_s g_s \langle \vec{v}_s \rangle^s) + \nabla \cdot (\rho_s g_s \langle \vec{v}_s \rangle^s \langle \vec{v}_s \rangle^s) = -g_s \nabla p + \nabla \cdot (\mu_s \nabla (g_s \langle \vec{v}_s \rangle^s)) + \rho_s g_s \vec{g} - \vec{M}_l^d + \langle \vec{v}_s \rangle^s \Gamma_s$
Solid viscosity	$\mu_s = \frac{\mu_l}{g_e} \left[\left(1 - \frac{g_e}{g_{ep}^{max}} \right)^{-2.5 g_{ep}^{max}} - (1 - g_e) \right]$

Momentum conservation, liquid phase

$$\frac{\partial}{\partial t} (\rho_l g_l \langle \vec{v}_l \rangle^l) + \nabla \cdot (\rho_l g_l \langle \vec{v}_l \rangle^l \langle \vec{v}_l \rangle^l) = -g_l \nabla p + \nabla \cdot (\mu_l \nabla (g_l \langle \vec{v}_l \rangle^l)) + \rho_l^b g_l \vec{g} + \vec{M}_l^d - \langle \vec{v}_s \rangle^s \Gamma_s$$

$$\text{when } g_e < g_{eb}: \vec{M}_l^d = \frac{3g_e \rho_l C_d}{4d_g} |\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s| (\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s)$$

$$\text{when } g_e \geq g_{eb}: \vec{M}_l^d = \frac{-g_l^2 \mu_l}{K} (\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s)$$

Permeability

$$K = \frac{g_l^3}{(1 - g_l)^2} \frac{l_c^2}{\pi^2 k_K \tau^2}$$

Density for the buoyancy term

$$\rho_l^b = \rho_{ref} [1 - \beta_T (T - T_{ref}) - \beta_C (\langle C_l \rangle^l - C_{ref})]$$

Nucleation

Activation of inoculant class i

If at time t_0 , $(T_m + m \langle C_l \rangle^l(t_0)) - T(t_0) > \Delta T_i$: $\phi_i = N_{nuc}^i \delta(t - t_0)$, where δ is the Dirac delta function.

Else: $\phi_i = 0$.

Solid mass generation due to nucleation

$$\Phi_s = \rho_s V_0 \sum_{i=1}^m \phi_i$$

Grain growth kinetics

Envelope growth rate

$$\Gamma_{env} = \frac{1}{\sqrt{3}} S_v^e V_{tip}$$

Mass balance, at the solid-liquid interface

$$\Gamma_s + \Gamma_l = 0$$

Solute balance, at the solid-liquid interface

$$\frac{\rho_s S_v^s D_s}{\delta_s} (k C_l^* - \langle C_s \rangle^s) + \left(\frac{1 - g_s}{1 - g_e} \right) \frac{\rho_l S_v^e D_l}{\delta_e} (C_l^* - \langle C_l \rangle^l) = \Gamma_s (1 - k) C_l^* - \rho_l (g_e - g_s) \frac{\partial C_l^*}{\partial t}$$

Diffusion lengths

$$\delta_s = d_s / 10$$

$$\delta_e = \left[\frac{2}{d_g} + \frac{1}{\frac{D_l}{v_{tip}} \left(1 - \frac{8D_l/V_{tip}}{d_f^2 - d_g^2} \left[\frac{d_g}{2} + \frac{D_l}{v_{tip}} - \left(\frac{d_f}{2} + \frac{D_l}{v_{tip}} \right) \exp\left(-\frac{d_f - d_g}{2D_l/V_{tip}}\right) \right] \right)} \right]^{-1} :$$

Primary arm length and equivalent diameter of envelope and of solid phase

$$R_{arm} = \left(\frac{3g_e}{4N_g} \right)^{1/3} \quad d_g = \left(\frac{6g_e}{\pi N_g} \right)^{1/3} \quad d_s = \left(\frac{6g_s}{\pi N_g} \right)^{1/3}$$

Specific surface areas of envelope and solid

$$S_v^e = 4\sqrt{3}R_{arm}^2 N_v \quad S_v^s = \frac{4}{\lambda_2} g_i^{1/2} g_e (1 - g_i^m) + g_i^m S_v^e$$

Thermodynamic equilibrium at the solid-liquid interface

$$C_s^* = kC_l^* \\ T = T_m + mC_l^*$$

Paraboloidal tip model [36,53]

Total tip undercooling

$$T - T_m - m\langle C_l \rangle^l = \frac{L_f}{c_p} \Omega_T + mC_0 \left[1 - \frac{1}{1 - (1 - k)\Omega_C} \right]$$

Dimensionless undercooling and supersaturation of a dendrite tip with convection [31,53]

$$\Omega_T = F(Pe_T, Pu_T, Re_T) \quad \Omega_C = F(Pe_C, Pu_C, Re_C)$$

$$F(Pe, Pu, Re) = 2Pe \int_1^\infty \exp \left[-\ln \eta + (1 - \eta^2) Pe - \frac{2Pu}{E_1(Re)} [1 - \eta^2 + \ln(\eta)(1 + \eta^2)] \right] d\eta$$

$$Pe_T = \frac{R_{tip} V_{tip}}{2\alpha} \quad Pe_C = \frac{R_{tip} V_{tip}}{2D_l}$$

$$Pu_T = \frac{R_{tip} |\langle \vec{v}_s \rangle^s - \langle \vec{v}_l \rangle^l|}{2\alpha} \quad Pu_C = \frac{R_{tip} |\langle \vec{v}_s \rangle^s - \langle \vec{v}_l \rangle^l|}{2D_l}$$

$$Re_T = \frac{Pe_T + Pu_T}{Pr} \quad Re_C = \frac{Pe_C + Pu_C}{Sc}$$

Tip selection criterion

$$R_{tip}^2 V_{tip} = \frac{\Gamma_{GT}}{\sigma^*} \left[\frac{m(1 - k)C_0}{D_l(1 - (1 - k)\Omega_C)} + \frac{L_f}{2\alpha c_p} \right]^{-1}$$

Hemispherical tip model [37]

Tip velocity

$$V_{tip} = \frac{D_l(T - T_m - m\langle C_l \rangle^l)^2}{\Gamma_{GT}(k - 1)mC_0\pi^2}$$

138 2.1. Three-phase model of grain growth

139 The equiaxed grains are described by their size and their morphology, which can be dendritic or
 140 globular. The description of the morphology in the volume-averaged model is based on the idea of
 141 the dendrite envelope, first proposed by Rappaz and Thévoz [34]. The solid volume fraction g_s is
 142 distinguished from the grain envelope volume fraction, g_e , as illustrated in Fig. 1a. When a grain is
 143 dendritic, its envelope, defined in the model by a regular octahedron linking the six grain tips, is larger
 144 than the solid volume of the grain. A part of the interior of the grain envelope is therefore liquid. The
 145 model distinguishes between the liquid outside the envelopes, named extradendritic liquid, and the
 146 liquid inside the envelopes, between the dendrite arms, named interdendritic liquid. Their volume
 147 fractions are g_{le} and g_{li} , respectively, and their sum is the liquid fraction $g_l = g_{li} + g_{le}$. The
 148 relationships between the different volume fractions are:

$$g_e + g_{le} = g_s + g_{li} + g_{le} = g_s + g_l = 1, \quad g_i = \frac{g_s}{g_e} \quad (1)$$

149 The indication of the grain morphology is given by the internal solid fraction g_i , which is the ratio
 150 between the volume of the solid and the envelope. If the internal solid fraction is close to one, the
 151 grains are considered to be globular, whereas when it is close to zero, the grains are considered to be
 152 dendritic. The final morphology of the solidified equiaxed structure is usually considered to
 153 correspond to the morphology at the instant of grain packing [31].

154 We assume that the interdendritic and extradendritic liquid phases move at the same flow velocity
 155 $\langle \vec{v}_{le} \rangle^{le} = \langle \vec{v}_{li} \rangle^{li} = \langle \vec{v}_l \rangle^l$ and have the same density $\rho_{le} = \rho_{li} = \rho_l$. This assumption is different from
 156 the treatment in the classical three phase model with convection [30] that accounts for the partitioning
 157 of the flow passing through and around the envelopes due to drag. Our description is not as detailed,
 158 but can be reasonable for coarse dendritic grains encountered in DC casting. It also considerably
 159 simplifies the macroscopic transport equations. The macroscopic conservation equations for the two
 160 liquid phases can be summed to a single equation for the liquid, identical to those in a two-phase
 161 model. The global liquid mass conservation equation is then:

$$\begin{aligned} \frac{\partial}{\partial t} (\rho_l g_{le} + \rho_l g_{li}) + \nabla \cdot (\langle \vec{v}_l \rangle^l (\rho_l g_{le} + \rho_l g_{li})) &= \\ = \frac{\partial}{\partial t} (\rho_l g_l) + \nabla \cdot (\langle \vec{v}_l \rangle^l (\rho_l g_l)) &= -\Gamma_s - \Phi_s, \end{aligned} \quad (2)$$

162 where Γ_s and Φ_s are the mass transfer rates from liquid to solid due to phase change and nucleation,
 163 respectively. Similarly, the solute conservation equation is formulated in terms of the average liquid
 164 concentration, $\langle C_l \rangle^l = (g_{li} \langle C_l \rangle^{li} + g_{le} \langle C_l \rangle^{le}) / g_l$.

$$\begin{aligned} \frac{\partial}{\partial t} (g_{le} \langle C_l \rangle^{le} + g_{li} \langle C_l \rangle^{li}) + \nabla \cdot (\langle \vec{v}_l \rangle^l (g_{le} \langle C_l \rangle^{le} + g_{li} \langle C_l \rangle^{li})) &= \\ = \frac{\partial}{\partial t} (g_l \langle C_l \rangle^l) + \nabla \cdot (g_l \langle \vec{v}_l \rangle^l \langle C_l \rangle^l) &= \frac{1}{\rho_l} (-C_s^* \Gamma_s - C_s^* \Phi_s - j_s^d) \end{aligned} \quad (3)$$

165 where Γ_k is the mass flux into phase k per unit volume at the phase interface due to phase change and
 166 Φ_k is the phase mass transfer rate due to nucleation. Note that the total of all interfacial fluxes is zero,
 167 therefore the sum of the fluxes into both liquid phases is equal to minus the sum of the fluxes into the
 168 solid phase.

169 The balance equations for interphase exchange of mass and solute at the microscopic scale are
 170 modified by the three-phase description. Nevertheless, Tveito [35] derived a formulation of the
 171 equations in terms of the average liquid concentration $\langle C_l \rangle^l$ that results into equations of a form similar

172 to that of the two-phase model. A detailed derivation of these equations is given in Appendix A. The
 173 interdendritic liquid is assumed to be perfectly mixed at the uniform concentration equal to the
 174 thermodynamic equilibrium concentration at the solid-liquid interface. Diffusion and convection
 175 govern the transport of solute in the extradendritic liquid around the envelope and finite diffusion is
 176 accounted for in the solid. A typical solute profile in and around a grain is illustrated in Fig. 1b.

177 The growth of the octahedral envelopes is driven by the growth of the primary tips corresponding to
 178 the six vertices. Analytical expressions for the dendrite tip velocity as a function of the undercooling
 179 exist for paraboloid [36] and hemispherical tip shapes [37]. A freely growing dendrite tip is accurately
 180 described by a paraboloid. However, Nielsen et al. [38] pointed out that the hemispherical tip model
 181 gave better predictions of the experimentally observed morphologies of interacting grains in
 182 inoculated Al-Cu alloys. Therefore, both tip models are considered in this study.
 183

184 2.2. Modeling of the slurry flow

185 In the slurry flow regime the free-floating equiaxed grains move with a velocity that is distinct from
 186 the liquid velocity. The velocity of each phase is determined with a separate momentum balance.
 187 Because of the assumption that the interdendritic and the extradendritic liquid have the same velocity,
 188 the mass and momentum balances can be summed and written globally for the liquid. Thus, only a
 189 solid and a liquid hydrodynamic phase need to be considered. The averaged mass balance for phase
 190 k ($k = s, l$) is given by:

$$\frac{\partial}{\partial t} (g_k \rho_k) + \nabla \cdot (g_k \rho_k \langle \vec{v}_k \rangle^k) = \Gamma_k + \Phi_k \quad (4)$$

191
 192 The momentum conservation equation for phase k is as follows:

$$\begin{aligned} \frac{\partial}{\partial t} (g_k \rho_k \langle \vec{v}_k \rangle^k) + \nabla \cdot (g_k \rho_k \langle \vec{v}_k \rangle^k \langle \vec{v}_k \rangle^k) \\ = -g_k \nabla p_k + \nabla \cdot (g_k \langle \tau_k \rangle^k) + g_k \rho_k^b \vec{g} + M_k^d + M_k^\Gamma + M_k^\Phi \end{aligned} \quad (5)$$

193 where M_k^d is the interfacial drag force per unit volume, M_k^Γ is the momentum flux across the solid-
 194 liquid interface due to phase change and M_k^Φ is the momentum transfer due to nucleation of the solid
 195 from the liquid phase. The nucleation term is negligible.
 196

197 The interfacial drag force is modeled as:

$$\vec{M}_s^d = -\vec{M}_l^d = \frac{3g_e \rho_l C_d}{4d_g} |\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s| (\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s), \quad (6)$$

