1. Introduction

For many engineering applications uniform properties of cast material are highly desirable. One of the major sources of non-uniformity is macrosegregation of solute, which evolves during solidification of an alloy. Unlike microsegregation, macrosegregation results in long range solutal inhomogeneity and, therefore, is difficult to eliminate by thermal processing. Some forms of macrosegregation, such as freckles, make the product unsuitable in critical application. Also, with increasing emphasis on near-net-shape casting, there shall be very few downstream processing steps of large plastic deformations and hence, little scope to alter/modify macrostructure of these semi-finished and finished products. Clearly, the need to control macrosegregation during solidification processing through better understanding of underlying mechanism is extremely important.

The cause of macrosegregation, despite several decades of intensive research, is understood mostly in qualitative terms. For example, it is known that macrosegregation is caused by long range transport of solute during the progress of solidification.2) Thermo-solutal convection, shrinkage and other convection generating forces induce fluid flow. Similarly, it is known that high superheat of the melt and large dimension of casting lead to increase in segregation of solute.2) For controlling macrosegregation, however, it is important to estimate the influence of various process and design parameters on macrosegregation with the help of some quantitative tool. Researchers in the past have studied the role of fluid flow on macrosegregation extensively using mathematical models, which have gradually become more and more sophisticated. Prescott and Incropera provide an excellent review on the subject.3) One of the important conclusions of these studies is that double-diffusive convection plays an important role in the evolution of macrosegregation.

Segregation is an important defect in steel casting and there have been few macrosegregation studies on iron–carbon and iron–carbon based steel.4–8) Amberg4) reported a numerical study on Fe–C system by using a continuum formulation based model. Singh and Basu5) have carried out simulations to study the role of double-diffusive convection on macrosegregation during solidification of binary Fe–1wt%C alloy. The effect of thermo-solutal convection on extent of segregation and segregation profile are discussed. Lesoult, Combeau and co-workers6,7) have reported the significant effects of permeability and carbon partition coefficient on axial segregation during solidification of multi-component steel. A significant conclusion of this work is that the axial segregation increases with an increase in permeability or a decrease in carbon partition coefficient. Schneider and Beckermann,8) using a fully coupled multi-component model, have shown that segregation profiles of carbon in multi-component steel show the same trend as in binary Fe–C owing to dominant role of carbon in solutal buoyancy and thermodynamic equilibria.

It is well known that heat flux has a significant effect on macrosegregation2) and steel is cast with a wide range of heat fluxes. Typical heat flux in the primary cooling zone of continuous caster is of the order of 2.5 MW/m² whereas
during some static casting, heat flux is as low as 10 kW/m². To the authors’ knowledge, however, there has been very little systematic effort to study the effect of cooling rate on macrosegregation during solidification of iron–carbon binary alloy or multi-component steel. The present study is aimed to understand the effect of cooling rate on macrosegregation during casting of steel with the help of a previously developed macrosegregation model5) based on mixture mass model of Voller et al.9) The model used in the study is briefly outlined below.

2. Numerical Model

The numerical methodology adopted for the present study is described in detail in an earlier work5) and therefore, the model is described below very briefly. The following equations are considered to represent solidification process in a rectangular cavity.

Continuity

\[ \nabla \cdot (\rho U) = 0 \] ............................................(1)

U-momentum

\[ \rho \frac{\partial u}{\partial t} + \rho u \nabla \cdot (uU) = - \frac{\partial p}{\partial y} + \nabla \cdot (\mu \nabla u) + \frac{\mu}{K} \frac{\partial u}{\partial y} \] ............................................(2)

V-momentum

\[ \rho \frac{\partial v}{\partial t} + \rho v \nabla \cdot (uU) = - \frac{\partial p}{\partial x} + \nabla \cdot (\mu \nabla v) + \frac{\mu}{K} \frac{\partial v}{\partial x} - \rho g[\beta_y(T - T_{ref}) + \beta_z(C - C_{ref})] \] ......................(3)

