Modelling of micro- and macrosegregation for industrial multicomponent aluminium alloys

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Abstract. Realistic predictions of macrosegregation formation during casting of aluminium alloys requires an accurate modeling of solute microsegregation accounting for multicomponent phase diagrams and secondary phase formation. In the present work, the stand alone Alstruc model, a microsegregation model for industrial multicomponent aluminium alloys, is coupled with the continuum model ALSIM which calculates the macroscopic transport of mass, enthalpy, momentum, and solutes as well as stresses and deformation during solidification of aluminium. Alstruc deals with multicomponent alloys accounting for temperature dependent partition coefficients, liquidus slopes and the precipitation of secondary phases. The challenge associated with computation of microsegregation for multicomponent alloys is solved in Alstruc by approximating the phase diagram data by simple, analytical expressions which allows for a CPU-time efficient coupling with the macroscopic transport model. In the present work, the coupled model has been applied in a study of macrosegregation including thermal and solutal convection, solidification shrinkage and surface exudation on an industrial DC-cast billet.

1. Introduction

Macrosegregation, the non-homogeneous chemical composition on the length scale of the casting, is the result of several phenomena including thermal convection, solutal convection, shrinkage induced flow and surface exudation. Mathematical models quantifying the formation of macrosegregation are well-established and based on mixing theory or volume-averaging of the conservation laws for mass, solute mass, enthalpy, and momentum [1]. These models need a sub-model, e.g. a microsegregation model, relating the enthalpy (or temperature) and total solute concentrations to the liquid fraction and solute concentrations. Realistic predictions of macrosegregation formation during casting of aluminium alloys requires an accurate modeling of solute microsegregation accounting for multicomponent phase diagrams and secondary phase formation.

Multi-component microsegregation models have only to a limited extent been coupled to macrosegregation models [2-6] as the models become quite computer time and storage demanding as the total solute concentration in the volume elements change and a unique solidification path cannot be used throughout the whole casting. The solidification path needs to be updated at each time step and for each volume element of the macro model.
In the present work, the Alstruc model [7], a microsegregation model for industrial multicomponent aluminium alloys, was applied as a micro-module for macrosegregation computations. The module was coupled with the continuum model ALSIM [8-10] which calculates the macroscopic transport of mass, enthalpy, momentum, and solute as well as stresses and deformation during casting. The challenge associated with computation of microsegregation for multicomponent alloys is solved in Alstruc by approximating the phase diagram data by simple, analytical expressions which allows for a CPU-time efficient coupling with the macroscopic transport model. Preliminary assessments demonstrating the usefulness of the new module has been presented elsewhere [11,12]. The module was also used in a simplified DC-casting demonstrating the effect of secondary phase precipitation on macrosegregation formation for a 5xxx alloy [13]. In this study the coupled models are applied in a study on an industrial DC-cast billet including the effects of thermal and solutal convection, solidification shrinkage and surface exudation on the ingot concentration fields. Modelling results are compared with measurements of chemical composition through the billet from the centre to the surface.

2. Model description

2.1. The Alsim model

Alsim [8-10] calculates the development of heat, fluid flow, stresses, and deformation during casting. For aluminium DC casting situations, the model addresses the thermal and fluid flow boundary conditions to a very high level of details regarding contact zones, air gap sizes, and water hit points. This also includes the effect upon the surface heat transfer and associated surface exudation caused by air-gap formation [10, 14]. Alsim also address the formation of macrosegregation.

