Positive segregation as a function of buoyancy force during steel ingot solidification

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Abstract
We analyze theoretically and experimentally solute redistribution in the dendritic solidification process and positive segregation during solidification of steel ingots. Positive segregation is mainly caused by liquid flow in the mushy zone. Changes in the liquid steel velocity are caused by the temperature gradient and by the increase in the solid fraction during solidification. The effects of buoyancy and of the change in the solid fraction on segregation intensity are analyzed. The relationships between the density change, liquid fraction and the steel composition are considered. Such elements as W, Ni, Mo and Cr decrease the effect of the density variations, i.e. they show smaller tendency to segregate. Based on the modeling and experimental results, coefficients are provided controlling the effects of chemical composition, secondary dendrite arm spacing and the solid fraction.

Keywords: segregation, steel ingot, solidification, buoyancy effect

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Solute redistribution during solidification of steel ingots determines the growth morphology, phase distribution and the amount of macro-segregation. Macro-segregation in steel ingots is caused by the different solubility of alloying elements in the liquid and solid metal, i.e. by the generation of concentration gradients in both zones, by the impossibility of these two gradients to equalize due to diffusion during solidification, and by the transport of liquid regions enriched by a solute.

One of the reasons for the transport of the segregated liquids may be density differences in the metal due to changes in temperature and composition. Hot liquid metal cools down near the cold surfaces, and the concomitant density increase causes it to sink. In the upper part of an ingot, regions with a higher concentration of alloying elements are formed, i.e. positive segregation occurs. On the other hand, negative segregation takes place in the lower part of the ingot that can be explained by the sedimentation of equiaxed crystals formed in the bulk liquid. Several segregation models, as well as experimental results on segregation intensity, were presented [1–3], and macro-segregation modeling was reviewed [4, 5].

Many variables affect the macroscopic distribution of a solute in a cast ingot, the most important of which are the size of the ingot, solidification rate, heat extraction, chemical composition and superheating. Furthermore, the solid framework of the mushy zone and the presence of equiaxed crystals are very important. The average intensity of the segregation in the central part of the ingot should be small if equiaxed crystals can not be transported [6]. High flux density in the two-phase region results in the early immobilization of equiaxed crystals and reduced segregation in the central part.

Based on theoretical analysis of the positive segregation and on the numerical simulation of the solidification of steel ingot [7], the effect of the temperature gradient on the velocity of interdendritic liquid flow in the two-phase region was analyzed.
The velocity component $V_y$ is

$$V_y = \frac{K \cdot g}{\eta \cdot f_l^1 (\rho_l^0 - \rho_l)}$$

where $K$ and $\eta$ are the permeability and viscosity of the liquid, $g$ is acceleration due to gravity, $\rho_l^0$ and $\rho_l$ are the liquid densities in the liquid and mushy zones, respectively, and $f_l$ is the liquid fraction.

Changes in $\eta$ are very small, and thus $V_y$ depends mostly on the changes in $K$ and $\eta$. Permeability $K$ can be determined empirically. In the direction perpendicular to the primary dendrite arms, it can be expressed [14] as

$$K = 1.73 \times 10^{-3} \frac{\lambda_1^{1.09}}{\lambda_2} \cdot \frac{\lambda_3^2 \cdot f_l^3}{(1 - f_l)^{0.749}}$$

Here $\lambda_1$ and $\lambda_2$ are the primary and secondary dendrite arm spacings, respectively.

The density change $d\rho_l/dT_l$ is difficult to determine, therefore, the average value $d\rho_l/df_l$ in the mushy zone is usually examined instead. Permeability $K$ can be determined empirically. In the direction perpendicular to the primary dendrite arms, it can be expressed [14] as

$$K = 1.73 \times 10^{-3} \frac{\lambda_1^{1.09}}{\lambda_2} \cdot \frac{\lambda_3^2 \cdot f_l^3}{(1 - f_l)^{0.749}}$$

If $d\rho_l/df_l < 0$ then the liquid in the mushy zone flows upward, and positive segregation occurs at the top of the ingot. The interdendritic liquid density depends on its temperature and chemical composition and it is a function of $f_l$

$$\frac{d\rho_l}{df_l} = \frac{\partial \rho_l}{\partial T_l} \cdot \frac{\partial T_l}{\partial f_l} + \frac{\partial \rho_l}{\partial c_l} \cdot \frac{\partial c_l}{\partial f_l}$$

Using the values of the partition coefficient for carbon and other elements, the following expression for $V_y$ can obtained

$$V_y = A \cdot Z_{\lambda_\lambda} \cdot Z_B \cdot Z_c$$

Here $A$ is a constant, and parameters $Z_{\lambda_\lambda}$, $Z_B$ and $Z_c$ control the dendrite arm spacing, liquid fraction and chemical composition, respectively.

