Sensitivity of macroscopic transport calculations to uncertain microscale relationships during metal alloy solidification

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Abstract. The formation of grains during metallic solidification results in a multiphase system consisting of many moving interfaces. Models have been formulated in terms of variables describing the microscopic details of interface motion during solidification, but the computational cost prohibits their extension to the length scale of industrial castings. For this reason, macroscopic transport equations are mathematically formulated using averaging methods to replace the instantaneous description of interfaces. The most common approach is the volume-averaged, Eulerian approach relying on both the proper derivation of macroscopic field equations as well as uncertain interfacial conditions used to account for the average behavior of the interphase transfer occurring on the sub-grid length scale. In this work, common models used to describe the microsegregation occurring during solidification are evaluated in terms of the uncertainty of parametric inputs invoked in each model and their effect on macroscale predictions. We show that the qualitative macroscale predictions are not significantly changed by using more sophisticated microsegregation models, however significant uncertainty in input parameters used to describe the unresolved sub-grid interface is accumulated. Therefore, the use of simplified models is preferred until the uncertainty in model inputs can be reduced.

1. Introduction
Macrosegregation during alloy solidification is initiated by solute partitioning at the solid-liquid interface (microsegregation) and propagated by the relative motion between solid and liquid phases. Two different numerical modeling approaches have been used for calculating macrosegregation in alloy castings. The multiphase approach attempts to resolve the microscopic features of solidification in significant detail by solving partial differential equations for each phase transport term at the expense of computational efficiency and the introduction of highly uncertain model inputs [1, 2]. The continuum mixture approach [3] instead solves transport equations in terms of mixture quantities using explicit relationships to describe the individual phase transport when needed. While the microsegregation relations used in mixture models have historically been limited to simplified analytical expressions neglecting liquid undercooling (e.g. Gulliver-Scheil and Lever rule), this is not a general requirement.

In order to justify the use of more complicated microsegregation models it must be first demonstrated that these models more accurately capture the physical phenomena occurring during equiaxed solidification. Perhaps the weakest area of the volume-averaged microsegregation models lies in the interfacial relationships used to describe the evolution of equiaxed grain morphologies. Unfortunately for intermediate grain radii, of critical importance in industrial castings, there are currently no analytical solutions to fully describe the growth kinetics. Since this class of microsegregation models are based on incomplete closure relations with uncertain input parameters, simplification is often desired. Beckermann and co-workers [4] recently showed that the qualitative macrosegregation predictions...
during columnar solidification do not significantly change when a volume-averaged microsegregation model is replaced with the simplified Gulliver-Scheil equation. Instead, only the prediction of mesosegregates changed which is now known to be limited by the length scale of the numerical grid rather than the interfacial features which produce such variations in the physical system [5]. Nevertheless, a simplified truncated-Scheil model which includes liquid undercooling at the primary dendrite tips was able to reproduce the full volume-average model results in about half the computational time. Motivated by this study of columnar solidification, the primary goal of this work is to investigate the degree to which macrosegregation predictions change using a volume-average microsegregation model compared to a Gulliver-Scheil model during equiaxed solidification.

2. Microsegregation modeling
A modified form of the volume-averaged microsegregation model of dendritic solidification formulated by Tveito et al. [6] is used here. The average solid and liquid volume fractions ($g_s, g_l$) and compositions ($C_s, C_l$) are calculated using the conservation equations:

$$\frac{\partial}{\partial t} (\rho_s g_s) + \nabla \cdot (\rho_s g_s \mathbf{U}_s) = \dot{m}_s$$  \hspace{1cm} (1)

$$\frac{\partial}{\partial t} (\rho_s g_s C_s) + \nabla \cdot (\rho_s g_s \mathbf{U}_s C_s) = \dot{m}_s k_p C_l^*$$  \hspace{1cm} (2)

$$\frac{\partial}{\partial t} (\rho_l g_l C_l) + \nabla \cdot (\rho_l g_l \mathbf{U}_l C_l) = -\dot{m}_s k_p C_l^*$$  \hspace{1cm} (3)

where $\rho_k$ is the phase density, $\mathbf{U}_k$ the phase velocity vector, $\dot{m}_s$ the mass phase change rate due to solidification, $k_p$ the equilibrium partition coefficient, $C_l^*$ the composition of the interdendritic liquid, and $g_s + g_l = 1$. Due to the highly coupled nature of these nonlinear equation, splitting methods, where the contributions of phase transport and growth are considered separately, are commonly adopted [7, 8].