Energy

\[ \rho \frac{\partial (cT)}{\partial t} + \rho c \nabla \cdot (U T) = \nabla \cdot \left( \kappa \nabla T \right) + \rho \Delta H \frac{\partial f_s}{\partial t} \] ............................................(4)

Solute

\[ \frac{\partial C}{\partial t} + \rho \nabla \cdot (UC) = \nabla \cdot \left( f_s D_1 \nabla C_1 + f_s D_2 \nabla C_2 \right) \] ......................(5)

The symbols used in above equations are explained below:

- \( \rho \): density;
- \( U \): velocity vector; \( u, v \): x- and y-components of \( U \); \( t \): time; \( x, y \): axes of two orthogonal coordinate system;
- \( \mu \): viscosity; \( p \): pressure; \( g \): acceleration of gravity; \( \beta_y \): coefficient of thermal expansion; \( \beta_z \): coefficients of solutal expansion; \( T_{ref} \): reference temperature; \( C_{ref} \): reference composition; \( T \): temperature; \( C \): composition; \( c \): specific heat; \( \kappa \): conductivity; \( D \): diffusivity; \( f_s \): fraction of a phase; subscripts l: for liquid; subscripts s: for solid.

The values of viscosity, conductivity, specific heat and diffusivity in the above equations are obtained through averaging as follows:

\[ \mu = \mu_l f_l + \mu_s f_s; \quad \kappa = \kappa_l f_l + \kappa_s f_s; \quad c = c_l f_l + c_s f_s; \quad D = D_l f_l + D_s f_s \] ......................(6)

\( K \) in momentum equations is the permeability of the mushy region. In this exercise, the value of permeability is calculated based on West’s correlation10); the expression is suitably modified for simulation of iron–carbon system.11) The expressions used for permeability are presented as follows:

\[ K = T_0 \left( Y_1 K_1 + T_2 Y_2 K_2 \right) \] ............................................(7)

where, \( K_i = f_{i2}^2; \) \( K_s = 0 \) for \( f_s < 1/3 \); \( K_i = f_{i3}^2 \times (3 + 4f_i - 3 \times (8f_i - 3)^{0.5}) \) for \( f_i > 1/3 \); \( Y_1 = 6.4 \times 10^{-17} \) m²; \( Y_2 = 8.8 \times 10^{-11} \) m²; \( T_0 = 137.5 \) and \( T_0 = 0.5 f_s / f_i \).

The dependence of the local liquid fraction on the thermal and solutal fields is mainly governed by the solidification conditions. There are several equations to represent this relationship. In case of slow solidification and/or interstitial solute, the process can be considered to be close to equilibrium. In such cases, the temperature and the solute concentration in the mushy region are related to the local liquid fraction through the phase diagram. The resultant Lever rule is shown as follows.

\[ f_s(T, C) = \frac{C - k(T - T_{ref})/m}{(1 - k)(T - T_{ref})/m} \] ......................(8)

For the purpose of present study, the solidification is considered in a rectangular cavity of length \( L \) (0.1 m); height \( H \) (0.1 m); the geometry of the cavity as well as initial conditions used in the present study is shown in Fig. 1. The boundary conditions used in the present study are as follows: no-slip conditions are applied on horizontal and left vertical walls of the cavity. The left vertical wall is the line of symmetry and the gradient of \( u \)-velocity along \( X \)-axis is zero. There is no solute flux through all four bounds of the cavity. As for the thermal boundary conditions, the horizontal walls of the cavity are adiabatic and there is no heat flux at the left wall. Solidification is initiated by imposing a heat flux along the right vertical wall of the cavity. The material chosen for the present study is Fe–1wt%C. \( T_{ref} \) for the present study is taken to be 1463°C. Thus, in all the studies, a superheat of 5°C is considered.

Various thermophysical data pertaining to this system are provided in Table 1.5) The main focus of the present study is to study the effect of variation of heat flux on double-diffusive convection and, in turn, on macrosegregation. In numerical implementation, the heat flux is varied by changing the value of \( q \) along the right wall (chill face) of the cavity.