Substituting the values of table 1 into equation (4) yields [15] the parameters in equation (5), which are listed in table 2.

Equation (4) reveals that temperature variation in the mold during solidification is important in calculating $d\rho_l/df_l$. Using the above relations, the density change and flow velocity can only be determined if temperature is continuously monitored.

2.1. Effect of dispersion of dendrite structure

The coefficients $Z_{\lambda_\lambda}$, $Z_B$ and $Z_c$ in equation (5) depend on various factors, but all of them are related to the temperature gradient. If the primary dendrites are defined as axially
symmetric ellipsoids then their arm space can be expressed as [16]

$$\lambda_1 = 4.3(\Delta T_m)^{1/2}(D\Gamma/\Delta T_m \cdot k)^{1/4}R^{-1/4}G^{-1/2}. \quad (6)$$

Here $\Delta T_m$ is the temperature difference between the liquidus and solidus lines, $D$ is the carbon diffusion coefficient, $\Gamma$ is the Gibbs–Thomson parameter, $k$ is the partition coefficient of carbon, $R$ is the rate of solidification and $G$ is the temperature gradient.

Ostwald mechanism is mostly used in the theoretical analysis of the formation of the secondary dendrite. Based on this mechanism, Imagembali [17] gives a general relation for the secondary arm spacing as

$$\lambda_2 = (2\pi)^{1-n}(D\Gamma/R \Delta T_m k)^{(1-n)/2}(8 R \Theta)^n. \quad (7)$$

Here $\Theta$ is the local solidification time, which can be calculated as

$$\Theta = \Delta T_m/R\tilde{G}. \quad (8)$$

The above equations reveal that the dominant factors affecting $Z_s$ in equation (5) are the change in temperature during the ingot solidification and the properties of the initial material. The solidification time depends on the dimensions and shape of the ingot, as well as its chemical composition and the casting temperature. Despite numerous studies, it is not clear whether the parameter $n$ in equation (7) is a constant or it changes across the surface of the solid–liquid boundary. Steels are multicomponent alloys where variations of $n$ are difficult to determine. Therefore, $n$ is usually taken as 1/3. Upon substituting this value in equation (7), the following expression is obtained:

$$\lambda_2 = 6.8(D\Gamma/R\tilde{G}k)^{1/3}. \quad (9)$$

### 2.2. Effect of solid fractions during solidification

The effect of the solidification dynamics on the liquid flow rate in the two-phase region (solid–liquid) can be obtained from $Z_s$ in equation (5):

$$Z_s = f_0^3(1 - f_1)^{-0.749} \quad (10)$$

or

$$Z_{gg} = (1 - f_g)^3 f_s^{-0.749}. \quad (11)$$

The critical value of $f_g$ for liquid metal is $\sim 0.2$, and thus, the changes in liquid density can be calculated in the range $f_g < f_1 < 1$. Solid fraction $f_s$ increases during solidification. Therefore, it is necessary to analyze the effect of temperature changes on $f_s$. For the more detailed analysis of the temperature field of an ingot cross section, a numerical model of the solidification process was developed [7] based on Fourier’s differential equation:

$$\rho c_p \frac{\partial T}{\partial t} = \lambda \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) + q. \quad (12)$$

Here $\rho$ is the steel density, $c_p$ is the heat capacity, $\lambda$ is the thermal conductivity and $q$ is latent heat. The change in the solid fraction is given by

$$\frac{df_s}{dT} = \frac{1}{\Delta T_m(1-2\pi)} \left[ 1 - \frac{\pi(T - T_i)}{2(T_s - T_i)} \right]. \quad (13)$$

The heat flux conducted across the ingot surface can be defined as the sum of convection and radiation heat until the formation of an ‘air gap’ (separation between the ingot surface and mold wall):

$$\frac{\partial T}{\partial x} = \frac{1.24(T_i - T_m)^{1.33} + \sigma \varepsilon (T_s^4 - T_m^4)}{\lambda}. \quad (14)$$

After formation of the air gap, the heat is only transferred by radiation and can be expressed as

$$\frac{\partial T}{\partial x} = \frac{\sigma (T_s^4 - T_m^4)}{\lambda(1/\varepsilon_m + 1/\varepsilon_i - 1)}. \quad (15)$$

Here $T_i$ is the surface temperature of the ingot, $T_m$ is the mold temperature, $\sigma$ is the Stefan–Boltzmann constant, $\varepsilon_i$ and $\varepsilon_m$ are the emissivities of the ingot and mold, respectively, and $T_i$ is the liquid metal temperature.