For a fixed solid (vs=0) the mass and momentum equations are given by:

\[
\frac{\partial}{\partial t} \rho + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = 0
\]

(1)

\[
\frac{\partial}{\partial t} (\rho \mathbf{v} \mathbf{v}) + \nabla \cdot (\rho \mathbf{v} \mathbf{v} \mathbf{v}) = -\frac{\rho}{\rho} \nabla p - \nabla \cdot (\mathbf{v} \mathbf{v})
\]

(2)

where \( \rho, \rho', g', g', v', h', v, f', g \) and \( K \) are the mixture density (\( \rho = \rho' g' + \rho' g' \)), liquid density (assumed constant except in the buoyancy term using the Boussinesq approximation), solid density, liquid volume fraction, solid volume fraction, liquid velocity, liquid pressure, kinematic viscosity, gravity vector and permeability, respectively. Solidification shrinkage, interfacial friction, and macroscopic viscous stress contribution are included. For solute conservation the following mixture equations are applied:

\[
\frac{\partial}{\partial t} (\rho c_i) + \nabla \cdot (\rho \mathbf{v} c_i \mathbf{v}) = \nabla \cdot (\rho D_i \nabla c_i)
\]

(3)

where \( c_i \) is the mixture concentration for element \( i \), and \( \rho c' = \rho g' c_i + \rho' g' c_i \). \( D_i \) is the mixture diffusion constant and \( \rho D_i = \rho g' D_i + \rho' g' D_i \). The term in the right-hand side of Eq. (3) is added for numerical stability reasons. Similar for energy conservation the following mixture enthalpy equation is applied:

\[
\frac{\partial}{\partial t} (\rho h) + \nabla \cdot (\rho \mathbf{v} h \mathbf{v}) = \nabla \cdot (\lambda \nabla T)
\]

(4)

where \( h \) is the mixture enthalpy and \( \rho h = \rho g' h' + \rho' g' h' \), and \( \lambda \) is the mixture thermal conductivity (\( \lambda = g' \lambda' + g' \lambda' \)).
The mechanical analysis is carried out in the fully solid regions of the ingot as well as in the coherent part of the mushy zone. The upper boundary of the coherent mushy zone corresponds to a solid volume fraction \( v_{\text{scoh}} \) which is an input to the model. Denoting the solid stress \( \sigma^s \), the mixture momentum balance in the coherent part of the mushy zone can be expressed by:

\[
\nabla \cdot ([g^s \sigma^s] - \nabla [(1 - g^s)p^l]) + \rho g = 0
\]

where \( g \) is the gravity vector. The total solid strain rate tensor is sub-divided into thermal, elastic, and viscoplastic parts and constitutive equations have to be provided for each contribution. Constitutive equations for the viscoplastic deformation of aluminium alloys during solidification are based on the work of Ludwig et al.\cite{15} where the mushy zone is considered as a partially cohesive porous solid saturated with liquid.

\[
\varepsilon^p = \frac{\alpha e^{-\theta R T}}{(c-a_0)^n} \left[-\frac{1}{9}A_2(g^s)J_1 I + \frac{3}{2}A_3(g^s)g^s \tau^s \right] \left[-\frac{1}{9}A_2(g^s)J_2 I^2 + 3A_3(g^s)J_2 \right]^{n-1/2}
\]

\[
\dot{\varepsilon}^p = \alpha (g^s, X) \left(1 - \frac{c}{c^*(g^s, X)} \right) \varepsilon^p
\]

\( J_1 \) and \( J_2 \) are the first and second invariants of the effective stress tensor \( \sigma^s = (g^s \sigma^g - g^l p^l I) + p^l I \), \( \tau^s \) is the solid deviatoric stress tensor, \( \varepsilon^p \) is the effective strain rate, and \( X \) is the stress triaxiality. Equation (7) describes the evolution of the partial cohesion of the coherent mushy zone. Expressions for the functions \( A_2, A_3, a \) and \( C^* \) are all given in Reference 7. Note that for the viscoplastic part of the strain, viscoplastic volumetric deformation is taken into account for temperatures between solidus and coherency, \( \varepsilon^p \neq 0 \). In the fully solid regions \( (g_s=1, A_2=0, A_3=1, C=1) \), Equation (6) simplifies to the classical flow rule with a power-law creep relation. In the fully solidified regions, strain hardening is also taken into account in the manner described in Reference 5.