198 where d_g is the equivalent spherical diameter of the grain envelope, $\langle \vec{v}_s \rangle^s$ and $\langle \vec{v}_l \rangle^l$ are the intrinsic
 199 average velocities of the liquid and solid phases, and C_d is the drag coefficient calculated as given in
 200 [6]. By using the envelope fraction, g_e , and the equivalent envelope diameter, d_g , the influence of
 201 the morphology on the drag is naturally taken into account. At a given solid fraction a dendritic grain
 202 experiences more drag than a globular grain and its slip velocity is smaller. The stress in the solid
 203 phase, generated by grain collisions, is modeled through a pseudo-fluid model [30,39]. Thus the stress
 204 term for both phases is formulated as:

$$\nabla \cdot (g_k \langle \tau_k \rangle^k) = \nabla \cdot (\mu_k \nabla (g_k \langle \vec{v}_k \rangle^k)). \quad (7)$$

205 The solid viscosity is calculated by the relation [40]:

$$\mu_s = \frac{\mu_l}{g_e} \left[\left(1 - \frac{g_e}{g_{ep}^{max}} \right)^{-2.5 g_{ep}^{max}} - (1 - g_e) \right], \quad (8)$$

206 with the maximum envelope packing fraction, g_{ep}^{max} , fixed at 0.6, the approximate value for a close
 207 random packing of monodisperse spheres. The pressure is assumed to be identical in both phases.
 208

209 The momentum conservation for the slurry zone may be rewritten for both phases as:

$$\begin{aligned} \frac{\partial}{\partial t} (\rho_k g_k \langle \vec{v}_k \rangle^k) + \nabla \cdot (\rho_k g_k \langle \vec{v}_k \rangle^k \langle \vec{v}_k \rangle^k) = \\ -g_k \nabla p + \nabla \cdot (\mu_k \nabla (g_k \langle \vec{v}_k \rangle^k)) + \rho_k^b g_k \vec{g} + \vec{M}_k^d + \Gamma_k \langle \vec{v}_k \rangle^k \end{aligned} \quad (9)$$

210 In the packed zone, considered as a porous medium, the solid is moving at the casting velocity and
 211 the drag force exerted on the liquid is
 212

$$\vec{M}_l^d = -\frac{g_l^2 \mu_l}{K} (\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s), \quad (10)$$

213 where K is the hydrodynamic permeability of the microstructure. It is modeled by the Kozeny-
 214 Carman relation:

$$K = \frac{g_l^3}{(1-g_l)^2} \frac{l_c^2}{\pi^2 k_K \tau^2}, \quad (11)$$

215 where l_c is the characteristic length of the porous microstructure. The Kozeny constant, k_K , and the
 216 tortuosity of the porous medium, τ , depend on its morphology. Values for a packed bed of spheres
 217 were used: $k_K = 5$ and $\tau = \pi/6$. The resulting momentum equation for the liquid in the porous zone
 218 is:

$$\begin{aligned} \frac{\partial}{\partial t} (\rho_l g_l \langle \vec{v}_l \rangle^l) + \nabla \cdot (\rho_l g_l \langle \vec{v}_l \rangle^l \langle \vec{v}_l \rangle^l) = \\ -g_l \nabla p + \nabla \cdot (\mu_l \nabla (g_l \langle \vec{v}_l \rangle^l)) + \rho_l^b g_l \vec{g} - \frac{g_l^2 \mu_l}{K} (\langle \vec{v}_l \rangle^l - \langle \vec{v}_s \rangle^s) - \Gamma_s \langle \vec{v}_s \rangle^s \end{aligned} \quad (12)$$

219 A summary of all model equations is presented in Table 1.

220

221 The conservation equations were solved with the finite volume method. An operator splitting scheme
 222 [32] was used to integrate the solute and solid mass conservation equations. The splitting scheme was
 223 extended to account for shrinkage induced flow and for transport of grain refiner particles [28]. The
 224 pressure-velocity coupling was resolved by the IPSA pressure correction algorithm for multiphase
 225 flows [41].
 226

227 3. DC casting experiments and their modeling

228 3.1. Experiments of Vreeman et al.

229 Vreeman and coworkers [14] cast two binary-alloy Al-Cu billets of 450 mm diameter, one inoculated
 230 with Al-3Ti-1B grain refiner and one without grain refiner. The other casting parameters were
 231 identical. The casting speed was 1 mm/s and hot-top molds were used. The macrosegregation was
 232 measured by X-ray fluorescence on 20 samples in each billet. More detailed information on the
 233 sampling was not given. The nominal composition of the two billets was slightly different with Al-
 234 6.1 wt.%Cu for the grain refined and Al-5.9 wt.%Cu for the non grain refined billet. In order to
 235 perform a direct comparison, the experimental data were shifted by $-0.1/+0.1$ wt.% to mimic an Al-

236 6 wt.%Cu billet. The simulations were also performed with the composition Al-6 wt.%Cu for both
237 billets. The main observation of the experiment was a substantially weaker centerline
238 macrosegregation in the non grain refined billet.

239 For the grain refined billet two additional experimental results are available. The temperature field
240 was characterized by inserting thermocouples into the metal during casting. The thermocouples were
241 inserted at three radial positions and were pulled down with the billet at the casting speed. The exact
242 three positions were at radial distances from the centerline of $r = 10$ mm (“centerline”), $r = 106$ mm
243 (“mid-radius”), and $r = 220$ mm (“subsurface”). The uncertainty of the thermocouple measurements
244 was ± 2 °C. Characterizations of the size, position and shape of the mushy zone were obtained by a
245 sudden addition of a molten Al-Si mixture. The etched macrograph of the billet section revealed the
246 solidus front and an indication of the packing front. All experimental results are shown along with
247 the simulations in the later sections.

248 Characterizations of the microstructure in terms of grain size and morphology were not reported by
249 Vreeman et al. A grain size of 75 μm used in their simulations was mentioned as representative of
250 the typical grain size in both billets. The structure of the grain refined billet was asserted to be fully
251 equiaxed. The structure of non grain refined billet was less clear and speculations about a possible
252 columnar-to-equiaxed transitions were made [14,42].

253

254 3.2. Modeling of the experiments

255 The billets were described by a 2D axisymmetric geometry. The computational mesh consisted of a
256 grid of 60×120 finite-volume elements, with a finer mesh in the liquid and mushy zone and a coarser
257 mesh in the fully solidified zone. The inlet was assumed to be across the whole cross-section and the
258 feeding velocity was assumed to be uniform. The melt temperature at the inlet was 943.35 K (670.35
259 °C), as specified in [14]. The mold height was 7 cm. The top 1 cm of the mold was supposed to be
260 thermally insulating and the heat transfer from the billet in the lower 6 cm of the mold was modeled
261 by a heat transfer coefficient of 30 $\text{W}/\text{m}^2\text{K}$ and a reference coolant temperature of 291 K (18 °C). The
262 heat extraction by the cooling water in the direct chill zone was described by the well-established
263 correlation of Weckman & Niessen [43]. The thermophysical properties of the alloy are summarized
264 in Table 2; they are the same as used in [14]. The thermal conductivity in the mushy zone is modeled
265 as a phase-fraction weighted average of the solid and liquid conductivities. The density of the solid
266 phase is considered as constant and a Boussinesq approximation is used for the liquid phase to account
267 for the buoyancy forces acting on the liquid and on the free-floating grains. The variation of the
268 densities during solidification is illustrated in Fig. 2. The densities in all mass balances are considered
269 as constant. Thus the density difference that induces flow due to solidification shrinkage is given by
270 a constant shrinkage coefficient of $\beta_{sl} = (\rho_s - \rho_{l,ref})/\rho_s = 0.032$. The size distribution of the grain
271 refiner particles used in the nucleation law was taken from the measurements of Tronche for an Al-
272 Ti-B grain refiner [44]. The distribution was discretized [28] into 13 classes of particles; the
273 nucleation law is shown in Fig. 3.

274

Table 2: Thermo-physical properties and model parameters

<i>Phase diagram (Al–Cu)</i>		<i>Process parameters</i>	
Melting temperature of pure Al [°C] ¹	677.8	Casting temperature [°C]	670.35
Nominal composition in Cu [wt.%]	6.0	Casting speed [m·s ⁻¹]	10 ⁻³
Eutectic temperature [°C]	547.85	Heat transfer coefficient, insulation [W·m ⁻² ·K ⁻¹]	0
Liquidus slope [°C·wt% ⁻¹]	-3.425	Heat transfer coefficient, mold [W·m ⁻² ·K ⁻¹]	30
Binary partition coefficient [-]	0.173	Insulation height [m]	0.01
		Mold height [m]	0.07
		Reference coolant temperature [°C]	18
<i>Thermo-physical data</i>		<i>Computational parameters</i>	
Specific heat [J·kg ⁻¹ ·K ⁻¹]	1300	Dimensions of the domain (R×Y) [m×m]	0.225×0.90
Solid thermal conductivity [W·m ⁻¹ ·K ⁻¹]	162	Number of grid volumes, R×Y	60×130
Liquid thermal conductivity [W·m ⁻¹ ·K ⁻¹]	95		
Reference density of the liquid, ρ _{l,ref} [kg/m ³]	2490		
Latent heat of fusion [J·kg ⁻¹]	3.92·10 ⁵		
Thermal expansion coefficient, β _T [°C ⁻¹]	1.17·10 ⁻⁴		
Solutal expansion coefficient, β _C [wt% ⁻¹]	-7.3·10 ⁻³		
Reference temperature for density [°C]	660		
Reference Cu concentration for density	6.0		
Dynamic viscosity [Pa·s]	1.28·10 ⁻³		
Diffusion coefficient, liquid [m ² ·s ⁻¹]	5.66·10 ⁻⁹		
Diffusion coefficient, solid [m ² ·s ⁻¹]	5.6·10 ⁻¹³		
Shrinkage coefficient β _{sl} = (ρ _s -ρ _{l,ref})/ρ _s	0.0319		
Characteristic length for permeability [m]	7.5·10 ⁻⁵		
Gibbs-Thompson coefficient [K·m]	1.9·10 ⁻⁷		

275

276

¹ Etrapolation of the linearized liquidus line to zero concentration.

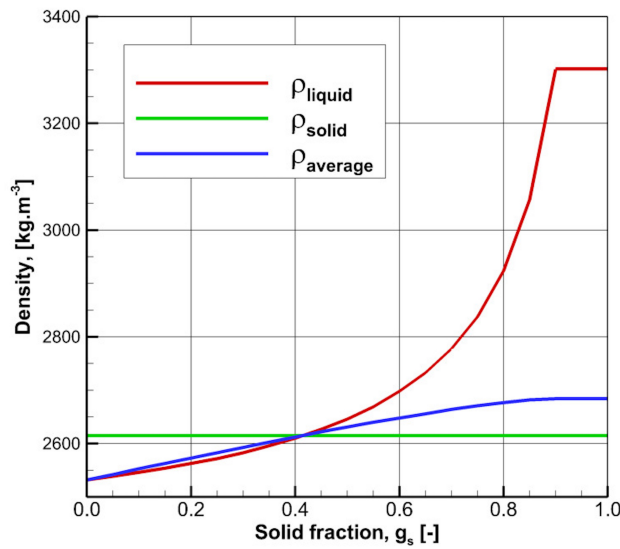


Figure 2: Evolution of solid, liquid and average densities of the alloy Al-6wt.%Cu with solid fraction. The solidification path is assumed to follow the Scheil-Gulliver model. The variation of the solid density due to the appearance of a secondary phase upon the eutectic reaction is not accounted for.

277

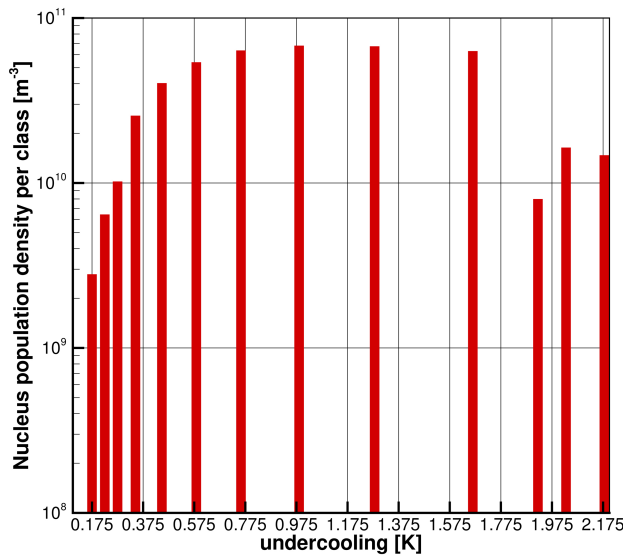


Figure 3: Discretization of the distribution of nucleation sites with respect to the critical undercooling. The distribution corresponds to the size distribution of TiB_2 particles for 1 kg/t of Al-5Ti-1B grain refiner [28,44].

