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Modelling of the thermosolutal convection and macrosegregation in the solidification of an Fe-C binary alloy

Received 1 January 2006
Accepted 9 July 2006

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Abstract

Purpose – The motivation for this work is to establish a model that not only includes the main factors resulting in macrosegregation but also retains simplicity and consistency for the sake of potential application in casting practice.

Design/methodology/approach – A mathematical model for the numerical simulation of thermosolutal convection and macrosegregation in the solidification of multicomponent alloys is developed, in which the coupled macroscopic mass, momentum, energy and species conservation equations are solved. The conservation equations are discretized by using the control volume-based finite difference method, in which an up-wind scheme is adopted to deal with the convection term. The alternative direction implicit procedure and a line-by-line solver, based on the tri-diagonal matrix algorithm, are employed to iteratively solve the algebraic equations. The velocity-pressure coupling is handled by using the SIMPLE algorithm.

Findings – Based on the present study, the liquid flow near the dendritic front is believed to play an important role in large-scale transport of the solute species. The numerical or experimental results in the literatures on the formation of channel segregation, especially those about the location of the initial flow as well as the morphology of the liquidus front, are well supported by the present investigation.

Research limitations/implications – The modelling is limited to dealing with the thermosolutal convection of two-dimensional cases. More complicated phenomena (e.g. crystal movement) and 3D geometry should be considered in future research.

Practical implications – The present model can be used to analyze the effects of process parameters on macrosegregation and, with further development, could be applied as a useful tool in casting practice.

Originality/value – The numerical simulation demonstrates the capability of the model to simulate the thermosolutal convection and macrosegregation in alloy solidification. It also shows that the present model has good application potential in the prediction and control of channel segregation.

Keywords Thermodynamics, Convection, Numerical control, Simulation, Solidification, Alloys

Paper type Research paper

International Journal of Numerical Methods for Heat & Fluid Flow
Vol. 17 No. 3, 2007
pp. 313-321

0961-5539
DOI 10.1108/09615530710730175

Nomenclature

c_p = specific heat, J/(kg °C)
C_l = species mass fraction in liquid, percentage
D_l = species diffusion coefficient in liquid, m^2/s
f_l = mass fraction of liquid
f_s = mass fraction of solid
g = gravity acceleration, m/s^2
K = permeability, m^2
K_0 = permeability coefficient, m^2
k = thermal conductivity, W/(m °C)
k_p = equilibrium partition ratio
L = latent heat, J/kg
m = liquidus slope, °C/percentage
p = pressure, Pa
T = temperature, °C
T_f = fusion temperature of pure metal, °C
t = time, s
u, v = x- and y-component of superficial velocity of liquid, m/s
$\mathbf{u}$ = superficial velocity of liquid, m/s
x, y = coordinates, m
ρ = density, kg/m^3
β_T = thermal expansion coefficient, 1/°C
β_C = solutal expansion coefficient, 1/percentage
μl = liquid viscosity, Pa s
φ = general dependent variable
Γ = diffusion coefficient
S = source term

Superscript
i = species

Subscript
ref = reference

Introduction

Macrosegregation is a type of chemical inhomogeneity in solidified castings, which is considered in some particular situations as a serious defect that deteriorates mechanical properties. Thermosolutal convection, induced and driven by uneven distribution of temperature and species in alloy solidification, is closely associated with the formation of macrosegregation. Therefore, the fluid flow, heat and mass transfer in casting solidification have significant impact on the casting quality.