2.1. Transport stage
In the transport stage equations (1)-(3) are solved without the influence of growth. The left-hand side of these equations are solved explicitly using the following expressions:

$$g_s^{tr} = g_s^o - \Delta t \nabla \cdot (g_s^o \mathbf{U}_s^o)$$  \hspace{1cm} (1a)

$$C_s^{tr} = \frac{1}{g_s^o + \varphi_s} \left( (g_s^o + \varphi_s) C_s^o - \Delta t \nabla \cdot (g_s^{tr} \mathbf{U}_s^o C_s^o) \right)$$  \hspace{1cm} (2a)

$$C_l^{tr} = \frac{1}{g_l^{tr} + \varphi_l} \left( (g_l^o + \varphi_l) C_l^o - \Delta t \nabla \cdot (g_l^{tr} \mathbf{U}_l^o C_l^o) \right)$$  \hspace{1cm} (3a)

where $\varphi_k$ is a stabilization coefficient used to bound the solution in the dilute limits of phase $k$, i.e. $\varphi_k = \max (10^{-6} - g_k, 0)$, the superscript $o$ denotes the values at the previous time step $\Delta t$, and the superscript $tr$ denotes the resulting transport values calculated in this stage. The MULES algorithm (multi-dimensional limiter for explicit solution) currently implemented in OpenFOAM [9] is used for solving equation (1a). Using the updated values of $g_s^{tr}$ and $g_l^{tr}$, the solute advection equations are solved (equations (2a) and (3a)), and the resulting transport fields are stored for reference in the subsequent growth stage. Solving the transport equations explicitly introduces a strict Courant number limit, and hence time step limit, however no time step sub-cycling is required in the transport stage leading to an overall decrease in computation time.
2.2. Growth stage

In the growth stage equations (1)-(3) are solved using the updated values from the transport stage. A simplified and efficient growth model is obtained by solving the energy equation for temperature and using the semi-implicit source method of Swaminathan and Voller [10] to couple to the solid fraction evolution to the temperature field. This is the same approach used for the analytical expressions, and therefore, should limit the numerical differences attributed to the solution procedure when comparing the predictions of the microsegregation models. Details of this method can be found in the original text, the most important detail being that phase fraction is updated using a truncated Taylor Series expansion of the solid fraction. Therefore, expressions for the change in solid fraction with respect to temperature \((dg_s/dT)\) and the temperature evaluated using the previous solid fraction value \(T^\ast\) are required. Using this method, the following explicit equations are used to update the phase fraction and composition fields due to the influence of growth:

\[
g_s^{n+1} = g_s^n + \frac{dg_s}{dT} (T^{n+1} - T^\ast)
\]

(1b)

\[
C_s^{n+1} = \frac{1}{g_s^{n+1} + \varphi_s} \left( (g_s^{n+1} + \varphi_s) C_s^{n+1} + \frac{\Delta t}{\rho_s} m_s k_p C_i^\ast \right)
\]

(2b)

\[
C_i^{n+1} = \frac{1}{g_i^{n+1} + \varphi_i} \left( (g_i^{n+1} + \varphi_i) C_i^{n+1} - \frac{\Delta t}{\rho_i} m_s k_p C_i^\ast \right)
\]

(3b)

where the superscript \(n\) denotes the time step sub-iteration level. The change in solid fraction with respect to temperature \((dg_s/dT)\) is obtained applying Leibniz’s rule to equation 1 assuming a constant phase density, i.e.:

\[
\frac{dg_s}{dT} = \frac{\partial}{\partial T} \left( \frac{\partial \rho_s g_s}{\partial t} \right) \equiv \frac{\Delta t}{\rho_s} \frac{dm_s}{dT}
\]

(4)

The mass phase change rate \(m_s\) is given by a solute mass balance at the solid-liquid interface neglecting diffusion into the solid phase:

\[
m_{s,i} = \left( \frac{1 - g_s}{1 - g_{env}} \right) \frac{S_{env} \rho_i D_i}{\delta_{env} (1 - k_p) C_i^\ast} = \frac{a_i (C_i^\ast - C_i)}{(1 - k_p) C_i^\ast}
\]