278 Some of our results are compared to the simulations of Vreeman et al. [14], who used a much simpler
 279 model, especially with respect to the microstructures. Their model considered the equiaxed grains to
 280 be *globular* and of a constant size (fixed to 75 μm), and described the solidification by the lever rule.
 281 Notably, with these assumptions the local grain population density must be adjusted in order to
 282 correspond to the solid fraction obtained from the lever rule and to the fixed grain size. Vreeman et
 283 al. used a similar criterion as the present work for packing of the grains. The grains were considered
 284 to be packed above a *solid* fraction of 0.3.
 285

286 4. Results and discussion

287 4.1. General features of the heat transfer, flow and microstructure

288 Model predictions of heat transfer, fluid flow, motion of the equiaxed grains and of solidification in
289 the sump of the billet are illustrated in Fig. 4. These are results from a simulation that used a packing
290 fraction of $g_{ep} = 0.20$ and the paraboloidal dendrite tip model.

291 The temperature and the solid fraction fields are shown in Figs. 4a and 4b, respectively. The radial
292 heat extraction from the metal that is fed into the mold from the top in liquid state creates the
293 characteristic roughly triangular shape of the solidification zone (sump). The heat extraction in the
294 mold is relatively weak due to the particular mold design. The ingot is cooled mainly by the direct
295 contact with the water, which starts abruptly below the mold. This explains why the sump in the
296 rapidly cooled zone at the billet surface bends perpendicularly to the surface. In the billet center the
297 temperature gradients are much smaller and a large part of the mushy zone in the core has a virtually
298 homogeneous temperature that is only slightly below the liquidus temperature of 930.25 K (657.25
299 °C).

300 The flow is driven by thermosolutal natural convection and by the settling equiaxed grains. Due to
301 the density relations in the Al-6 wt%Cu alloy (Fig. 2), all three driving forces cooperate. The colder,
302 solute enriched liquid with a higher fraction of solid grains, forming at the slopes of the sump, has a
303 higher density than the hotter solute-lean liquid in the core. The buoyancy forces induce a descending
304 current along the mushy zone and develop a flow in a roughly triangular pattern with one major loop,
305 as shown in Fig. 4c. More precisely, below the inlet the flow deflects horizontally towards the mold
306 and then rushes downwards parallel to the packing front towards the billet center. The flow velocity
307 is the largest in the current descending along the packing front, due to the conjunction of strong
308 buoyancy forces and drag forces induced by grain sedimentation (as the solid phase is heavier than
309 the surrounding liquid). Then the flow slowly turns upward in the central part of the billet and runs
310 again towards the mold.

311 The field of the intrinsic velocity of the solid phase is shown in Fig. 4d. The solid forms as equiaxed
312 grains nucleating on grain refiner particles. The nucleation zone is located adjacent to the inclined
313 part of the packing front [1,45], which is a result of a particular competition between nucleation,
314 growth and grain transport [28]. The solid grains are heavier than the liquid and tend to sediment until
315 packing. The packing is modeled by a limit envelope volume fraction. In the example shown in Fig.
316 4 the packing fraction is $g_{ep} = 0.2$ and the packing front is shown by a black line. The grains descend
317 towards the billet center in a current parallel to the packing front. Some of them pack on the way,
318 most of them accumulate in a wide packing layer at the bottom of the sump in the billet center, and
319 some grains are dragged upwards by the liquid flow. This migration of the solid phase from the
320 periphery to the billet core results in a global advection of latent heat in the inverse direction. This
321 cools down the core and also produces the isothermal zone in the core of the sump.

322

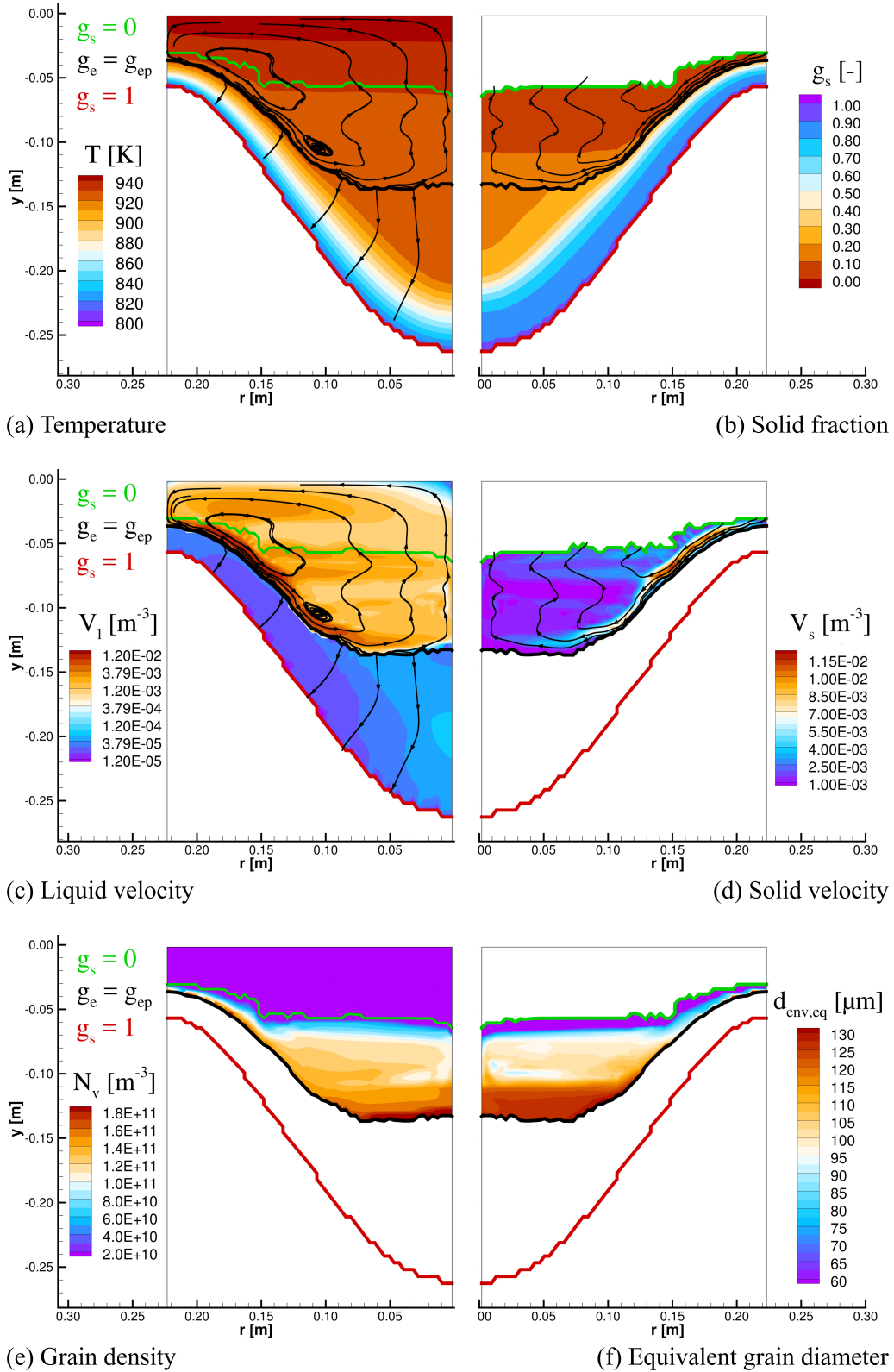


Figure 4: Heat transfer and flow in the billet: (a) temperature field, (b) solid fraction, (c) norm of the liquid velocity and liquid velocity streamlines, (d) norm of the solid velocity and solid velocity streamlines. The grain refined billet simulated with a packing fraction of $g_{ep}=0.2$ and the paraboloidal dendrite tip model is shown. The relative velocities are shown in the reference frame of the moving product.

323 Although no information on the grain size was reported in the experiments, it is interesting to have a
 324 look to the at the distributions of grain density and grain size in the slurry zone, shown in Fig. 4e and
 325 4f, respectively. The variations of both grain size and grain density are mainly in the vertical direction
 326 and the highest values of both are located at the packing front. With the exception of a thin layer close
 327 to the liquid region, in most of the slurry region the grain density varies from around $8 \cdot 10^{10}$ to $1.8 \cdot 10^{11}$
 328 grains/m³ and the grain size from around 75 to 130 μm . These variations are not exceedingly large
 329 and they can justify the assumption of a fixed grain size that was adopted by Vreeman et al. [14]. The
 330 main limitation of such an assumption is that the grain size becomes an input parameter of the model
 331 that is difficult to determine a priori.

332 The microstructure was simulated with two different dendrite tip models, leading to significantly
 333 different predictions of the equiaxed grain morphology. Fig. 5 shows the internal solid fraction in the
 334 mushy zone obtained with both tip models. A packing fraction of $g_{ep} = 0.2$ was used in both
 335 simulations. The paraboloidal tip model predicts fully globular grains in the whole mushy zone, i.e.
 336 a globular morphology from nucleation until packing of the grains. Contrarily, the hemispherical
 337 model predicts a rapid dendritization of the grains and an evolution of the morphology. Nevertheless,
 338 the grains remain dendritic until they pack. The internal solid fraction at the packing front, g_{si} , is
 339 characteristic of the final grain morphology. The predicted morphology profiles across the billet
 340 radius in terms of this criterion are shown in Fig. 6. The profiles are given for simulations with both
 341 dendrite tip models and using envelope packing fractions between 0.10 and 0.55. The plot clearly
 342 shows that the parabolic tip model predicts globular grains and the hemispheric tip model predicts
 343 dendritic grains. The latter model also indicates somewhat less pronounced dendritic morphologies
 344 in the core than at the billet surface as an overall trend. The predictions depend weakly on the packing
 345 fraction, with the tendency of giving somewhat more globularized morphologies at higher packing
 346 fractions, especially in the region that coincides with the most inclined part of the packing front. The
 347 tendency of the hemispherical tip model to predict a more pronounced dendritization of the grains
 348 than the parabolic tip model has already been reported in literature [38].

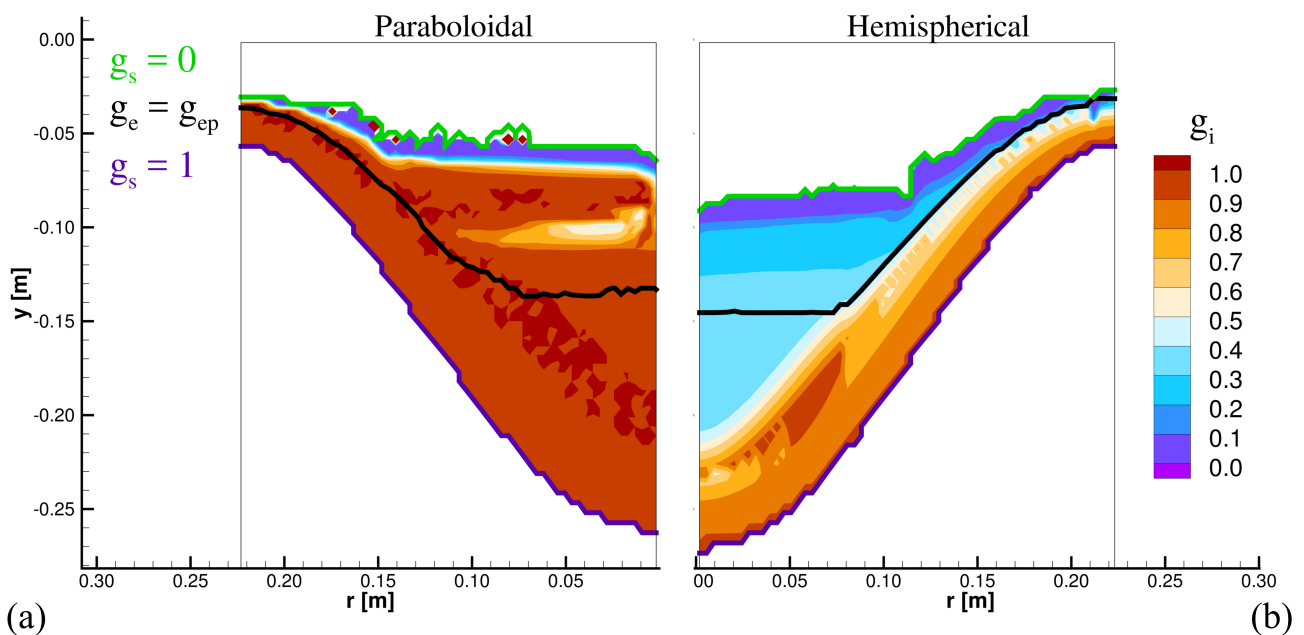


Figure 5: Internal solid fraction in the mushy zone obtained with both dendrite tip models: (a) paraboloidal, (b) hemispherical tip model. The packing fraction is $g_{ep} = 0.20$ in both cases.

349

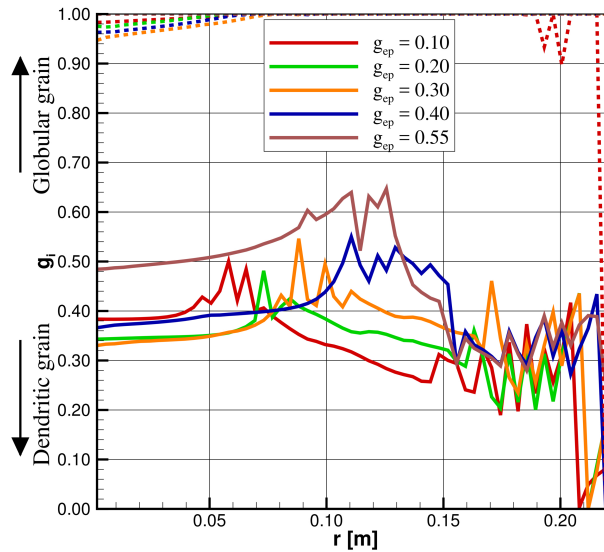


Figure 6: Internal solid fraction at packing (g_{ip}) for both tip growth models, hemispherical (solid lines) and paraboloidal (dashed lines).