Modelling of macrosegregation in castings has more than 40 years of history. At the end of the 1960s, Flemings and Nereo (1967, 1968) and Flemings *et al.* (1968) proposed the famous local solute redistribution equation (LSRE), which has been considered the beginning of modelling on macrosegregation. In the succeeding 20 years, a combination of the LSRE and Darcy's law that describes fluid flow in a porous medium was a basic approach for macrosegregation modelling. In the late 1980s, a continuum model (Bennon and Incropera, 1987a, b) and a volume-averaged model (Beckermann and Viskanta, 1988) were proposed, which have been recognized as an important milestone in the history of this field. In 1990s, a volume-averaged two-phase model (Ni and Beckermann, 1991; Schneider and Beckermann, 1995) and a multiscale/multiphase model (Wang and Beckermann, 1996; Beckermann and Wang, 1996) were developed, in which the effect of nucleation, crystal movement, and microscopic interfacial undercooling on macrosegregation were taken into account.

Channel segregation (also termed as freckles) is a typical macrosegregation occurring in unidirectional solidification of, for example, nickel-base superalloys (Giamei and Kear, 1970). The formation of this segregation is attributed principally to the thermosolutal double-diffusive convection. Copley *et al.* (1970) were the first investigators who found the presence and the role of double-diffusive convection in a bottom-chilled NH_4Cl-H_2O system. Based on direct observation, they concluded that freckles are caused by upward flowing liquid jets that result from a density inversion in the mushy zone. However, Sample and Hellawell (1984) stated that channels

originate, not within the dendritic array at any depth, but immediately ahead of the growth front as a result of perturbation from the less dense boundary layer into the bulk liquid. This conclusion was well supported by the work of Neilson and Incropera (1991) and Felicelli *et al.* (1991) who carried out numerical simulation on NH_4Cl-H_2O and Pb-Sn alloys, respectively. In recent years, mathematical models for freckle formation in multi-component alloys have been developed (Schneider *et al.*, 1997; Felicelli *et al.*, 1998).

In this paper, a mathematical model for describing the transport phenomena in multi-component alloys is presented. The motivation for this work, as a potential application in casting practice, is to establish a model that not only includes the main factors resulting in macrosegregation but also retains simplicity and consistency. By using the developed model, the solidification of a Fe-C alloy in rectangular domains is simulated, in which two cases are considered:

(1) side-cooling boundary condition case, where the distribution of flow vector, temperature and solute concentration is presented; and

(2) bottom-cooling condition case, in which the origin and development of channel segregation are discussed.

Mathematical model

The mathematical model is developed based on mainly the continuum conservation equations proposed by Bennon and Incropera (1987a), and the following assumptions are invoked:

- laminar, constant viscosity, Newtonian flow in the liquid phase;
- equal and constant phase densities except for variations in the buoyancy terms, and validity of the Boussinesq approximation;
- stationary solid without deformation and internal stress;
- isotropic permeability in the mushy zone;
- negligible flow induced by phase transformation shrinkage;
- equal and constant phase specific heat and thermal conductivity;
- local thermal and phase equilibrium in the mushy zone; and
- negligible species diffusion in the solid phase.

Based on these assumptions, the mass, momentum, energy, and species conservation can be described by the following equations:

$$\text{Continuity}: \quad \nabla \mathbf{u} = 0 \tag{1}$$

$$\text{Momentum}: \quad \frac{\partial}{\partial t}(\rho u) + \nabla(\rho \mathbf{u} u) = \nabla(\mu_l \nabla u) - \frac{\mu_l}{K} u - \frac{\partial p}{\partial x} \tag{2}$$

$$\frac{\partial}{\partial t}(\rho v) + \nabla(\rho \mathbf{u} v) = \nabla(\mu_l \nabla v) - \frac{\mu_l}{K} v - \frac{\partial p}{\partial y} + \rho g \left[\beta_T (T - T_{\text{ref}}) + \sum_i \beta_C^i \left(C_l^i - C_{l,\text{ref}}^i \right) \right] \tag{3}$$

$$\text{Energy}: \quad \frac{\partial}{\partial t}(\rho T) + \nabla(\rho \mathbf{u} T) = \nabla\left(\frac{k}{c_p} \nabla T \right) + \rho \frac{L}{c_p} \frac{\partial f_s}{\partial t} \tag{4}$$

$$\text{Species}: \quad \frac{\partial}{\partial t}\left(\rho C_l^i\right) + \nabla\left(\rho \mathbf{u} C_l^i\right) = \nabla\left(\rho f_l D_l^i \nabla C_l^i\right) + \left(1 - k_p^i\right)\frac{\partial}{\partial t}\left(\rho f_s C_l^i\right) \tag{5}$$