All the simulation studies are carried out on a non-uniform grid of 30×30 nodal points; the choice of nodal points is based on a grid independency test.5) A variable time step of 0.5–25 sec is employed to simulate the tran-
sient time marching solution till the end of solidification. On an average, a CPU time of 1 min is required for 5 sec of real time simulation on a SUN ULTRA-60 workstation.

3. Results and Discussion

The main focus of this study is to understand the effect of variation of heat flux on double-diffusive convection and other transport variables and, finally, on resultant macrosegregation during solidification of binary iron–carbon alloy. Simulations were carried out with heat fluxes varying in the range of 5 to 6000 kW/m². Since the time taken for complete solidification is different for various cases, overall solid fraction is chosen as a basis for comparing the results for various heat fluxes. The results presented below are at three instances of overall solidification, namely, at 20, 50 and 80%. Beyond 80% solidification, the flow field is restricted near the hot wall and is very weak in magnitude except for the higher heat fluxes. In addition to these, the macrosegregation profiles for various heat fluxes are also compared at the end of solidification.

Figures 2 through 4 show the streamlines, isotherms, mush profiles and macrosegregation profiles (at 20, 50 and 80% solidification) for heat fluxes of \( q = 10, 60 \) and 360 kW/m\(^2\), respectively. In these figures, the right vertical wall of the cavity represents the chill face, whereas, the left vertical wall, which is at the line of symmetry, represents the hot face. The maximum and minimum values of composition, temperature, stream function and fraction solid are listed in Table 2.

Table 1. Data for test problem [5].

| Property               | Value       |
|------------------------|-------------|
| Initial temperature    | 1463 °C     |
| Initial composition    | 1.0 wt% C   |
| Cavity dimension       | 0.1 m x 0.1 m |
| Density                | 6940.0 kg/m\(^3\) |
| Specific heat of liquid| 753.0 J/kgK |
| Specific heat of solid | 753.0 J/kgK |
| Conductivity of liquid | 30 W/mK     |
| Conductivity of solid  | 60 W/mK     |
| Coefficient of thermal expansion | -2.71x10\(^{-4}\)/K |
| Coefficient of solutal expansion | -0.686 |
| Liquid viscosity       | 6.94 x 10\(^{-3}\) kg/ms |
| Latent heat            | 2.72 x 10\(^{5}\) J/kg |
| Eutectic temperature   | 1136 °C     |
| Eutectic concentration | 4.3 wt% C   |
| Partition coefficients | 0.42        |
| Melting point of pure iron | 1536 °C   |
| Diffusivity of carbon in solid | 4.8x10\(^{-12}\) m\(^2)/s |
| Diffusivity of carbon in liquid | 1.0x10\(^{-9}\) m\(^2)/s |

Figure 2 shows the results for heat flux of 10 kW/m\(^2\). At 20% overall solidification, Fig. 2(a), the flow pattern is very complex and the presence of multiple vortices is readily seen. The isotherms, shown in Fig. 2(b), are affected by the flow to a large extent; they are parallel to the horizontal axis showing the dominance of convection over conduction. Although the overall solidification is only 20%, the mush is spread over the large part of the cavity and both mush profiles and macrosegregation patterns, shown in Figs. 2(c) and 2(d), respectively, are very complex. Since the mush covers the large part of the cavity, the resistance to the flow is high leading to the reduction in strength of thermal buoyant flow. Isotherms are almost parallel to horizontal wall near the hot wall of the cavity; the fluid near the bottom corner is thermally stable and a solutal buoyancy driven cell is readily seen.

With the progress of solidification, the strength of flow diminishes and isotherms show its vertical nature near the cold wall. The macrosegregation profile is highly evolved by this time. Important feature of mush profile at this junction is that even though the overall solidification is around 50%, pure solid regime is yet to start. As the solidification progresses further, the flow strength becomes negligible. At 80% the flow is almost absent and isotherms are mostly vertical in nature. There is very little difference between macrosegregation profiles of this time and those at 50% solidification.