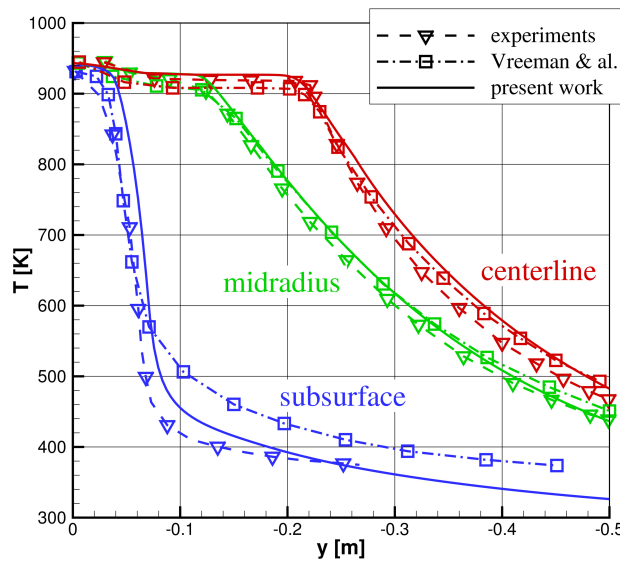
350

351 4.2. Comparison to measurements of temperature and sump shape

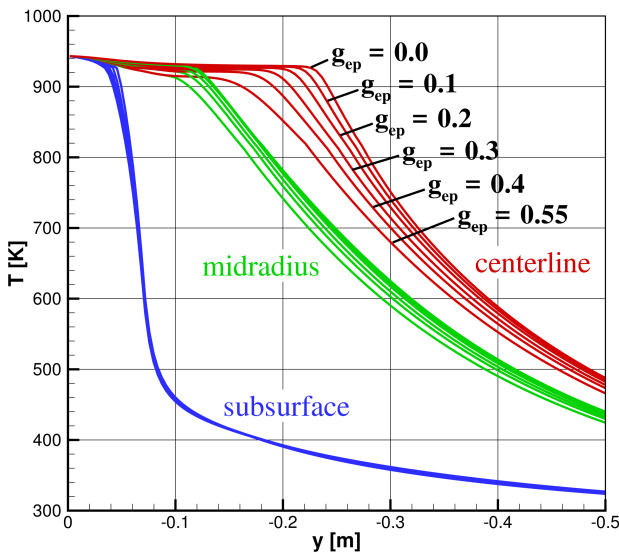
352 Before analyzing the model predictions in more detail, it is essential to validate the computed thermal
 353 field by comparison to the experiment. The comparison between the experimental temperature
 354 profiles and the present model (using $g_{cp}=0.20$ and the paraboloidal tip model), as well as with the
 355 model of Vreeman et al. [14] is shown in Fig. 7a. The agreement between the experimental and the
 356 predicted profiles is very good in the center of the ingot and at mid-radius. The subsurface profile is
 357 also accurately predicted and globally fits better to the experiment than the simulations of Vreeman
 358 et al., despite the use of the generic Weckman-Niessen thermal boundary condition instead of a fit to
 359 the measurements. The centerline temperature profiles indicate that the mushy zone is nearly
 360 isothermal up to a depth of 20 cm in the ingot center. This isothermal zone reaches down to more
 361 than half way through the packed mushy layer (and does not end at the packing front as suggested in
 362 [14]).

363 The effect of the packing fraction on the thermal field is illustrated in Figs. 7b and 7c, for the
 364 paraboloidal and the hemispherical tip model, respectively. An increase of the packing fraction leads
 365 to a significant temperature decrease in the core of the billet, especially at the centerline. When the
 366 grains pack at a higher volume fraction, a larger proportion of the solid in the mushy zone is mobile.
 367 Because of this, the latent heat transport from the billet core to the periphery, induced by grain motion
 368 in the opposite direction, is intensified. This effect is more significant when the grains are globular
 369 (Fig. 7b) than when they are dendritic (Fig. 7c). The reason is that the solid fraction of globular grains
 370 at packing is higher than that of dendritic grains by a factor of g_{ip}^{-1} and therefore a larger amount of
 371 solid is set in motion.

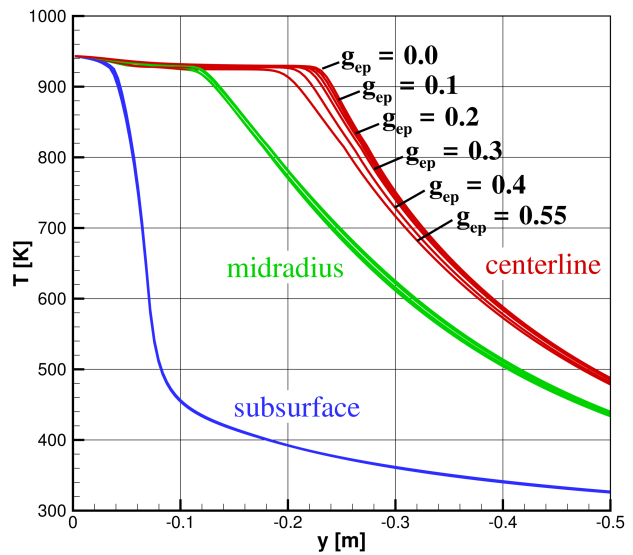
372



(a) Experiments and simulation



(b) Globular grains



(c) Dendritic grains

Figure 7: Comparison of measured and computed vertical temperature profiles at three different radius in the ingot (subsurface in blue, midradius in green and centerline in red). (a) Comparison of simulations with globular grains (paraboloidal tip model) and $g_{ep}=0.2$ with experiments and with simulations of Vreeman et al. [14]. (b) Influence of the packing fraction on the temperature profiles for globular grain morphology (paraboloidal tip model). (c) Influence of the packing fraction on the temperature profiles for dendritic grain morphology (hemispherical tip model).

373 A comparison between experimental and simulated sump geometries is shown in Fig. 8. In Fig. 8a
 374 the sump shape measured in the refined billet is superimposed on the packing and the solidus fronts
 375 predicted by the model (using $g_{ep}=0.20$ and the paraboloidal tip model, which gave the most accurate
 376 results). The data are shifted such that the solidus fronts are aligned at the centerline. We can see that
 377 the fit is very good, both in terms of shape and overall dimensions. In order to determine the best
 378 value of g_{ep} by comparison with the experiment, two characteristic dimensions of the sump were

379 extracted from the experimental measurements. The distance between the packing front and the
 380 solidus at mid-radius, $L1$, represents the thickness of the mushy zone ($L1_{exp} = 5$ cm) and $L2$ is the
 381 height of the solidus front ($L2_{exp} = 19.3$ cm). In Fig. 8b these two dimensions from a series of
 382 simulations using both dendrite tip models and a wide range of packing fractions are compared to the
 383 experimental values. We can see that the solidus height $L2$ is well estimated overall. It decreases with
 384 increasing packing fraction. This is a consequence of an enhancement of the cooling of the core
 385 through latent heat transport by grain motion, as explained earlier. This effect is more pronounced
 386 with globular grains, as predicted by the paraboloidal tip model, than with dendritic grains, predicted
 387 by the hemispherical tip model. The prediction of the mushy zone thickness $L1$ is much more sensitive
 388 to the packing fraction. The simulations give rather accurate results for small packing fractions (g_{ep}
 389 = 0.10 – 0.20). For larger packing fractions, the mushy zone thickness at mid-radius is clearly
 390 overestimated. The main reason is that at higher packing fractions (between 0.30 and 0.40) the plateau
 391 of the packing front in the core widens and its characteristic inflection shifts further outwards
 392 (compare Figs. 9a,c to Figs. 9b,d). This clearly indicates that the extent of the packed layer is
 393 overestimated when using high packing fractions. The predicted increase of the packing layer with
 394 an increasing packing fraction is stronger with the hemispherical tip model.

395

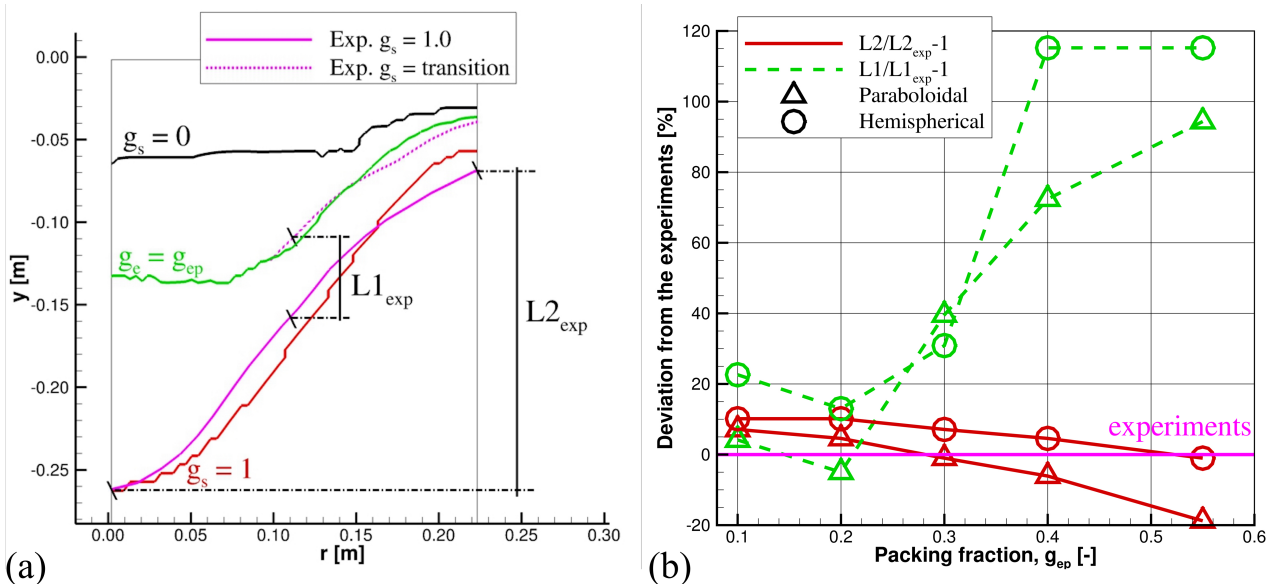


Figure 8: (a) Comparison of the sump shape obtained with the paraboloidal tip growth model and $g_{ep} = 0.20$ to the experimental sump shape. (b) Relative error of the lengths $L1$ and $L2$ as a function of the packing fraction, shown for simulations with both tip models.

396 The validation of the model by comparison to measurements of temperature in the billet and to
 397 experimentally determined sump shape and dimensions has shown that the model predictions
 398 significantly depend on the packing fraction of the free-floating equiaxed grains. The packing fraction
 399 determines the intensity of the heat transfer due to the transport of latent heat by grain motion in the
 400 sump. This effect is particularly strong when the grains are globular due to a larger proportion of
 401 moving solid in the mushy zone. Dendritic grains have a considerably weaker influence on the heat
 402 transfer and less impact on the thermal state in the casting.

403

404 4.3. Macrosegregation in the grain refined billet

405 The formation of the macrosegregation in the billet is shown in Fig. 9 for four cases: with globular
406 and dendritic grain morphologies (stemming from the use of the two dendrite tip models), each for
407 two packing fractions ($g_{ep} = 0.20$ and $g_{ep} = 0.40$). The field of the average “mixture” composition,
408 $C_m = (1 - g_s)\langle C_l \rangle^l + g_s\langle C_s \rangle^s$, and the streamlines of the liquid velocity are represented. We can
409 distinguish four zones of segregation formation [1]:

- 410 • the slurry zone, where a natural convection flow caused by thermosolutal buoyancy and grain
411 motion determines the solute transport;
- 412 • the packing front, where the sudden accumulation of solute-lean grains causes a negative jump
413 of the solute concentration;
- 414 • the moderately permeable packed layer (moderate g_l), where the natural convection and the
415 shrinkage-induced flow determine the path of the liquid and the segregation caused thereby;
- 416 • the weakly permeable packed layer (low g_l), where shrinkage flow dominates the solute
417 transport, leading to a negative segregation tendency in the center and positive at the
418 periphery.

419 The coupling and superposition of these phenomena, the roots of the final macrosegregation pattern,
420 were investigated in detail in [1,45]. It was shown that the grain transport is not only directly
421 responsible for the creation of the negative centerline segregation, but also changes the sump shape
422 and thus modifies the trajectory of the natural convection and shrinkage flow and the segregation they
423 cause. The cooperation of the listed transport phenomena results in a radial segregation profile, shown
424 in Fig. 10. The predicted profile is qualitatively similar to the experiment and both show a strong
425 negative segregation in the billet center, a weak positive segregation from the mid-radius outwards,
426 and a neutral or slightly negative segregation at the surface of the billet.