### 3. Experimental procedure and results

The proposed model is applied to the solidification of an ingot of high-alloy tool steel. The chemical composition of the studied steel is given in Table 3.

Temperature during ingot solidification was measured with Pt-Rh18-Pt thermocouples fixed at different positions at two-thirds of the ingot height. Ingot has square cross-section.
Table 3. Chemical composition of the studied steel.

| Element | C   | Si  | Mn  | Cr  | Ni  | V   | W   | Mo  | P   | S   | Fe   |
|---------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|------|
| wt%     | 1.65| 0.35| 0.32| 12.5| 0.31| 0.22| 0.45| 0.64| 0.03| 0.03| 83.7 |

Figure 2. Temperature distribution across the ingot cross section as a function of time (Casting temperature 1693 K).

Figure 3. Temperature distribution across the ingot cross section as a function of distance from the ingot surface (Casting temperature 1770 K).

There, we can assume \( x = y \) in equation (12) and analyze only one quadrant of the cross-section. The temperature distributions during the ingot solidification, calculated as a function of time and the distance from the surface, are presented in figures 2–4.

The temperature of the liquid steel during solidification was measured by three thermocouples at different positions shown in figure 5. The agreement between the measured and calculated values is good, particularly at the ingot surface.

The values of temperature gradient and solidification rate obtained from the model can be used to calculate the spacing of the primary and the secondary dendrite arms using equations (6) and (9), respectively. The theoretical values of \( \lambda_1 \) and \( \lambda_2 \) were compared with the experimental values for different casting temperatures (figures 6 and 7).

Interdendritic arm spacing increases from the surface to the center of the ingot. The differences between calculated and experimental values are caused by a number of solidification parameters; thus, it is difficult to derive analytical relations for \( \lambda_1 \) and \( \lambda_2 \). The obtained values of \( \lambda_1 \) and \( \lambda_2 \) can be used to calculate the controlling parameter \( Z_\lambda \) in equation (5), as shown in figure 8.

In figure 8, \( Z_\lambda \) is plotted versus the temperature gradient. Decrease in the temperature gradient increases the effect of \( Z_\lambda \). This effect is more pronounced at the beginning of the solidification process while at the end it tends to be reduced (figure 9). Explanation of those changes can be found in the fact that the end of the solidification process is followed by the formation of zone equiaxed crystals in the center of the ingot without a pronounced dendrite structure.

The liquid velocity in the mushy zone depends on the fraction \( f_l \), i.e. on the increment of the solid phase \( f_s \). With
increasing solid phase fraction the space available for the liquid metal to flow decreases due to the buoyancy effect. Using the numerical model we obtained \( \frac{df_{l}}{dt} \) during ingot solidification, as shown in figure 10.

Based on the data of table 2, we can determine changes in the correction factor \( Z_{\beta} \). Its dependence on the distance from the surface is shown in figure 11.

It is clear that near the ingot surface \( Z_{\beta} \) has little effect on \( V_{y} \). The reason for that is the dendrite structure does not form near the surface but undergoes almost instantaneous bulk solidification.

In the central region, there also is no change in \( Z_{\beta} \). The latter has small value because solidification is completed and liquid fraction is small \( (f_{l} = 0.1) \) in this region. Considering the effect of the dendrite structure dispersion and the liquid fraction, i.e. the amount of solid phase in the solidification equation (5), it is possible to derive the overall dependence of liquid flow velocity on these parameters. For steels with a fixed chemical composition \( Z_{\gamma} \), equation (5) is constant; it does not affect the variation of \( V_{y} \), although it has some effect on its absolute value.