In the mushy zone, the energy and species conservation equations are fully coupled via the following equation:

$$T = T_f + \Sigma_i m^i C_l^i \tag{6}$$

In the conservation equations, the superficial velocity of liquid, $\mathbf{u}$, is defined as:

$$\mathbf{u} = f_l \mathbf{u}_l \tag{7}$$

where $\mathbf{u}_l$ is the intrinsic velocity of the liquid. The permeability in the mushy zone is calculated by using the Kozeny-Carman equation:

$$K = K_0 \frac{f_l^3}{(1 - f_l)^2} \tag{8}$$

where K_0 is a parameter depending on the morphology and size of the dendrites.

Solution methodology

The above conservation equations can be written in the following unified form:

$$\frac{\partial}{\partial t}(\rho\phi) + \nabla(\rho \mathbf{u}\phi) = \nabla(\Gamma \nabla\phi) + S \tag{9}$$

where φ is a general dependent variable, Γ is the diffusion coefficient, and S is a source term. The unified conservation equation is discretized by using the control volume-based finite difference method, in which an up-wind scheme is adopted to deal with the convection term. A detailed description of the discretization procedure can be found in the literature (Patankar, 1980). The solid fraction field is updated using an approach based on the coupling of temperature and liquid concentrations in the mushy zone, by equation (6). The alternate direction implicit procedure and a line-by-line solver, based on a tri-diagonal matrix algorithm, are employed to iteratively solve the algebraic equations. The velocity-pressure coupling is handled by using the SIMPLE algorithm (Patankar, 1980).

Results and discussion

The solidification of a Fe-C alloy in rectangular domains is simulated. Two cases, i.e. side- and bottom-cooling boundary conditions are studied. The domain geometry and boundary conditions are shown in Figure 1. In both cases, the initial temperature and carbon concentration of the alloy are 1,510°C and 0.8 per cent (mass fraction), respectively, and the initial state of the alloy is assumed to be quiescent. Non-slip boundary conditions are imposed on all of the domain boundaries. The main parameters used in the simulation are listed in Table I.

Case 1. Side-cooling boundary condition

In the early cooling stage, no solid phase appears and no solute redistribution happens, thus the solute distribution is uniform in the whole domain. However, the heat dissipation at the side boundary results in unevenness in temperature, and consequently an uneven distribution of liquid density in the considered domain. Figure 2 shows the calculated flow vector and temperature distribution at different times in the early cooling stage. It can be seen that at the beginning, a downward flow appears near the

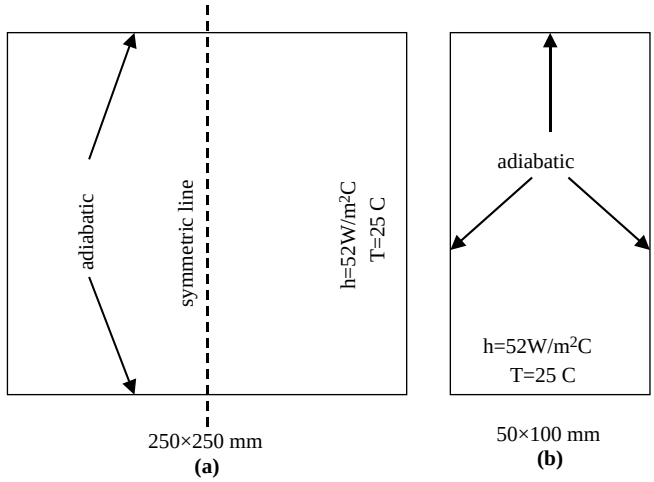

Figure 1.
The domain geometry and boundary conditions (a) for Case 1; (b) for Case 2