### 2.2. The ALSTRUC model

ALSTRUC\cite{7} calculates the solidification path of AlMgCuFeMnSi alloys in the Al rich corner of the phase diagram. ALSTRUC also handles additions of Cr, V, Sr, Ti, and Zn in commercial alloys. The model uses the well-established assumptions of full miscibility in the liquid and thermodynamic equilibrium at the solid/liquid interface and accounts for solid-state diffusion and particle growth undercooling. A constant total alloy composition is input along with the cooling rate and the final characteristic dendrite arm length. By gradually changing the solid fraction in a step-wise manner, Alstruc computes the solute concentrations in the liquid phase, the temperature, the alloying element concentration profiles and the amount and composition of the various secondary phases in a simplified geometrical representation of the dendrite arm. Phase diagrams are formulated as simple, approximate, analytical expressions. Computer time and storage demanding calls to a thermodynamic data base, or alternatively, use of numerical mapping of such data \cite{4, 6}, is thus avoided. The amount and concentration of the secondary phases are calculated at the end of each step by considering solubility products assuming that all secondary phases are precipitated from the liquid phase. The model parameters have been tuned to metallographic investigations of solidification microstructures in samples from numerous experiments \cite{16-21}. 
2.3. A lsim/Alstruc coupling
The ALSIM/ALSTRUC coupling is carried out through a dynamically linked library (DLL). The solid phase diffusion of the alloying elements in the dendrite arms is neglected (Scheil-Gulliver approximation). This simplification also keeps the computation time and data storage low because the alloy concentration profiles do not need to be traced at the nodal points of Alsim. Input to the micro-model are the Alsim predicted values of the enthalpy and average alloy compositions at the start and end of the time step along with values for the solid fraction, alloying element concentrations in the liquid phase, and temperature at the beginning of the step. To compute the change in solid fraction, the Alstruc micro-model requires the change in liquid composition without considering solidification as input. The latter is computed from the total average concentration at the end of the time step in the following manner:

\[ c^l_{Alstruc} = (c(t) - f(t-1) \cdot c^s(t-1)) / (f(t-1) - 1) \]  

where \( c(t) \) is the average total concentration at the end of the step, \( f(t-1) \) is the fraction of solid and \( c^s(t-1) \) the composition of the solid at the beginning of the step. The approach is justified by the different time and length scales of the macroscopic and microscopic concentration changes.

The solid fraction, alloying element concentrations in the liquid phase and temperature at the end of the time step are calculated by integrating differential versions of the Scheil-Gulliver-like equations before precipitation of secondary phases.

3. Model application
The model was applied to study the macrosegregation formation in a DC cast 6xxx 270mm (11 inch) billet produced at Alcoa Lista [22] with alloy composition as shown in Table 1. A fully coupled heat and fluid flow, macrosegregation, stresses and deformation calculation was performed. The simulation included the ingot, the starting block, the mould, the gas, oil and insulation (transition) rings above the mould and the hot-top. Elements are added to the computational domain as the simulation proceeds so the domain increases in size similar to the casting itself. The contact area and the air-gap size against the mould were dynamically calculated by the model and back-coupled to the thermal boundary conditions. Fluid outflow from the semi-solid surface (exudation) is allowed if there is a local air-gap at the current element surface. The minimum air-gap size for exudation was set to 1 micrometer. If the calculated air-gap is larger than 1µm a pressure boundary condition is applied in the solution of the liquid mass and momentum equations. The pressure at the melt surface is equal to zero and the pressure at the local air-gap surface is calculated from the height of the metal column above the surface. This pressure drop is then the driving force for the fluid outflow, while the pressure drop through the semi-solid material is the limiting factor. More details about thermal and mechanical boundary conditions can be found in [22]. Thermal properties for the alloy were calculated by the microstructure model Alstruc [7]. The permeability of the semi-solid material is assumed to follow the Kozeny-Carman relation with \( K_0=1.39 \times 10^{-11} \). Diffusion constants applied in this study was \( 10^{-8} \text{m}^2/\text{s} \) and \( 10^{-12} \text{m}^2/\text{s} \) for the liquid and the solid, respectively (for all alloy components). The solid mechanical properties were taken from the 6063-alloy in [23]. The parameters for the cohesion mechanical model of the semi-solid material were taken from [7], as measured for a grain-refined Al-Cu-alloy and the upper boundary of the coherent mushy zone was set to a solid volume fraction equal to 0.6.