427 Figs. 9 and 10 show that the shape and the intensity of the predicted macrosegregation are strongly
428 dependent on the morphology of the equiaxed grains. To a smaller, but significant degree the
429 predicted segregation also depends on the packing fraction used in the computations. The segregation
430 in case of globular morphologies is systematically stronger than in case of dendritic morphologies.
431 The most striking difference is a stronger negative centerline segregation. The negative centerline
432 segregation is favored by settling of solute-lean grains and expulsion of solute-rich liquid. If the grains
433 have a globular morphology, they settle into a compact packing, carrying a large volume of solute-
434 lean solid phase. In comparison, ramified dendritic grains create a loose packing, with a smaller
435 volume of solid phase replacing the solute-rich liquid. Supposing that the globular and dendritic
436 grains pack at the same *envelope fraction*, the ratio of *solid fraction* in the packed layer between
437 dendritic and globular grains is of the order of g_{ip} , the internal solid fraction of the dendritic grains at
438 the packing front. The difference in the segregation induced by settling can be seen in the average
439 concentration of the sedimentation layer at the packing front, which is much lower in the case of
440 globular grains (Fig. 9a) than in the case of dendritic grains (Fig. 9b).

441

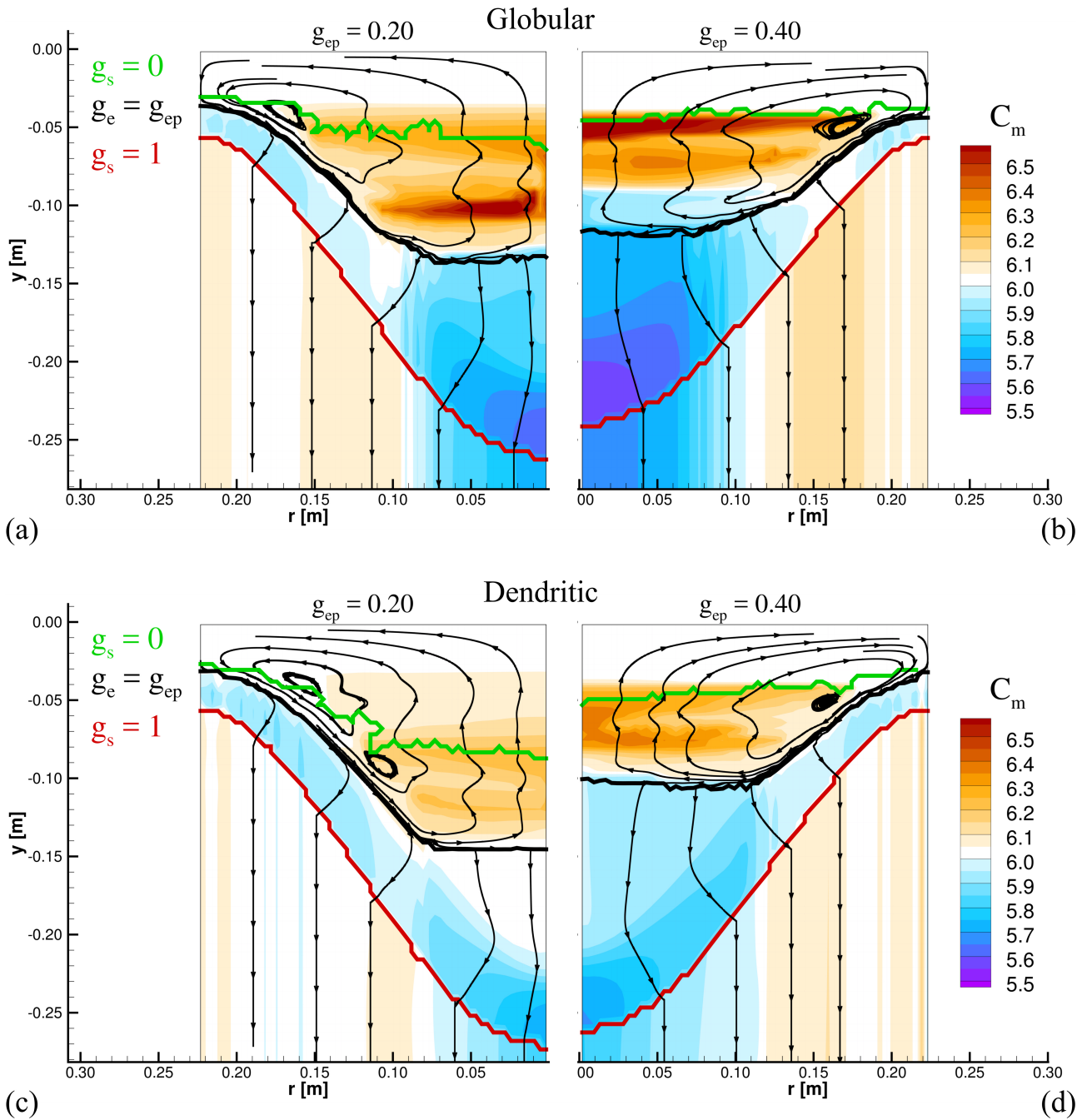


Figure 9: Macrosegregation of copper in the billet. Fields of average concentration for packing fractions of $g_{ep} = 0.20$ and $g_{ep} = 0.40$ and both dendrite tip models: (a) paraboloidal tip model and (b) hemispherical tip model.

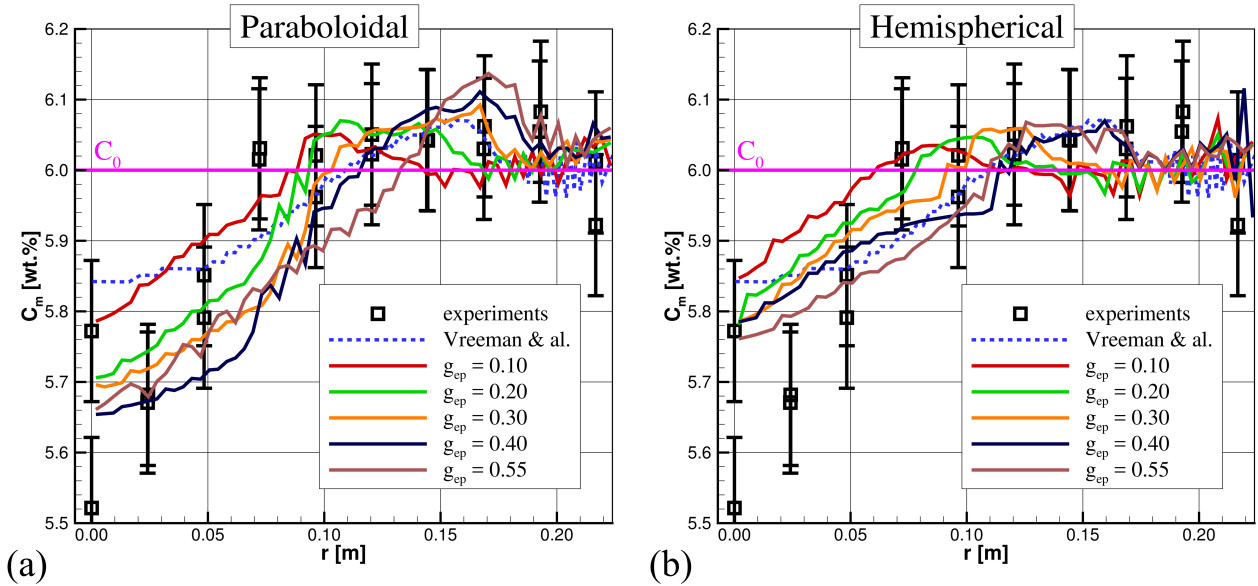


Figure 10: Macrosegregation in the grain-refined billet. Profiles of copper concentration from simulations with different packing fractions and both dendrite tip models: (a) paraboloidal tip model and (b) hemispherical tip model. The model predictions are compared to the experimental profiles and to simulations of Vreeman et al. [14].

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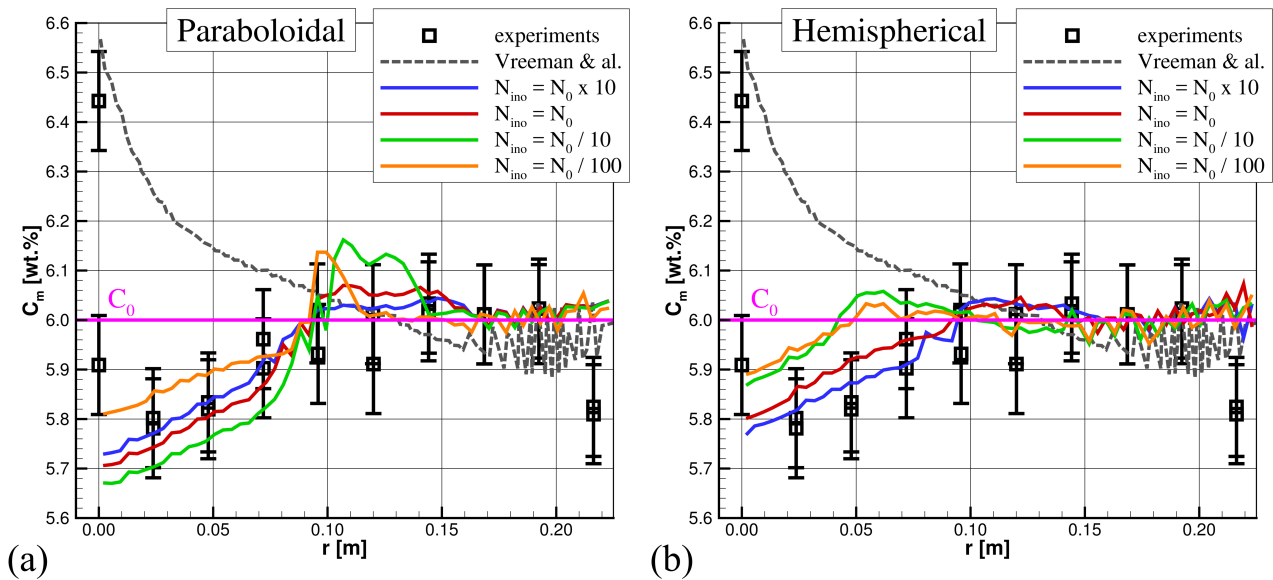


Figure 11: Macrosegregation in the non grain-refined billet. Profiles of copper concentration from simulations with different nucleation densities and both dendrite tip models: (a) paraboloidal tip model and (b) hemispherical tip model. The model predictions are compared to the experimental profiles and to simulations of Vreeman et al. [14]. N_0 refers to the distribution of nucleation sites shown in Fig. 3.

444

445 A further effect of the morphology is the drastic influence on the hydrodynamic permeability of the
446 packed layer. The more *closely* packed globular grains result in a packed layer with a solid fraction
447 of g_{ep} . This packed zone is less permeable than the *loosely* packed layer of dendritic grains, whose
448 solid fraction is lower, i.e., $g_{ip} g_{ep}$. The flow of intergranular liquid in the packed layer is a result of a
449 balance between buoyancy forces, suction due to solidification shrinkage, and drag forces exerted by
450 the matrix of packed grains. Inertial forces become negligible and in steady state the force balance
451 can be approximated by the Darcy equation: $(g_l^2 \mu_l / K)(\langle \bar{v}_l \rangle^l - \langle \bar{v}_s \rangle^s) \approx -g_l \nabla p + \rho_l^b g_l \vec{g}$. At low
452 permeability, such as in the closely packed globular layer, the drag forces increase, making the
453 buoyancy negligible. The percolation through the packed layer is then governed by a subtle interplay
454 of drag and solidification shrinkage and its trajectory largely depends on the shape of the field of g_l
455 (the shape of the packed layer and the progression of solidification within). In a dendritic packed
456 layer the permeability at the packing front is larger, which allows the buoyancy forces to exert an
457 influence on the flow up to a certain depth into the packed layer. This modifies the trajectory of the
458 liquid through the mushy zone and thus impacts the macrosegregation, generally diminishing the
459 negative segregation in the center [1]. Our interpretations are based on simulations that use the same
460 characteristic microstructure length for the permeability ($l_c = 75 \mu\text{m}$ in Eq. (11)) for globular and for
461 dendritic packed layers. This is done due to lack of accurate information on the grain size and the
462 dendrite spacing from the experiment. It is not clear whether this assumption is always reasonable.
463 The characteristic length for permeability is often approximated by the secondary dendrite arm
464 spacing (SDAS) in dendritic layers and by the grain size in globular layers. Dendritic grains are larger
465 than globular grains, but if their SDAS is very fine (which is not the case for coarse dendrites often
466 encountered in DC casting), the characteristic length for permeability in a dendritic packed layer can
467 be smaller than that of the globular packed layer. Therefore, the permeability of a dendritic packed
468 layer is not necessarily larger.

469 The effect of the packing fraction is similar for both grain morphologies (Fig. 10). An increase of the
470 packing fraction leads to an amplification of the negative centerline segregation and to a shift of the
471 positive segregation towards the billet surface. More extensive settling of solute-lean grains amplifies
472 the negative segregation in the ingot center. In reaction, it enhances the positive segregation in the
473 rest of the billet. However, due to the cylindrical geometry the amplification of the positive
474 segregation is less marked in the concentration profile. The shift of the positive segregation towards
475 the ingot surface is linked to the modification of the flow pattern. The raise of the packing front
476 displaces the main flow loop towards the billet surface (Fig. 9a–b and c–d) and thus impacts the flow
477 that is responsible for the macrosegregation formation.