Parameter	Value
Density, kg/m^3	7.3×10^3
Specific heat, J/(kg °C)	7.23×10^2
Thermal conductivity, W/(m °C)	28.4
Latent heat, J/kg	2.7×10^5
Liquid viscosity, Pa s	6.0×10^{-4}
Mass diffusivity in liquid, m^2/s	2.0×10^{-9}
Partition ratio	0.3
Liquidus slope, °C/percentage	−55.0
Solutal expansion coefficient, 1/percentage	1.1×10^{-2}
Thermal expansion coefficient, 1/°C	2.0×10^{-4}
Fusion temperature of pure iron, °C	1533.0
Permeability coefficient, m^2	5.56×10^{-11}

Table I.
The parameters used in the numerical simulation

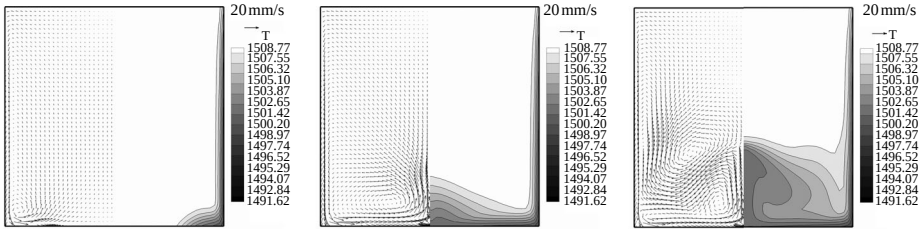

Figure 2.
The flow vector and temperature distribution in the early cooling stage

cooling boundary where the liquid density increases due to heat dissipation, and eddying flows result at the bottom corner of the domain. Then, the liquid flows along the base of the domain. When this flow encounters that coming from the opposite direction, the flow direction is changed and an upward flow is formed at the symmetric line. In the further cooling process, the fluid flow exhibits complex multi-eddy features. The variation of the temperature distribution basically reflects the development of the

fluid flow. Meanwhile, a low temperature region appears at the bottom corner of the domain.

A solid phase appears firstly at the bottom corner of the domain, and a mushy zone forms near the lower part of the side boundary. Owing to the large resistance of the solid dendrites to the fluid flow, the bulk liquid changes its flow direction and starts to move along the newly formed solidifying front (Figure 3). It has also been found that an upward flow occurs in the mushy zone due to the local accumulation of carbon, however, the flow is quite slow as compared with that in the bulk liquid. On the other hand, after commencement of the solidification, the temperature distribution in the bulk liquid tends to become even whereas in the mushy zone a large temperature gradient develops in the normal direction of the isothermals.

Figure 4 shows the variation of local averaged solute concentration (solid plus liquid) during the solidification stages. The simulation results show that in the region where the solid phase appears earlier, the local averaged solute concentration is lower than the initial concentration of the alloy. This implies that the rejected solute during solidification is carried away by the inter-dendritic liquid convection and diffusion, thus resulting in a local solute depletion. Additionally, it is seen that the rejected solute is carried into the bulk liquid mainly by the flow near the solidifying front. Therefore, the liquid flow near the dendritic front is believed to play an important role in large-scale transport of the solute species.

Case 2. Bottom-cooling boundary condition

In this case, the simulation and discussion focus mainly on the origin and development of channel segregation in the unidirectional solidification process.