Table 1. Nominal composition for the alloy.

| Conc. [wt%] | Si  | Mg  | Fe  | Mn  | Cu  |
|-------------|-----|-----|-----|-----|-----|
|             | 0.593| 0.213| 0.170| 0.445| 0.170|
Figure 1: a) The temperature field and flow velocities after 280 sec. of casting. b) Silicon concentration field and flow velocities after 280 sec. of casting. The contour lines show the fraction of solid from 0.1 to 1.0 with a spacing of 0.1.

4. Results
The temperature and flow fields are shown in Figure 1a, and the silicon concentrations field is shown in Figure 1b. Figure 1 also show the air-gap formed against the bottom block and the mould. The metal flows downwards along the solidification front, partly flows into the mushy zone to feed the solidification shrinkage and flows upwards in the ingot center creating two vortices with flow in the clockwise direction. Positive segregation is obtained towards the bottom block is due to solidification shrinkage against a chill surface. Due to initial solidification during the initial holding phase, the impression of the transition ring can also be seen about 20mm up from the bottom block edge. As the bottom block starts to move the continuous casting enhanced air-gap is developing and so is the exudation. In the steady state we find a depleted center due to solidification shrinkage. A positive surface is obtained due to the inverse segregation and exudation. Exudation creates a depleted zone behind the positively segregated surface since melt enriched with Si due to microsegregation is pushed through the mushy zone into the air-gap formed in the mould region. The concentration in the exuded layer was calculated to be 3.46wt% and the thickness of the layer was 0.14 mm. The contribution of
different mechanisms to the obtained macrosegregation pattern was investigated in [24], showing that solidification shrinkage is the dominating mechanism.

**Figure 2:** a) Horizontal concentration profiles in the steady state for all alloy components. b-f) Comparison between measured concentrations (diamond symbols) [24] and model results (continuous line).
Figure 3: The liquid concentration of alloying elements as a function of solid fraction for the nominal alloy composition.

The calculated horizontal steady state concentration profiles for all alloying elements are shown in figure 2a. All elements have a similar profile as the one discussed for Silicon, with a depleted center, an enriched surface and a depleted zone behind the positively segregated surface. The largest relative segregation is calculated for the elements where the remaining liquid is largely enriched with alloying elements late in the solidification interval (from fraction of solid 0.5). Si has the largest relative segregation and Mn has a relatively flat profile. The concentration development in the liquid during solidification as calculated by the microsegregation model is shown in Figure 3 for the nominal alloy composition. The calculated profiles were compared to available measurements [24] and is shown in Figure 2 b-f. There is a good overall agreement with the measured concentration profiles. The model predicts slightly lower concentrations in the center for Si and Mg (deviation 2-3 %), and the Mn concentration in the depleted region close to the surface is slightly higher than measured concentration. However, the depletion behind the surface is captured by the model due to the coupling with an advanced micromodel. A simplified micromodel with a linearized phase diagram and a constant partition coefficient typically set to ~1 for Mn, would not capture the depletion.

5. Conclusion

In this study macrosegregation in an industrial DC-cast billet including the effects of thermal and solutal convection, solidification shrinkage and surface exudation has been calculated using coupling to an advanced microsegregation model. The proposed approach has been validated by comparing modelled concentration profiles from the centre to the surface of the billet in the steady state with measurements of chemical composition.

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