478 The packing fraction impacts on the flow in two ways. First, the flow intensity increases due to an
479 enhanced sedimentation of the solid grains, which drag along the liquid. When the packing fraction
480 varies from 0.20 to 0.40, the maximum liquid velocity increases from 1.2 cm/s to 1.6 cm/s for globular
481 grains and from 0.09 cm/s to 1.5 cm/s for dendritic grains. The enhanced sedimentation is mainly due
482 to a larger grain size. Because the grains grow up to a higher envelope fraction before packing, the
483 equivalent grain diameter at packing increases from 125 to 150 μm for globular grains and from 140
484 to 170 μm for dendritic grains. Because the ratio of buoyancy to drag forces increases with the grain
485 size, the settling velocity thus increases with the packing fraction. At the same equivalent diameter,
486 the settling velocity of dendritic grains is smaller than that of globular grains due to the larger drag.
487 The second effect of the grain packing fraction concerns the region where the liquid flow develops.
488 Delaying the packing in terms of grain fraction does not imply a deepening of the sump as could
489 possibly be expected. Instead, a global upward shift of the whole mushy zone occurs due to enhanced
490 cooling and due to enhanced motion of grains to the center. The depth of both the mushy zone and of
491 the liquid zone decrease. It is interesting to note that this decrease is more pronounced with globular

492 grains. The reason is a larger enhancement of the heat transfer due to a more intense transport of the
493 solid phase. Conversely, the decrease of the liquidus depth is stronger with dendritic grains, because
494 a larger portion of the mushy zone is affected by the modification in the packing fraction (the grains
495 globularize, this means that the increase of the envelope fraction is slower than the increase of the
496 solid fraction).

497 Overall, the best agreement of the simulations with the experiment on the grain refined billet is
498 obtained with the paraboloidal tip model (predicting globular grains) and with an *envelope* packing
499 fraction of $g_{ep} = 0.20$. This holds for all compared characteristics: the segregation profile, the thermal
500 field and the sump geometry. This packing fraction is smaller than the *solid* packing fraction of $g_{sp} =$
501 0.30 that was used in previous simulations of this billet [14]. Vreeman et al. commented that a better
502 fit for the sump shape would be obtained with a solid packing fraction lower than 0.30 . Note that the
503 packing fractions used in both model studies are much lower than the maximum value for random
504 close packing of monodisperse spheres, which is around 0.65 [46,47]. Values of solid fraction at
505 packing usually used in DC casting modeling range from 0.15 to 0.3 [1,22,26,45,48,49]. It is also
506 interesting to note that in the Al-6 wt%Cu alloy, the density of the liquid phase becomes higher than
507 the solid density when the solid fraction increases above 0.4 . Thus the grains have the tendency of
508 floating instead of settling. It is possible that the packing of the grains is affected by this inversion of
509 the density difference.

510

511 4.4. Influence of the amount of grain refiner on the macrosegregation and 512 discussion on the non grain refined billet

513 In DC casting, inoculants are usually added to the melt in order to refine the grain size. The absence
514 of grain refinement affects the microstructure as well as the macrosegregation. Generally,
515 experimental observations have shown an increase of grain size, a dendritization of the grain
516 morphology and weaker macrosegregation at the centerline [2,12,17,19]. In the billet analyzed by
517 Vreeman et al. [14], the removal of grain-refiner led to a slight decrease of macrosegregation. This
518 can be seen by comparison of the experimental profiles shown in Figs. 10 and 11. Although
519 characterizations of the microstructure have not been performed in the experiments, we can
520 reasonably assume that in a non grain refined ingot the average grain size is larger and the grain
521 morphology is more dendritic. The model should be able to reproduce such a transition of
522 microstructure and macrosegregation between a refined and a non-refined casting.

523 The modeling of a non grain refined casting is not straightforward. Vreeman et al. assumed a fixed
524 solid phase in their simulations of the non grain refined billet. As seen in Fig. 11, this strategy gave
525 incorrect results. The assumption of a fixed solid would be adequate for a columnar microstructure.
526 However, the measured macrosegregation profiles as well as other experiments on non grain refined
527 DC castings [12,18,50] indicate that the microstructure remains mostly equiaxed even in the absence
528 of grain refinement. The present model is well adapted for describing heterogeneous athermal
529 nucleation on inoculant particles with a known size distribution. In the absence of inoculant particles
530 the population and the distribution of nucleation sites are not known. We tried to reproduce the
531 influence of the grain refiner by altering the total population of the inoculant particles in the
532 simulations. The nucleation was thus still described by the athermal heterogeneous nucleation model,
533 but with a smaller number of nucleation sites. The level of refiner addition was varied by factors 10
534 with respect to the nominal refiner density of $N_0 = 1$ kg/t. Four levels of grain refinement were
535 simulated: 10 kg/t ($N_0 \times 10$), 1 kg/t (N_0), 0.1 kg/t ($N_0/10$) and 0.01 kg/t ($N_0/100$). The same type of
536 particle size distribution (as shown in Fig. 3) was used; only the population density of all inoculant

537 particle classes was modified by the same factor. The envelope packing fraction was set to $g_{ep} = 0.20$
 538 in all simulations, since it gave the most accurate results for the grain-refined billet. The goal is not
 539 only to check whether the model is able to reproduce the experimentally observed macrosegregation
 540 in the non-refined billet, but also to analyze the effect of the refiner level on the structure.

541 Figs. 12a and 12b provide information about the microstructure in the billet predicted by our model.
 542 They show the effect of the refiner addition level on the average grain size (represented by the mean
 543 final equivalent grain diameter, \bar{d}_s) and on the grain morphology (represented by the mean internal
 544 solid fraction at the packing front, g_{ip}), as predicted by both dendrite tip models. The mean final
 545 equivalent grain diameter is calculated as the equivalent grain diameter after complete solidification
 546 and by excluding the eutectic fraction:

$$\bar{d}_s = \left(\frac{6(1-\bar{g}_{eut})}{\pi \bar{N}_s} \right)^{1/3}, \quad (13)$$

547 where \bar{g}_{eut} is the mean eutectic volume fraction predicted by the model and \bar{N}_s is the mean grain
 548 density. The effect of the inoculant density (grain refiner level) is similar for the two dendrite tip
 549 models: a reduction of the refiner level leads to an increase of the grain size and to a more dendritic
 550 morphology. On the opposite, an increase of the refiner level leads to smaller and more globular
 551 grains. However, important differences remain in the microstructure predictions, depending on the
 552 tip model used. The refiner level has a greater impact on the microstructure when the hemispherical
 553 model is considered, both in terms of grain size and morphology. With the hemispherical tip growth
 554 model, grains are already dendritic for the highest refiner level, and become even more dendritic
 555 when the refiner level is lowered. With the paraboloidal tip growth model, only the lowest refiner
 556 level leads to a significant reduction of the mean internal fraction. With higher refiner levels grains
 557 remain globular.

558

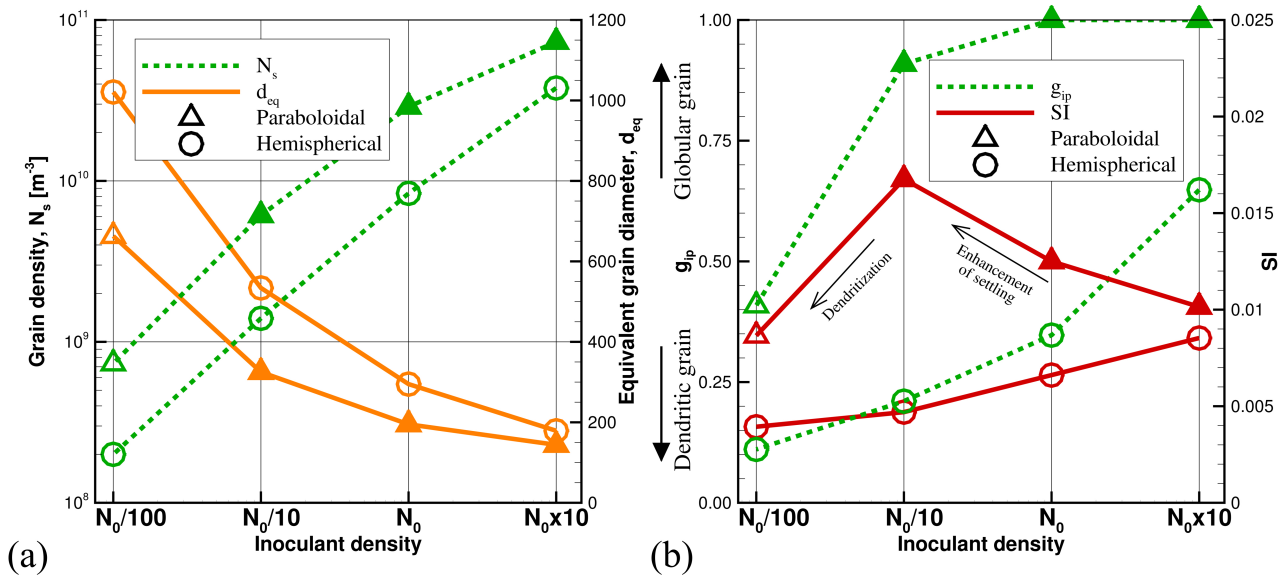


Figure 12: Microstructure and macrosegregation as a function of the nucleation density, predicted by both dendrite tip models. N_0 refers to the distribution of nucleation sites shown in Fig. 3. (a) Mean final grain density and mean equivalent grain diameter. (b) Mean internal solid fraction at packing (g_{ip}) and mean segregation index. Full symbols indicate globular and empty symbols dendritic grain morphology.

559 Fig. 12a also shows the evolution of the mean final grain density as a function of the grain-refiner
 560 level. We can see that the dependence is not linear for either tip model. The nonlinearity is particularly
 561 strong with the paraboloidal model in the domain of globular grain morphologies (i.e. for inoculant
 562 densities of $N_0/10$ and greater). This is a result of the complex coupling between the nucleation, the
 563 kinetics of grains growth and the transport of nucleant particles and grains [28,29,51]. When the
 564 number of nucleation sites is reduced, the consequence is that a smaller number of grains nucleate at
 565 a given undercooling. The rejection of solute into the liquid during their growth is less efficient and
 566 therefore the enrichment of the surrounding liquid is slower. The constitutional undercooling thus
 567 keeps increasing. As a consequence, nucleation sites requiring larger undercooling for their activation
 568 are activated. The reduction of the grain density is therefore not proportional to the reduction of the
 569 nuclei density but is less. This effect is stronger with globular grains due to the stronger coupling with
 570 grain transport [28].

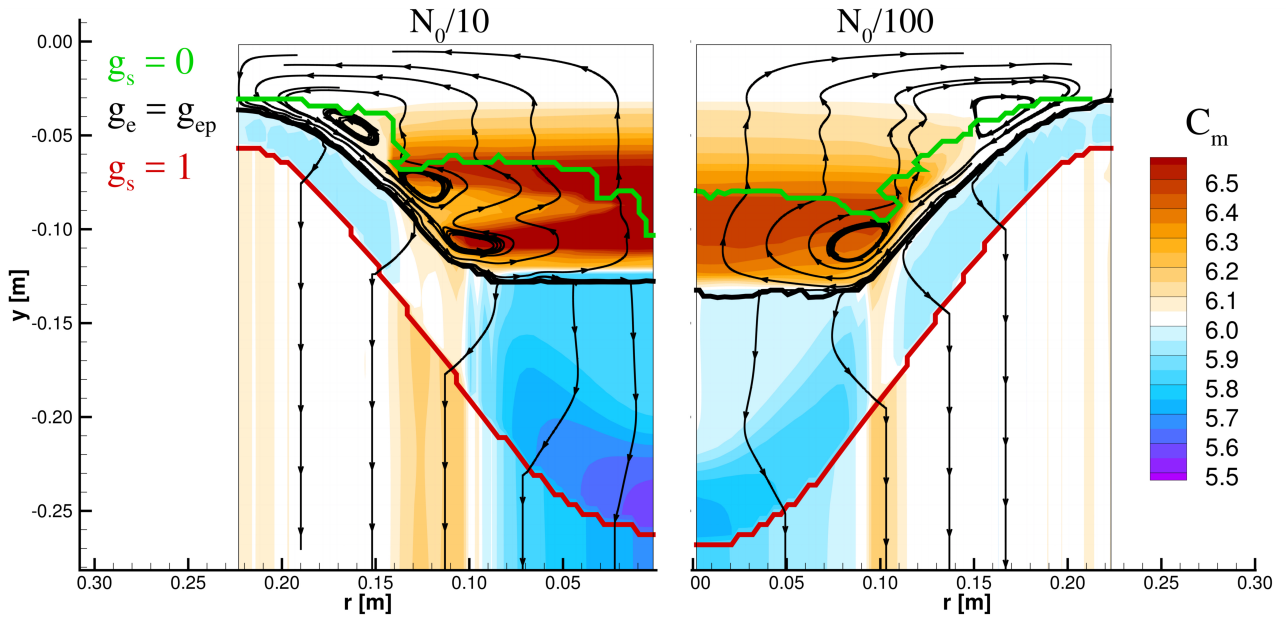
571 The intensity of macrosegregation can be quantified in a compact form as a segregation index, SI .
 572 The segregation index is defined as the standard deviation of the macrosegregation across the billet
 573 radius:

$$SI = \sqrt{\frac{2}{R_b^2} \int_0^{R_b} (C - C_0)^2 r dr}, \quad (14)$$

574 The segregation indices for all simulations and their correlation with the grain morphology are shown
 575 in Fig. 12b. A comparison of the results obtained with both tip models indicates that the intensity of
 576 the macrosegregation is generally higher when the grains are more globular. This is further supported
 577 by the abrupt decrease of the SI upon the morphology transition predicted by the paraboloidal tip
 578 model. We can also see that with increasing grain size the SI increases, for globular grains whereas
 579 it decreases for dendritic grains.