Convection onset and channel origin. Figure 5 shows the calculated velocity vectors and solute concentration in the liquid phase during the early stages of solidification. The solid fraction contours during this stage are shown in Figure 6. It can be seen that

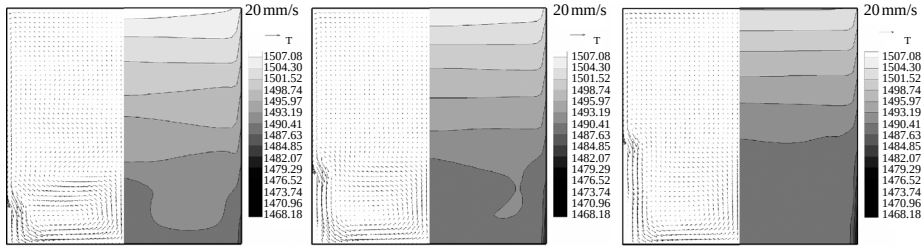

Figure 3.
The flow vector and temperature distribution during solidification process

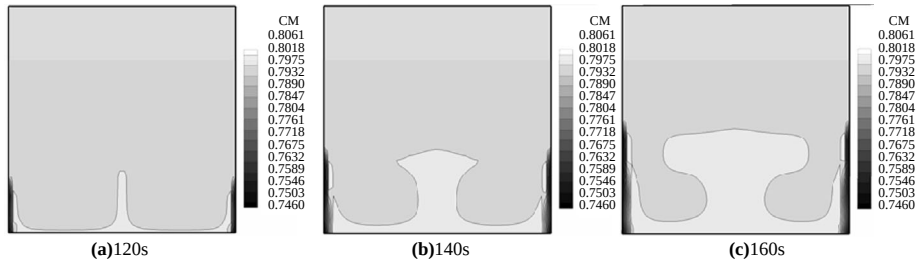

Figure 4.
The distribution of local averaged solute concentration during solidification process

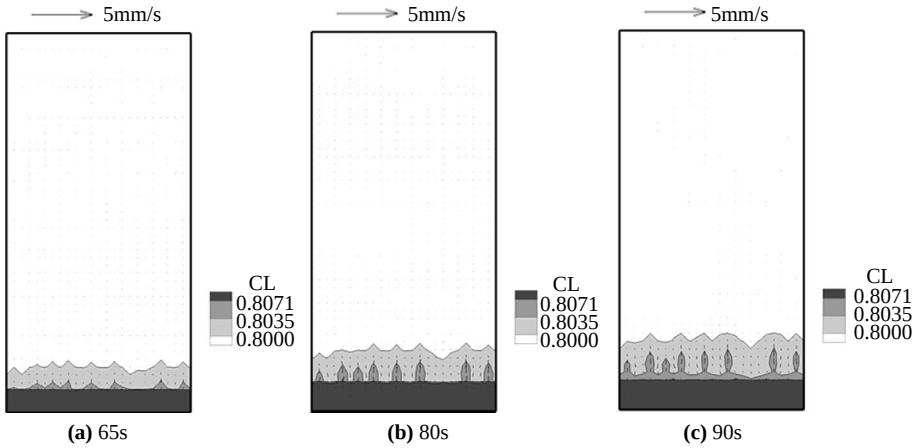

Figure 5.
The velocity vector and solute concentration in the liquid phase during the channel origin stage