Figure 3 shows the results for heat flux of 60 kW/m\(^2\). At 20% solidification, the strength of flow is higher than that of the case of 10 kW/m\(^2\). However, the isotherms show conduction dominance near the cold wall due to large \( f_\text{s} \). The macrosegregation patterns are less complex in this case compared to those for \( q = 10 \) kW/m\(^2\). At 50% solidification, the flow strength is considerably diminished and mush profiles and macrosegregation profiles are now well evolved. As solidification progresses further, the flow is restricted in a narrow zone near the hot wall. At 80% solidification, the isotherms are vertical in most part of the cavity. Comparing the macrosegregation patterns at 50 and 80% solidification, it is noted that though the evolution of the macrosegregation patterns are largely complete at 50% for \( q = 60 \) kW/m\(^2\), the differences in patterns at 50 and 80% are more than those for \( q = 10 \) kW/m\(^2\).

Figure 4 shows the results for heat flux of 360 kW/m\(^2\). At 20% solidification, the strength of flow is higher than the other two cases. In addition to the major vortex (in pure liquid region), a minor vortex is clearly visible in this case (in the mushy region). The mushy region is very narrow in this case and isotherms in mushy region are largely conduction dominated due to high resistance to flow. There is virtually no temperature gradient in the pure liquid region, Fig. 4(a), due to high strength of flow in the pure liquid zone. Macrosegregation pattern is much simpler in this case and the extent of segregation is less than the other two cases.

As solidification progresses, the strength of major and minor vortices goes down. Mushy region continues to be narrow and all three regions, namely, liquid, solid and mushy regions are clearly seen. As solidification progresses further, the flow is restricted in a narrow zone near the hot wall. At 80% solidification, the isotherms are vertical in
Fig. 2. Streamlines, isotherms, mush profile and macrosegregation pattern for 20, 50 and 80% solidification for heat flux of 10 kW/m²: (a) $\psi_{\text{max}}$ = 1.10, $\psi_{\text{min}}$ = 1.97 & $\Delta \psi$ = 0.25, (b) $T_{\text{max}}$ = 1450°C, $T_{\text{min}}$ = 1438°C & $\Delta T$ = 5°C, (c) $f_{\text{max}}$ = 0.66, $f_{\text{min}}$ = 0.0 & $\Delta f$ = 0.1, (d) $C_{\text{max}}$ = 1.19 wt%, $C_{\text{min}}$ = 0.76 wt% & $\Delta C$ = 0.1 wt%, (e) $\psi_{\text{max}}$ = 0.44, $\psi_{\text{min}}$ = 0.01 & $\Delta \psi$ = 0.05, (f) $T_{\text{max}}$ = 1431°C, $T_{\text{min}}$ = 1417°C & $\Delta T$ = 5°C, (g) $f_{\text{max}}$ = 0.86, $f_{\text{min}}$ = 0.0 & $\Delta f$ = 0.10, (h) $C_{\text{max}}$ = 1.37 wt%, $C_{\text{min}}$ = 0.76 wt% & $\Delta C$ = 0.1 wt%, (i) $\psi_{\text{max}}$ = 0.01, $\psi_{\text{min}}$ = 0.0 & $\Delta \psi$ = 1.5 x $10^{-3}$, (j) $T_{\text{max}}$ = 1393°C, $T_{\text{min}}$ = 1382°C & $\Delta T$ = 5°C, (k) $f_{\text{max}}$ = 1.00, $f_{\text{min}}$ = 0.29 & $\Delta f$ = 0.1, (l) $C_{\text{max}}$ = 1.45 wt%, $C_{\text{min}}$ = 0.76 wt% & $\Delta C$ = 0.1 wt%.