The dependence \( V_{y} = f(Z_{\beta}, Z_{\gamma}) \) is shown in figure 12. The velocity of liquid flow \( V_{y} \) decreases with decreasing \( Z_{\beta} \), but increases with increasing \( Z_{\gamma} \). During solidification, these two parameters behave differently: \( f_{l} \) decreases, but \( \lambda_{1} \) and \( \lambda_{2} \) increase. In the model of solidification of high alloy tool steel, the dominant effect on \( V_{y} \) is the increase in interdendrite arm spacing (figure 13).

In zone II, the increase in velocity \( V_{y} \) is approximately linear versus distance from the ingot surface, whereas \( V_{y} \) in zones I and III exhibits a different trend. In the area with a developed dendrite structure \( V_{y} \) increases almost linearly, while \( V_{y} \) decreases in the central zone. The \( V_{y} \) value is higher for a higher casting temperature due to the increased formation of the dendrite structure and the effect of parameter \( Z_{\gamma} \).

4. Effect of \( V_{y} \) on the formation of positive segregation profile

An equation for global segregation intensity can be derived by integrating the local segregation intensity [5]:

\[
S_{g}^{\phi} = \frac{\iiint_{V} \left( C - C_{o} \right)^{2} \, dV}{V}.
\]

Here \( C_{i} \) is the concentration of a certain element at a certain point and \( C_{o} \) is the average concentration of the element.

Using the experimentally obtained values of local segregation intensity across the ingot cross section, we derived the dependence of the macro-segregation intensity on the liquid metal velocity (figure 14). The dependence of \( S_{g}^{\phi} \) on \( V_{y} \) is approximately linear, and it is most pronounced for elements having small partition coefficient (sulfur, phosphorus) and thus prone to segregation. These elements have the greatest effect on the value of the correction factor \( Z_{\gamma} \) (table 1); thus, they indirectly increase the flow rate of the liquid and \( S_{g}^{\phi} \). In this way, the experimental values of \( M \)-shaped segregation profiles can be explained, particularly in the upper part of the ingot.

Comparing the liquid flow rate and profile segregation contour with the images of the microstructure across the cross section of the ingot reveals similar changes (figure 15). The
Figure 9. Micrographs of dendrite microstructure of investigated steel for different casting temperatures (×50).

Figure 10. Changes in solid fraction during ingot solidification at different distances from surface (x).

Figure 11. Change in $Z_n$ during solidification for different distances from surface (x).

Figure 12. Changes in $V_y$ as function of $Z_n$ and $Z_{\lambda}$.

Figure 13. Variation of $V_y$ for different values of casting temperature (I: zone of equiaxed crystals, II: columnar zone, III: central zone).

Similarity is particularly pronounced in the columnar dendrite zone. However, there are differences at the surface and in the central zone containing equiaxed crystals. The liquid flow rate and the overall segregation intensity increase linearly in the zone of dendrite crystals. In the surface zone, these differences can be neglected, while in the central zone the difference is due to the very small amount of liquid flow because of the high value of the solid fraction $f_s$.

The maximum values of $V_y$ and $S_{gi}$ coincide not at the ingot center but at the border between the columnar dendrite zone and the central zone. This fact can verify the conclusion.
that the overall segregation intensity depends greatly on the liquid velocity in the interdendritic region, but only in the range of $f_s = 0.15$–0.70.

5. Conclusions

Positive macro-segregation in the upper part of a steel ingot during solidification is directly affected by the liquid flow in the mushy zone due to the buoyancy effect and the density decrease. The dispersion of dendrite structures and the chemical composition also contribute to the rate of liquid transfer.

The effects of these three factors can be expressed by the parameters $Z_\lambda$, $Z_c$ and $Z_{fl}$. Based on the developed here numerical model of solidification, the areas of predominance of each factor have been determined.

In the zone of equiaxed crystals, the prevailing factor is $Z_c$, i.e. the chemical composition, while in the zone of columnar dendrites, the $Z_B$ and $Z_s$ dominate. Global macro-segregation intensity ($S_g^i$) depends almost linearly on the liquid velocity. The greatest value of $S_g^i$ is between the columnar dendrite zone and the central zone, in agreement with the experimental data.

Using the measured temperature changes (i.e. the temperature gradient) and the derived mathematical equations, the relationships were obtained between the buoyancy effect, the dispersion of dendrite structures and the presence of segregated elements. The segregation intensity increases with increasing $V_y$, particularly in for elements with small partition coefficients such as S and P.

Using the steel solidification model and the parameters $Z_\lambda$, $Z_c$ and $Z_{fl}$, it is possible to determine the liquid velocity and its relation with the segregation intensity.

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