580 We can take a closer look at what happens if we analyze the flow and the grain and solute transport
 581 in the sump. Fig. 13 shows the effect of the morphology transition on the macrosegregation. It shows
 582 the concentration field in the billet for nucleation-site densities of $N_0/10$ and $N_0/100$, corresponding
 583 to the transition from globular to dendritic morphology with the paraboloidal tip model. When the
 584 nucleation density is reduced from N_0 down to $N_0/10$ (compare Fig. 9a to Fig. 13a), the segregation
 585 is enhanced everywhere in the billet. The main reason is that the grains, which remain globular, ($\bar{g}_{ip} \approx$
 586 0.9) are almost twice larger. The effect of the grain settling is therefore increased. This is shown
 587 quantitatively in Fig. 14, where the difference of the velocity between the solid and the liquid in the
 588 slurry zone is plotted against the local grain size. We can see that across the whole range of nucleation
 589 densities the settling velocity globally increases with increasing grain size. When the grains are very
 590 small (around 80 μm in Case $N_0 \times 10$), their velocity is close to that of the liquid and thus
 591 macrosegregation is reduced. Larger grains settle faster, therefore the negative centerline
 592 macrosegregation increases when the nuclei density is reduced down to $N_0/10$. When a transition to
 593 dendritic morphology ($\bar{g}_{ip} \approx 0.4$) is triggered by further lowering the nuclei density to $N_0/100$ (Fig.
 594 13b), the segregation is suddenly strongly reduced. It is interesting to note that the flow pattern
 595 remains very similar. Although the settling velocity further increases (Fig. 14), the expulsion of
 596 enriched liquid at the packing is lessened due to the smaller solid fraction at the packing front. This
 597 effect is shown in detail in Fig. 15 that displays the evolution of the mean concentration of copper
 598 and of the envelope and solid fractions along the billet centerline for cases with $N_0/10$ (globular) and
 599 $N_0/100$ (dendritic). We can see that with $N_0/10$ (Fig. 15a) the intense settling of globular grains results
 600 into a large concentration jump and a negative centerline segregation ($C_m \approx 5.8$) at the packing front.
 601 The negative segregation is then exacerbated mainly by the shrinkage flow through the packed layer.
 602 With $N_0/100$ (Fig. 15b), the accumulation of dendritic grains at the packing front does not cause any

603 significant segregation. The average concentration at the packing front is close to 6% – the nominal
 604 alloy concentration. The final negative centerline segregation is caused mainly by the shrinkage flow.
 605 This shows that the morphology transition reduces the influence of the grain packing on the
 606 macrosegregation pattern.



(a) Globular grains

(b) Dendritic grains

Figure 13: The influence of the morphology transition on the macrosegregation. Fields of the average copper composition, (a) $N_{ino} = N_0/10 = 0.1$ kg/t, (b) $N_{ino} = N_0/100 = 0.01$ kg/t. Both simulations are with the paraboloidal dendrite tip model.

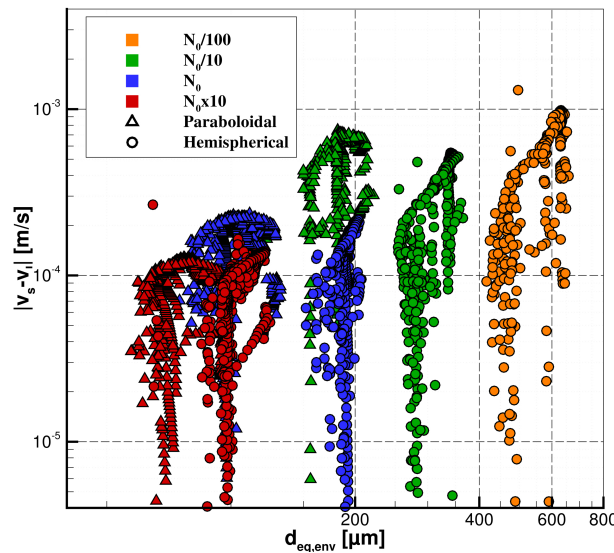


Figure 14: The slip velocity of the equiaxed grains in the slurry zone as a function of the local equivalent grain diameter, $d_{eq,env}$. Colored markers indicate a downward oriented slip velocity (settling) and (scarce) black markers indicate an upward oriented slip velocity (flotation). N_0 refers to the distribution of nucleation sites shown in Fig. 3.

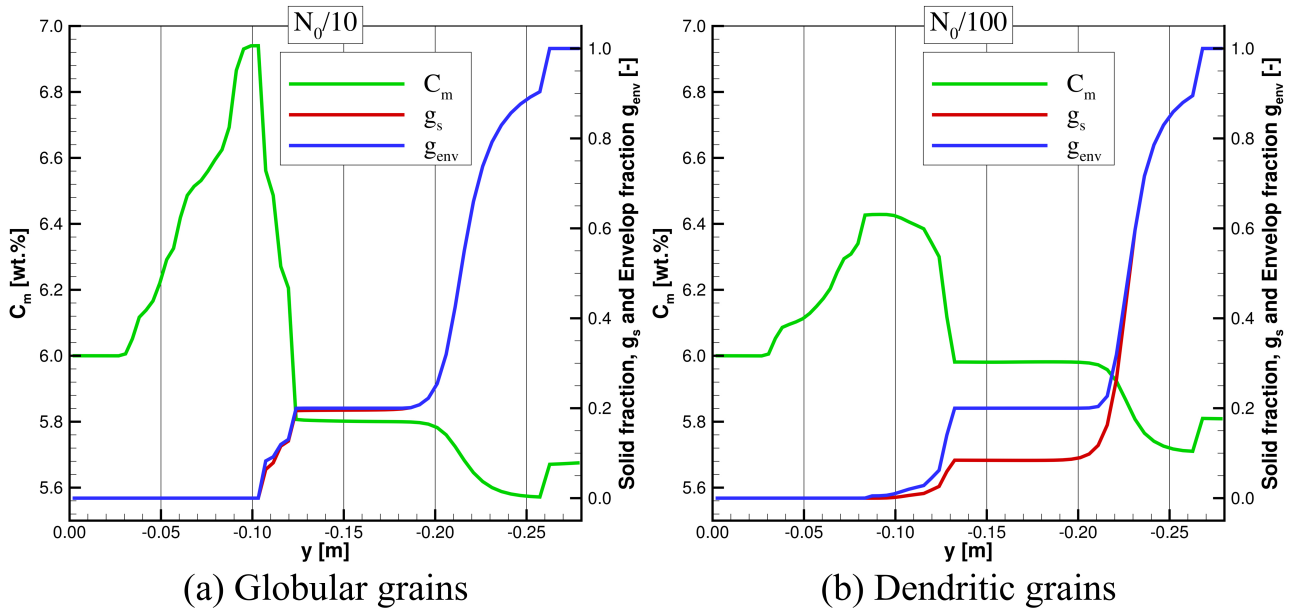


Figure 15: The formation of the centerline macrosegregation with different grain morphologies. Evolution of the average copper composition C_m , solid fraction g_s and envelope fraction g_e along the billet centerline for (a) $N_0/10$ and (b) $N_0/100$, both obtained with the paraboloidal tip model.

607 Macro-segregation profiles for all nucleation densities are presented in Fig. 11a, for the paraboloidal
 608 model and in Fig. 11b for the hemispherical model. The evolution of the profiles as a function of the
 609 nucleation density is consistent with the microstructure evolution shown in Fig. 12. With the
 610 paraboloidal dendrite tip model the grain size increases and the morphology remains globular for
 611 nucleation densities from $N_0 \times 10$ down to $N_0/10$. This leads to an amplification of the negative
 612 centerline macrosegregation. The segregation intensity is reduced only for the extreme case of $N_0/100$,
 613 where the grains become dendritic. With the hemispherical dendrite tip model, the segregation
 614 intensity decreases all along as the density of nucleation sites is reduced. The main reason is the
 615 diminished grain settling due to the larger drag on the grains with a more pronounced dendritic
 616 morphology. In the extreme case of $N_0/100$, the solid fraction at packing is only around 2 %.

617 Overall, our results indicate that the absence of grain refinement triggers a modification of the grain
 618 morphology from globular to dendritic. This morphology transition induces a weakening of the
 619 macrosegregation in the whole casting. The main reason is that the smaller solid fraction of the
 620 dendritic grains causes less expulsion of the solute-enriched liquid at the packing front. This trend is
 621 consistent with the experimental observations in the two billets and is predicted with a correct order
 622 of magnitude.

623

624 5. Conclusions

625 We performed a detailed study of the formation of macrosegregation in industrial size DC cast billets
 626 using a multiphase and multiscale model. This model stands out by the careful description of the
 627 intricate couplings between the kinetics of microstructure formation and the macroscopic transport
 628 of heat, mass, momentum and solute. It represents the nucleation, the growth kinetics and the transport
 629 of equiaxed grains; all taking into account the transitions between globular and dendritic grain
 630 morphologies. The model study was supported by previously published experimental results, which
 631 were used as a reference.

632 Predictions of radial macrosegregation profiles, of lengthwise temperature profiles, and of the shape
633 and the dimensions of the mushy zone were compared to the experimental measurements in a grain
634 refined and a non grain refined billet. A sensitivity study with respect to the grain packing fraction
635 was performed, in order to fit this poorly known but key model parameter. It showed that the rather
636 low packing fraction of 0.2 clearly captures best both the heat transfer and the macrosegregation in
637 the billets.

638 While the grain refiner addition level in the grain refined billet was known, the absence of grain
639 refiner in the non grain refined billet was modeled by lowering the population density of nucleation
640 sites. These simulations indicate that a morphology transition triggered by the grain refinement is the
641 key to the explanation of the differences of the macrosegregation patterns in the two billets. In the
642 grain refined billet, globular grain morphologies clearly gave better predictions of the thermal field
643 and of the macrosegregation than dendritic morphologies. The reduction of the macrosegregation in
644 the non grain refined billet was caused by a transition from globular to dendritic grain morphologies.
645 It was shown that a globular grain morphology promotes the settling of equiaxed grains and thus
646 exacerbates the associated transport of solute. It results into more pronounced negative centerline
647 segregation than a dendritic grain morphology.

648 Previous work has shown that the choice of the dendrite tip model is not unambiguous when
649 describing coarse dendritic morphologies. Two different dendrite tip models were therefore used to
650 model the grain morphology and they gave disparate predictions. In the same solidification
651 conditions, the hemispheric tip model tends to predict more dendritic grain morphologies than the
652 paraboloidal tip model. The hemispheric tip model indicated dendritic grains in both the grain refined
653 and the non grain refined billet, thus failing to capture a morphology transition and the associated
654 reduction in macrosegregation. Only the paraboloid tip model was able to capture the transition from
655 globular to dendritic grains as a function of grain refinement and to quantitatively reproduce its
656 influence on the macrosegregation. A proper choice of the nucleation density was required, reducing
657 the number of nucleation sites by around 100 times with respect to the grain refined case.

658 The formation of macrosegregation in DC casting is a result of a subtle interplay of several flow
659 phenomena: thermosolutal convection, grain motion and packing, shrinkage-induced flow, and flow
660 induced by thermomechanical deformation. The prominence of one or another of these phenomena
661 may strongly depend on the casting size and shape (billet or ingot), on the alloy composition, on the
662 mold type, and on the process parameters (casting speed, temperature, grain refinement). This work
663 thus cannot provide a generally valid explanation of the macrosegregation mechanism in DC casting.
664 It does however make a large step forward to a complete description of DC casting at the process
665 scale, accounting for the coupling of microstructure and macrosegregation. Further work should focus
666 on the refinement of the microstructure model, in order to ensure a quantitative prediction of the
667 morphology transition between globular and dendritic grains. This is particularly important in the DC
668 casting process, since the observed morphology of the grains is often coarse dendritic, i.e. close to
669 the transition. A more extensive comparison of the models to well characterized industrial-scale
670 experiments and application to multi-component alloys are also required.