Fig. 3. Streamlines, isotherms, mush profile and macrosegregation pattern for 20, 50 and 80% solidification for heat flux of 60 kW/m²: (a) $\psi_{\text{max}}$ = 7.0, $\psi_{\text{min}}$ = 0.17 & $\Delta \psi$ = 1, (b) $T_{\text{max}}$ = 1455°C, $T_{\text{min}}$ = 1410°C & $\Delta T$ = 5°C, (c) $f_{\text{max}}$ = 0.80, $f_{\text{min}}$ = 0.0 & $\Delta f$ = 0.1, (d) $C_{\text{max}}$ = 1.22 wt%, $C_{\text{min}}$ = 0.71 wt% & $\Delta C$ = 0.1 wt%, (e) $\psi_{\text{max}}$ = 1.65, $\psi_{\text{min}}$ = 0.10 & $\Delta \psi$ = 0.55, (f) $T_{\text{max}}$ = 1448°C, $T_{\text{min}}$ = 1378°C & $\Delta T$ = 5°C, (g) $f_{\text{max}}$ = 1.00, $f_{\text{min}}$ = 0.0 & $\Delta f$ = 0.10, (h) $C_{\text{max}}$ = 1.31 wt%, $C_{\text{min}}$ = 0.71 wt% & $\Delta C$ = 0.1 wt%, (i) $\psi_{\text{max}}$ = 1.31, $\psi_{\text{min}}$ = 0.01 & $\Delta \psi$ = 0.02, (j) $T_{\text{max}}$ = 1408°C, $T_{\text{min}}$ = 1336°C & $\Delta T$ = 5°C, (k) $f_{\text{max}}$ = 1.0, $f_{\text{min}}$ = 0.11 & $\Delta f$ = 0.1, (l) $C_{\text{max}}$ = 1.66 wt%, $C_{\text{min}}$ = 0.68 wt% & $\Delta C$ = 0.1 wt%. 

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most part of the cavity. Comparing the macrosegregation patterns at 50 and 80% solidification, it is noted that though the evolution of the macrosegregation patterns are largely complete at 50% for $q=360\,\text{kW/m}^2$ the differences in patterns at 50 and 80% are more than the other two cases.

Macrosegregation patterns at the end of complete solidification are shown in Figs. 5(a)–5(c). It is readily noted that overall nature of macrosegregation patterns undergoes drastic changes with heat flux. The severity of segregation ($C_{\text{max}}-C_{\text{min}}$) at $q=60\,\text{kW/m}^2$ is higher than those at 10\,\text{kW/m}^2 and 360\,\text{kW/m}^2.

For the quantitative comparison of macrosegregation, a parameter called global extent of segregation (GES) is used which is defined as root mean square of deviation from nominal composition of all the nodal points.

$$\text{GES} = \frac{100}{C_{\text{avg}}} \left[ \frac{1}{\sum \sum (\Delta C)^2 \, dx \, dy} \right]^{1/2}$$ (9)

The global extent of segregation, GES, as a function of heat flux is shown in Fig. 6. It is seen from the graph that there is a drop in GES with an increase in heat flux in the beginning. However, GES starts to rise at around $q=10\,\text{kW/m}^2$ and goes through a peak at $q=60\,\text{kW/m}^2$. Beyond this point, there is a steady fall in GES with an increase in heat flux. Thus it is seen that there are three regimes in GES curve. The first regime corresponds to $q<10\,\text{kW/m}^2$ where GES falls monotonically with an increase in heat flux. This observation is in line with the observations of Tewari et al. who studied vertical solidification of Pb–Sn alloys. The main observation was that the decrease in the rate of