(5)

where \(g_{env}\) is the volume fraction of the grain envelope, \(S_{env}\) is the interfacial area of the grain envelope, \(\delta_{env}\) is the liquid diffusion length, \(a_i = S_{env} \rho_i D_i / \delta_{env}\) is the diffusion rate coefficient characterizing the length and time-scales of solute diffusion to the extradendritic liquid, and \(f = (1 - g_s) / (1 - g_{env})\) is a modification factor to formulate the solute balance in terms of the average liquid composition in place of the extradendritic liquid composition. Equation (5) can be specified as a differentiable function with respect to temperature following the condition of interfacial equilibrium:

\[
T = T_m + m_i C_i^\ast
\]

(6)

where \(T_m\) is the melting temperature of the pure solvent and \(m_i\) is the linear liquidus slope take from the binary phase diagram. The terms required for the semi-implicit source-based method are found following the algebraic rearrangement of equations 5 and 6:

\[
\frac{dg_s}{dT} = \frac{\Delta t}{\rho_s} \left( f \frac{m_i C_i a_i}{(1 - k_p)(T - T_m)^2} \right)
\]

(7)

and
\[ T^* = T_m + m_l C_l \left( f \frac{a_l}{(k_p - 1) m_{s,l} + a_l} \right) \]  

It is noted that \( k_p C_l^* \) in equations (2b) and (3b) is replaced with \( C_{s,lr} \) for remelting (i.e. \( m_{s,l} < 0 \)) and \( C_{lr} \) during a eutectic reaction. In order to close the numerical model auxiliary relationships are required for determining \( g_{env}, S_{env}, \) and \( \delta_{env} \). These relationships are taken from Tveito et al. \[6\] and are calculated at the end of a given time step once the solid fraction and phase compositions have converged to a relative tolerance of \( 10^{-6} \). Details of the full numerical model can be found in upcoming papers from the same authors; however, it is noted that the microsegregation model was verified against the recalescence benchmark of Rappaz and Thevoz for an Al-5wt\%Si alloy \[11\].

3. Results and discussion

Before investigating the sensitivity of macrosegregation predictions to different microsegregation model inputs, the controlling transport phenomenon occurring during solidification are illustrated using the Gulliver-Scheil microsegregation model. The test case is based on SMACS benchmark \[12\] for a Sn-10wt\%Pb alloy modified for equiaxed solidification using the following expression for the relative phase velocity in the slurry:

\[ (U_s - U_l) = \frac{4}{3} \frac{(1 - g_{env}) g_{si} (\rho_s - \rho_l) d_g^2}{C_p Re \mu_l g} \]  

where \( C_p \) is the drag coefficient taken from drag model of Wang and Beckermann \[13\], \( Re \) the grain Reynolds number, \( \mu_t \) the dynamic viscosity of the liquid, and \( d_g \) the variable grain diameter. A constant envelope packing fraction is assumed \( (g_{env}^C = 0.3) \), the grains are assumed to be fully globular \( (g_{si}^C = 1) \) assuming a value of \( d_g (g_{env} = 0.3) = 75 \mu m \) at packing, and a constant relative phase density is used \( (\rho_s - \rho_l = 280 \text{ kg/m}^2) \).

Contour plots of the composition field, packing front, and streamlines are shown in figure 1. Since no superheat is provided and the microsegregation model assumes zero liquid undercooling, the slurry region quickly occupies the entire domain upon cooling from the right wall. Buoyancy forces near the chill drive a clockwise flow cell and negative segregation develops near the bottom, left corner as the depleted grains separate from the mixture flow due to settling. At the onset of packing, the grains become immobilized and the penetration of the buoyancy-driven flow transports enriched solute from the rigid mush to the slurry further depleting the rigid mush and enriching the slurry. By 200 s, the entire domain is occupied by the rigid mush and the clockwise cell of buoyancy-driven flow transports enriched interdendritic liquid from the top right to the bottom right corner. The final composition field due to this combined transport is shown in figure 1(c).

![Figure 1](image.png)

**Figure 1.** Lead composition distribution with clockwise rotating flow at (a)=50s, (b)=100s, and (c)=600s.
Figure 2. Final macrosegregation fields for the Scheil model and the kinetics models at extremes of the parameter space for $D_l$ and $N$.