671

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768 Appendix A

769 In the three-phase solidification model, the growth kinetics of the equiaxed grains is described by
 770 solute balances over the three hydrodynamic phases and on two moving interfaces, the solid–
 771 interdendritic-liquid interface and the envelope–extradendritic-liquid interface. The volume averaged
 772 mass transfer rate per unit volume from the interdendritic liquid to the solid due to phase change at
 773 the solid-liquid interface is denoted Γ_s . The mass transfer rate from the extradendritic liquid to the
 774 envelope at the envelope-liquid interface is Γ_e . The solid-liquid interface is at equilibrium and the
 775 equilibrium concentrations of solid and liquid are linked by the partition coefficient: $C_s^* = kC_l^*$. The
 776 interdendritic liquid is also entirely at the liquid equilibrium concentration C_l^* . The solute transfer
 777 rates due to this interfacial growth are $\Gamma_s C_s^*$ and $-\Gamma_s C_l^*$ at the solid and liquid sides of the solid-liquid
 778 interface, and $\Gamma_e C_l^*$ and $-\Gamma_e C_l^*$ at the exterior and the interior sides of the envelope-liquid interface,
 779 respectively. The diffusion fluxes from the solid-liquid interface into the solid phase, j_s^d , and from
 780 the envelope into the extradendritic liquid, j_l^d , are modeled as:

$$j_s^d = \frac{\rho_s S_v^s D_s}{\delta_s} (C_s^* - \langle C_s \rangle^c) \quad j_l^d = \frac{\rho_l S_v^e D_l}{\delta_e} (C_l^* - \langle C_l \rangle^e) \quad (15)$$

781 The diffusion length in the solid is $\delta_s = d_s/10$, where d_s is the equivalent diameter of the solid. The
 782 diffusion length in the liquid, δ_e , is modeled by an expression that accounts for the effects of liquid
 783 convection and of a moving interface and was derived in [52]. The diffusion fluxes from both
 784 interfaces into the interdendritic phase, j_{is}^d and j_{ie}^d , become indeterminate under the assumption of
 785 infinite diffusion in the extradendritic liquid. The solute balance at the solid–interdendritic-liquid
 786 interface is

$$\Gamma_s C_s^* + \frac{\rho_s S_v^s D_s}{\delta_s} (C_s^* - \langle C_s \rangle^s) = \Gamma_s C_l^* - j_{is}^d \quad (16)$$

787 The solute balance at the envelope–extradendritic-liquid interface is

$$\Gamma_e C_l^* + j_{ie}^d = \Gamma_e C_l^* - \frac{\rho_l S_v^e D_l}{\delta_e} (C_l^* - \langle C_l \rangle^{le}) \quad (17)$$

788 To avoid the indeterminate terms we can write down the sum of both interfacial balances as the solute
 789 balance for the interdendritic phase [35]:

$$\frac{\partial}{\partial t} (\rho_l g_{li} C_l^*) = \Gamma_e C_l^* - \Gamma_s C_s^* - \frac{\rho_l S_v^e D_l}{\delta_e} (C_l^* - \langle C_l \rangle^{le}) - \frac{\rho_s S_v^s D_s}{\delta_s} (C_s^* - \langle C_s \rangle^s). \quad (18)$$

790 We take into account that $g_{li} = g_e - g_s$ and $g_l = 1 - g_s$, that $g_l \langle C_l \rangle^l = g_{le} \langle C_l \rangle^{le} + g_{li} C_l^*$ and we
 791 obtain

$$\frac{\rho_s S_v^s D_s}{\delta_s} (kC_l^* - \langle C_s \rangle^s) + \left(\frac{1-g_s}{1-g_e} \right) \frac{\rho_l S_v^e D_l}{\delta_e} (C_l^* - \langle C_l \rangle^l) = \Gamma_s (1-k) C_l^* - \rho_l (g_e - g_s) \frac{\partial C_l^*}{\partial t}. \quad (19)$$

792 This equation is solved in conjunction with:

- 793 • the heat conservation equation that gives the evolution of the local average enthalpy, defined
 794 as $\bar{h} = c_p T + (1 - g_s) L_f$;
- 795 • the phase diagram that gives the link between the equilibrium liquid concentration and the
 796 temperature: $T = T_m + m C_l^*$;
- 797 • and a dendrite-tip model that gives the velocity of the envelope growth as a function of the
 798 local supersaturation $(C_l^* - \langle C_l \rangle^{le})$ and thus enables us to calculate the envelope fraction g_e .

799 This gives four equations for the four additional unknowns Γ_s , C_l^* , T and g_e .

800 Several supplementary geometric relations are required. The grain envelopes are assumed to be
 801 regular octahedra, the distance between the center and the vertices of the octahedron corresponding

802 to the length of the primary dendrite arms. The primary arm length, R_{arm} , is calculated by integrating
803 the tip velocity over time. Knowing the primary arm length, the envelope fraction and the specific
804 surface of the envelope are:

$$g_e = \frac{4}{3} N_v R_{arm}^3 \quad \text{and} \quad S_v^e = 4\sqrt{3} R_{arm}^2 N_v, \quad (20)$$

805 where N_v is the grain population density. The specific surface of the solid, S_v^s , is described by a
806 generalized correlation that continuously switches between a relation for a strongly dendritic grain,
807 in which the surface area is controlled by the secondary dendrite arm spacing, λ_2 , and a relation for a
808 globular grain, where $S_v^s = S_v^e$ [31]:

$$S_v^s = \frac{4}{\lambda_2} g_i^{1/2} g_e (1 - g_i^m) + g_i^m S_v^e. \quad (21)$$

809 The interpolation is done by the internal solid fraction, $g_i = g_s/g_e$, and the exponent m was set to 6
810 [38]. The secondary dendrite arm spacing is approximated as be a fraction of the envelope radius:
811 $\lambda_2 = R_e/5 = R_{arm}/(5\pi^{1/3})$.

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813

814 **Figure captions**

815

Figure 1: (a) Schematics of the hydrodynamic phases in the three-phase model. (b) Solute profiles in the three phases, solute concentrations and volume fractions.

816

Figure 2: Evolution of solid, liquid and average densities of the alloy Al–6wt.%Cu with solid fraction. The solidification path is assumed to follow the Scheil-Gulliver model. The variation of the solid density due to the appearance of a secondary phase upon the eutectic reaction is not accounted for.

817

Figure 3: Discretization of the distribution of nucleation sites with respect to the critical undercooling. The distribution corresponds to the size distribution of TiB₂ particles for 1 kg/t of Al-5Ti-1B grain refiner [28,44].

818

Figure 4: Heat transfer and flow in the billet: (a) temperature field, (b) solid fraction, (c) norm of the liquid velocity and liquid velocity streamlines, (d) norm of the solid velocity and solid velocity streamlines. The grain refined billet simulated with a packing fraction of $g_{ep}=0.2$ and the paraboloidal dendrite tip model is shown. The relative velocities are shown in the reference frame of the moving product.

Figure 5: Internal solid fraction in the mushy zone obtained with both dendrite tip models: (a) paraboloidal, (b) hemispherical tip model. The packing fraction is $g_{ep} = 0.20$ in both cases.

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Figure 6: Internal solid fraction at packing (g_{ip}) for both tip growth models, hemispherical (solid lines) and paraboloidal (dashed lines).

820

Figure 7: Comparison of measured and computed vertical temperature profiles at three different radius in the ingot (subsurface in blue, midradius in green and centerline in red). (a) Comparison of simulations with globular grains (paraboloidal tip model) and $g_{ep}=0.2$ with experiments and with simulations of Vreeman et al. [14]. (b) Influence of the packing fraction on the temperature profiles for globular grain morphology (paraboloidal tip model). (c) Influence of the packing fraction on the temperature profiles for dendritic grain morphology (hemispherical tip model).

821

Figure 8: (a) Comparison of the sump shape obtained with the paraboloidal tip growth model and $g_{ep} = 0.20$ to the experimental sump shape. (b) Relative error of the lengths L1 and L2 as a function of the packing fraction, shown for simulations with both tip models.

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Figure 9: Macrosegregation of copper in the billet. Fields of average concentration for packing fractions of $g_{ep} = 0.20$ and $g_{ep} = 0.40$ and both dendrite tip models: (a) paraboloidal tip model and (b) hemispherical tip model.

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Figure 10: Macrosegregation in the grain-refined billet. Profiles of copper concentration from simulations with different packing fractions and both dendrite tip models: (a) paraboloidal tip model and (b) hemispherical tip model. The model predictions are compared to the experimental profiles and to simulations of Vreeman et al. [14].

824

Figure 11: Macrosegregation in the non grain-refined billet. Profiles of copper concentration from simulations with different nucleation densities and both dendrite tip models: (a) paraboloidal tip model and (b) hemispherical tip model. The model predictions are compared to the experimental profiles and to simulations of Vreeman et al. [14]. N_0 refers to the distribution of nucleation sites shown in Fig. 3.

825

Figure 12: Microstructure and macrosegregation as a function of the nucleation density, predicted by both dendrite tip models. N_0 refers to the distribution of nucleation sites shown in Fig. 3. (a) Mean final grain density and mean equivalent grain diameter. (b) Mean internal solid fraction at packing (g_{ip}) and mean segregation index. Full symbols indicate globular and empty symbols dendritic grain morphology.

826

Figure 13: The influence of the morphology transition on the macrosegregation. Fields of the average copper composition, (a) $N_{ino} = N_0/10 = 0.1$ kg/t, (b) $N_{ino} = N_0/100 = 0.01$ kg/t. Both simulations are with the paraboloidal dendrite tip model.

827

Figure 14: The slip velocity of the equiaxed grains in the slurry zone as a function of the local equivalent grain diameter, $d_{eq,env}$. Colored markers indicate a downward oriented slip velocity (settling) and (scarce) black markers indicate an upward oriented slip velocity (flotation). N_0 refers to the distribution of nucleation sites shown in Fig. 3.

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Figure 15: The formation of the centerline macrosegregation with different grain morphologies. Evolution of the average copper composition C_m , solid fraction g_s and envelope fraction g_e along the billet centerline for (a) $N_0/10$ and (b) $N_0/100$, both obtained with the paraboloidal tip model.

829 **Table captions**

830

Table 1: Summary of the model equations

Table 2: Thermo-physical properties and model parameters

831

832 **Nomenclature**

Latin symbols

C_0	Nominal solute concentration
C_d	Drag coefficient
C_k^*	Solute concentration at the solid-liquid interface in phase k
$\langle C_k \rangle^k$	Average solute concentration in phase k
C_m	Average “mixture” concentration
c_p	Specific heat
C_{ref}	Reference solute concentration for the liquid density
d_g	Equivalent grain envelope diameter
D_k	Solute diffusion coefficient in phase k
d_s	Equivalent diameter of the solid phase
\bar{d}_s	Mean final equivalent grain diameter
\vec{g}	Gravity acceleration
g_{ep}	Packing grain-volume fraction
g_{ep}^{max}	Maximum envelope packing fraction
\bar{g}_{eut}	Mean final eutectic volume fraction
g_i	Internal solid volume fraction
g_{ip}	Mean internal solid volume fraction at the packing front
g_k	Volume fraction of phase k
h_k	Mass enthalpy of phase k
j_k^d	Diffusion flux from the solid-liquid interface into phase k
k	Equilibrium partition coefficient
K	Permeability
k_k	Kozeny constant
L_1	Distance between the packing front and the solidus at mid-radius
L_2	Height of the solidus front
l_c	Characteristic length of the porous microstructure
L_f	Latent heat
m	Liquidus slope
M_k^d	Interfacial drag force on phase k per unit volume
M_k^r	Momentum flux to phase k due to phase change
M_k^Φ	Momentum transfer to phase k due to nucleation

N_0	Nominal population density of grain refiner particles
N_g	Grain density (number of grains per unit volume)
N_{nuc}^i	Density of i^{th} class of inoculant particles
\bar{N}_s	Mean final grain density
p	Pressure
Pe_C, Pe_T	Solutal, thermal Péclet number of the dendrite tip related to growth
Pr	Prandtl number
Pu_C, Pu_T	Solutal, thermal Péclet number of the dendrite tip related to convection
R_{arm}	Primary dendrite arm length
R_b	Billet radius
Re_C, Re_T	Solutal, thermal Reynolds number
R_{tip}	Dendrite tip radius
Sc	Schmidt number
$SDAS$	Secondary dendrite arm spacing
SI	Segregation index
S_v^e	Specific surface of the dendrite envelope
S_v^s	Specific surface of solid phase
t	Time
T	Temperature
T_m	Melting temperature of pure aluminum
T_{ref}	Reference temperature for the liquid density
ΔT	Undercooling
V_0	Initial grain volume at nucleation
$\langle \vec{v}_k \rangle^k$	Intrinsic average velocity of phase k
V_{tip}	Dendrite tip velocity

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Greek symbols

α	Thermal diffusivity
β_C	Solutal expansion coefficient for the liquid density
β_T	Thermal expansion coefficient for the liquid density
β_{sl}	Constant solidification shrinkage coefficient
Γ_k	Mass flux into phase k per unit volume at the interface due to phase change
Γ_{GT}	Gibbs-Thomson coefficient
δ	Dirac delta function
δ_e	Solute diffusion length in the extra-dendritic liquid phase
δ_s	Solute diffusion length in the solid phase
λ	Thermal conductivity
λ_2	Secondary dendrite arm spacing
μ_k	Dynamic viscosity of phase k
ρ_k	Density of phase k
ρ_l^b	Liquid density in the buoyancy term
ρ_{ref}	Reference density for the buoyancy term
σ^*	Tip selection constant
τ	Tortuosity of the porous medium
$\langle \tau_k \rangle^k$	Shear stress tensor in phase k
Φ_k	Mass flux into phase k per unit volume due to nucleation
ϕ_k	Mass flux due to nucleation
Ω_C	Dimensionless supersaturation
Ω_T	Dimensionless undercooling

835

Sub- and superscripts

e	Envelope
l	Liquid
le	Extradendritic liquid
li	Intradendritic liquid
s	Solid

836