| Heat Flux (K/W/m²) | % Solidification | Composition (wt%) | Temperature (°C) | Streamlines | Fraction solid |
|-------------------|------------------|-------------------|------------------|-------------|----------------|
| 10                | 20               | 1.19 0.76 | 1450 1438 | 1.10 1.97 | 0.66 0.0 |
| 50                | 1.37 0.76 | 1431 1417 | 0.44 0.01 | 0.86 0.0 |
| 80                | 1.45 0.76 | 1393 1382 | 0.01 0.0 | 1.0 0.29 |
| 100               | 1.45 0.76 | 1267 1258 | - - | 1.0 1.0 |
| 60                | 20               | 1.22 0.71 | 1445 1419 | 1.70 0.17 | 0.80 0.0 |
| 50                | 1.31 0.71 | 1448 1378 | 1.65 0.10 | 1.0 0.0 |
| 80                | 1.66 0.68 | 1408 1338 | 0.20 0.01 | 1.0 0.11 |
| 100               | 1.66 0.68 | 1232 1182 | - - | 1.0 1.0 |
| 360               | 20               | 1.19 0.80 | 1463 1413 | 10.66 0.80 | 1.0 0.0 |
| 50                | 1.23 0.79 | 1462 1416 | 12.33 0.88 | 1.0 0.0 |
| 80                | 1.42 0.80 | 1460 1395 | 1.57 0.24 | 1.0 0.0 |
| 100               | 1.46 0.80 | 1276 0926 | - - | 1.0 1.0 |
solidification led to the increases in strength of flow and macrosegregation. Although the present study is on horizontal solidification, at the lower heat fluxes solutal buoyancy plays a significant role in the mushy region. The drop in GES can be attributed to the net drop in strength of solutal buoyancy.

Between $q = 10 \text{ kW/m}^2$ and 100 $\text{kW/m}^2$, GES first rises and then starts to decrease. This peculiar behavior is due to the opposing nature of thermo-solutal convection, which becomes important at the lower heat fluxes. In this zone, thermal buoyancy completely overcomes solutal buoyancy and causes rise in macrosegregation. However, this rise is arrested beyond $q = 60 \text{ kW/m}^2$ as the higher heat fluxes also causes higher rate of solidification, which reduces the time for the evolution of macrosegregation. Prescott and Incropera\textsuperscript{13}) in their numerical study on Pb–Sn alloy, have observed similar phenomena. The main reasons for the fall in macrosegregation level at very high heat fluxes are due to lowering of solidification time and narrow mushy region, which allows very little solute transport to the pure liquid region. Thus it is clearly seen that due to opposing nature of thermo-solutal convection, GES curve shows a hump between two monotonically decreasing portions of GES curve. The above results clearly showed a complex variation of GES with heat flux.

4. Conclusions

The aim of the present work was to study the effect of cooling rate on the double-diffusive convection and its role in evolution of macrosegregation during solidification of Fe–1wt%C alloy. A comprehensive model was used to simulate the effect of heat flux on thermo-solutal convection and, in turn, on the macrosegregation. The heat flux was varied in the range of 5 and 6000 $\text{kW/m}^2$. Some of the important findings of the present study are highlighted below.

- GES goes down monotonically up to $q = 10 \text{ kW/m}^2$. Solutal buoyancy plays a crucial role in this regime and the lower the rate of solidification is, the higher the GES is.
- Between heat flux of 10 and 100 $\text{kW/m}^2$, the GES goes through a maximum. This is due to opposing nature of thermo-solutal convection. In this regime, thermal buoyancy plays an important role in evolution of macrosegregation.
- For higher heat flux (>100 $\text{kW/m}^2$), the GES curve goes down monotonically with an increase in heat flux. The main reasons for a decrease in GES are lowering of solidification time and narrow mushy zone which do not allow solute transport from the mushy region.

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Nomenclature

- $C$: Composition
- $D$: Diffusivity ($\text{m}^2/\text{s}$)
- $H$: Width of the cavity (m)
- $\Delta H$: Latent heat of fusion (J/kg)
- $K$: Permeability of the mush ($\text{m}^2$)
- $L$: Length of the cavity (m)
- $S$: Source term
- $U$: Velocity vector ($\text{m/s}$)
- $c$: Specific heat ($\text{J/kg K}$)
- $dxdy$: Area element ($\text{m}^2$)
- $f$: Fraction