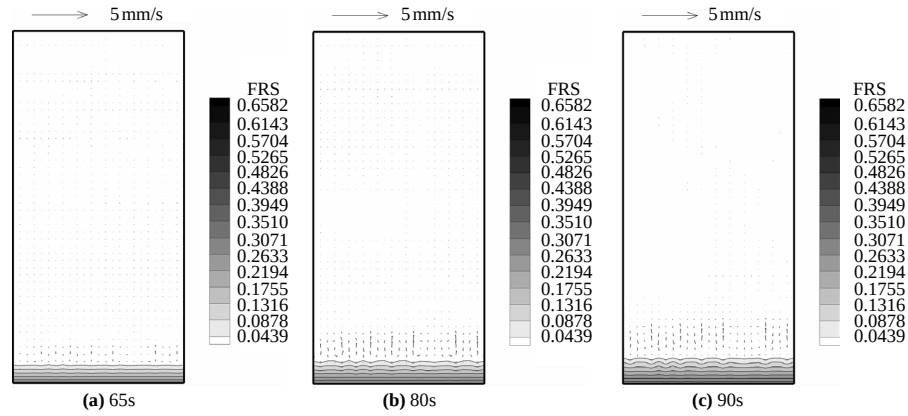

Figure 6.
The solid fraction contours during the channel origin stage

in the early stage of solidification, an uneven distribution of liquid composition develops at the dendritic front, namely, the liquid at a lower position has a higher carbon concentration, which results in an inverse distribution of the liquid density, i.e. the liquid density near the dendritic front is less than that of the bulk liquid. This is really an instable configuration in the system and once the instability increases to a certain level, an onset of upward fluid motion occurs, as shown in Figure 5(b). Further development of the initial fluid motion, after a short time, is shown in Figure 5(c). This simulation result perfectly supports the explanation given by Sample and Hellawell (1984) for the mechanism of channel origin. The initial flow occurs just ahead of the liquidus front, instead of at a deep location in the mushy zone. The simulation results (not shown here due to length limitation) also show that the temperature decreases monotonously from the top to the bottom of the domain, which could counteract in some extent the inverse distribution of liquid density. However, it may finally be seen that the distribution of liquid concentration dominates the process.

Thermosolutal convection development and channel segregation. Figure 7 shows the velocity and local averaged concentration during the full development of the channel

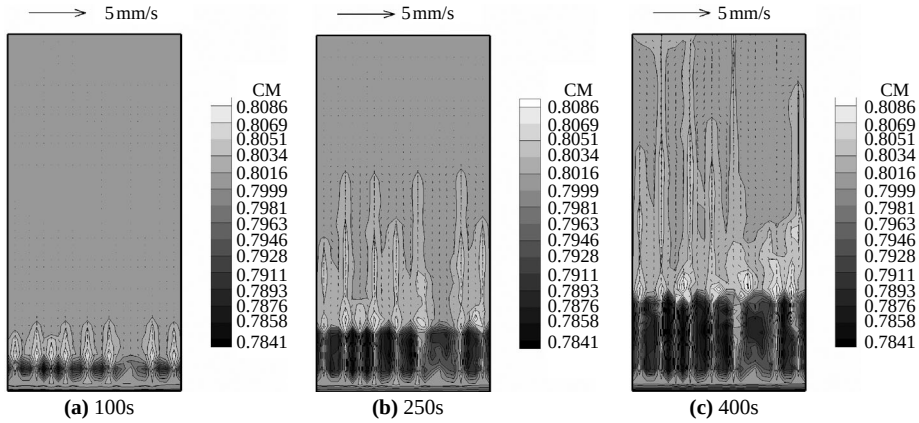

Figure 7.
The velocity vector and the local averaged concentration during the channel development stage

flows. The developed convection is characterized by fountain-like flow patterns, namely, upward liquid jets at the centre of the channels are surrounded by downward flowing liquid. This feature was observed by Sample and Hellawell (1984) in NH_4Cl-H_2O system. Furthermore, in the present study, it was found that the liquidus front bends downwards at the locations where the channel flow is fully developed. This feature was also captured by Schneider *et al.* (1997) and Felicelli *et al.* (1998) who called these concaves "volcanoes". In addition, intensive convection only occurs in the region above the liquidus front. It was reported that the liquid velocities in the mushy zone are about 2-3 orders of magnitude lower than those in the bulk liquid (Neilson and Incropera, 1991) due to the large resistance of the solid skeleton to inter-dendritic liquid flow. This is probably the main reason why the channels originate near the liquidus front.

Conclusions

A mathematical model for describing the fluid flow, heat and mass transfer in the solidification of multicomponent alloys is developed. The solidification of a Fe-C alloy in rectangular domains with side- or bottom-cooling conditions was simulated and discussed, which shows the capability of the model to simulate the thermosolutal convection and macrosegregation in alloy solidification. The modelling of the origin and development of channel segregation shows that the present model has good application potential in predicting and controlling channel segregation.

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