3.1. Influence of microsegregation model

The total possible effect of liquid undercooling on the macrosegregation predictions during equiaxed solidification is demonstrated by varying the two primary model inputs to the microsegregation model: the liquid mass diffusivity $D_l$ and the grain number density $n_g$. These inputs are varied over the range expected for equiaxed solidification, $D_l = (10^{-9}, 5.5 \times 10^{-9}, 10^{-8}) \text{ m}^2/\text{s}$ and $n_g = (10^9, 10^9, 10^{11}, 10^{12}) \text{ m}^{-3}$, for a total of 12 simulations. The liquid undercooling increases with decreasing $D_l$ and $n_g$, and therefore, the highest deviation from the assumption made in the Gulliver-Scheil equation should occur for $D_l = 10^{-9} \text{ m}^2/\text{s}$ and $n_g = 10^9 \text{ m}^{-3}$. Contour plots of the final Pb composition fields are shown for the two extremes of microsegregation model inputs and show excellent agreement with the predictions using the Gulliver-Scheil equation (figure 2). This result indicates the liquid undercooling does not significantly affect the final macrosegregation pattern and can be explained by the fact that the bulk liquid phase becomes solutally well-mixed at the solid fractions upon which equiaxed grains typically pack (0.1-0.5) and the more complicated microsegregation model becomes functionally equivalent to the Gulliver-Scheil solution. Therefore, the only change in macrosegregation is due to the relative motion of the solid, interdendritic liquid, and extradendritic liquid in the slurry.

The change in the macrosegregation prediction using the volume-averaged microsegregation model from the previous Gulliver-Scheil results is quantified using the mean absolute percentage error

$$MAPE = \frac{100}{n} \sum_{i=1}^{n} \left| \frac{F_i - A_i}{A_i} \right|$$

where $F_i$ is the local composition at each control volume $i$ and $A_i$ is the local composition predicted using the Gulliver-Scheil model. Shown in figure 3, the difference is less than 1.5% over the entire physical parameter space indicating that solutal undercooling alone does not play a significant role in the macrosegregation formation for the current case of equiaxed solidification. For reference, it was previously shown by Schneider and Beckermann [14] that assumption of linear solidus and liquidus slopes taken from binary phase diagrams and used in most solidification models is of a similar difference.

3.2. Influence of grain hydrodynamics

To investigate the effect of grain morphology on the macrosegregation, the internal solid fraction upon packing ($g_{si}$) is varied over a physically reasonable range for dendritic grains 0.75-1.0. An octahedral grain envelope is assumed, characterized by a packing fraction of $g_{env} = 0.4$, therefore the grains become immobilized between a solid fraction of 0.3-0.4. It is assumed that both the inter- and extra-
dendritic liquid have the same velocity. A response surface of the global macrosegregation index (GMI) is shown in figure 4 for the grain number density and internal solid fraction. It is clear that the grain number density has a negligible effect on final macrosegregation prediction compared to internal solid fraction of the grain. This is further demonstrated by quantifying the GMI sensitivities for various model inputs using the elementary effects (EE) method in figure 4(b).

![Figure 3](image)

**Figure 3.** The mean absolute percent error (MAPE) measuring deviation of the macrosegregation predictions using the VA microsegregation models from the Gulliver-Scheil predictions for different values of liquid mass diffusivity and grain number density.

![Figure 4](image)

**Figure 4.** (a) The response surface of the mean absolute percent error (MAPE) measuring deviation of the macrosegregation predictions from those predicted using the Gulliver-Scheil equation for different values of liquid mass diffusivity and grain number density. (b) GMI sensitivities for the inputs.

### 4. Conclusions

First, the degree to which the macrosegregation prediction varies due to the microsegregation changes of the complicated volume-averaged models can vary from those of the Gulliver-Scheil equation are investigated. The mean absolute percent error (MAPE) over the entire physical parameter space is shown. From these results, it appears that previous studies directing the need for complicated equiaxed
dendrite models were primarily motivated by the observed change in hydrodynamic behavior compared to globular grain morphologies and not the need to capture the amount of liquid undercooling. Further effort in modeling solidification should focus more on the closure relationships used for the grain structure predictions. If the uncertainty in model inputs and/or sensitivity of the grain structure predictions to said inputs cannot be reduced using these kinetics-based models, then more pragmatic relationships should be investigated.

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