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Influence of crystal fragmentation on the formation of microstructure and macrosegregation during directional solidification under forced convection condition

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Abstract. Directional solidification experiment under forced convection condition was conducted. The AlSi7 alloy was solidified in an alumina cylindrical crucible (Ø8 mm) in a Bridgeman furnace; and forced convection was induced by applying rotating magnetic field (RMF). The RMF induced flow in the sample during solidification leads to the formation of equiaxed crystals by the mechanism of crystal fragmentation (assumption). The current study is to use a mixed columnarequiaxed solidification model to simulate this experiment by considering the crystal fragmentation as sole origin of equiaxed crystals. An inward flow (Ekman effect) forms in the front of the (columnar) mushy zone under the RMF. Solute-driven remelting, as enhanced by the interdendritic flow, leads to fragmentation near the columnar tip front. Some fragments are transported by the forced convection to the sample centre and remelted there, while many of them are captured by the columnar structure near the sample centre. The modelling result on the mixed columnar-equiaxed structure agrees with the post-mortem analysis of as-solidified sample. As conclusion following impacts of the crystal fragmentation on solidification are suggested: (1) it widens the central segregation channel and promotes the formation of side-arms; (2) it leads to the formation of relatively high volume fraction of equiaxed crystals near the sample centre.

1. Introduction

To investigate the effect of the forced convection during alloy solidification on the evolution of microstructure and macrosegregation, a series of unidirectional solidification experiments on Al-alloys were conducted in a Bridgeman furnace as equipped with the rotating magnetic field (RMF) [1-4]. Under weak RMF or natural convection condition, the sample solidified normally as columnar structure. With the increase of RMF, a transition of microstructure from columnar to equiaxed (CET) occurred in the center part of a sample, whereas in the out-of-center region columnar dendrites still existed. Under strong RMF, a CET occurred in the whole cross section. Formation of fragments under forced convection was believed to be the main source of equiaxed crystals [5-7]. The RMF induced flow has also strong influence on the macrosegregation.

This paper will use a three-phase mixed columnar-equiaxed solidification model [8-10] to simulate the unidirectional solidification of a binary Al-7.0 wt%Si alloy under the RMF. The model was recently extended by incorporating the fragmentation of columnar dendrites as the source of equiaxed crystals [11, 12]. One goal of this study is to verify the numerical model by comparison with the experiments; a more ambitious goal is to study the influence of crystal fragmentation on the formation of microstructure and macrosegregation.

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2. Model descriptions

A three-phase mixed columnar-equiaxed solidification model has been described elsewhere [8-10]. The three phases refer to the liquid melt, equiaxed crystals and columnar dendrite trunks, and their amounts are quantified by their volume fractions, f_{ℓ} , $f_{\rm e}$ and $f_{\rm c}$, respectively. Both columnar dendrites and equiaxed crystals are treated as two separate solid phases. The primary and secondary arm spaces are taken from the experiment ($\lambda_1 = 300 \ \mu m$ and $\lambda_2 = 50 \ \mu m$). The columnar phase is stationary, i.e. $\vec{u}_{\rm c} \equiv 0$; while the motion of equiaxed phase, $\vec{u}_{\rm e}$, is calculated by solving the corresponding momentum conservation equation.

In this study crystal fragmentation is considered as sole origin of equiaxed crystals; heterogeneous nucleation is ignored. The method to treat crystal fragmentation in the mixed columnar-equiaxed solidification model was described previously [11, 12]. The fragmentation-induced mass transfer rate from the columnar phase to the equiaxed phase (M_{ce}) can be calculated by:

$$M_{\rm ce} = -\gamma \cdot \bar{u}_{\ell} \cdot \nabla c_{\ell} \cdot \rho_{\rm e} \tag{1}$$

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where \bar{u}_{ℓ} is velocity of liquid; ∇c_{ℓ} is the liquid concentration gradient; and $\rho_{\rm e}$ is the density of equiaxed phase. A fragmentation coefficient γ is assigned to bridge the unknown contributions to $M_{\rm ce}$. Those unknown contributions to the fragmentation could be the curvature effect of the dendrites, latent heat induced thermal fluctuation, solute diffusion in the interdendritic liquid, etc. The value of γ should be estimated and modified according to some available experimental results. The initial diameter of the fragment ($d_{\rm e,frag}^0$) is assumed to be proportional to λ_2 and local volume fraction of columnar phase ($f_{\rm c}$).

$$d_{\rm e,frag}^0 = \lambda_2 f_{\rm c} \tag{2}$$

As the ideal sphere is assumed for the morphology of equiaxed crystals [12], the production rate of fragments can be calculated with the following equation:

$$N_{\rm frag} = \frac{M_{\rm ce}}{\rho_{\rm e} \cdot \frac{\pi}{6} \left(d_{\rm e,frag}^0\right)^3} \tag{3}$$

In this paper, the mixture concentration, as calculated by equation (4), was used to analyse the macrosegregation.

$$c_{\rm mix} = \frac{c_{\ell} \cdot \rho_{\ell} \cdot f_{\ell} + c_{\rm e} \cdot \rho_{\rm e} \cdot f_{\rm e} + c_{\rm c} \cdot \rho_{\rm c} \cdot f_{\rm c}}{\rho_{\ell} \cdot f_{\ell} + \rho_{\rm e} \cdot f_{\rm e} + \rho_{\rm c} \cdot f_{\rm c}} \times 100\% \tag{4}$$

where, c_{ℓ} , c_{e} , c_{c} and c_{mix} are concentrations of liquid, equiaxed, columnar, and mixture phase respectively; ρ_{ℓ} , ρ_{e} and ρ_{c} are corresponding densities of liquid, equiaxed and columnar phase.

3. Simulation settings

The numerical model is in accordance with the experiments [1], as shown in figure 1. The simulation is carried out in 2D axisymmetry. The cylindrical sample of AlSi7 alloy with a diameter of 8 mm solidifies in an Al₂O₃ crucible unidirectionally. A Dirichlet thermal boundary condition is imposed on the top (T_{Top}) and bottom (T_{Bottom}) of the sample. The outer wall of the sample is considered to be adiabatic. The fragmentation coefficient in equation (1) γ is equal to 0.2 for current study. It was determined through numerical parameter study by fitting the modelling results with the available experimental results. An RMF inductor is installed outside the sample. As shown by the dotted line in figure 1, the RMF is

activated at t = 210 s. An analytical approximation of the azimuthal component of the electromagnetic force (\vec{F}_{θ}) is valid (equation (5)) [13]:

$$\bar{F}_{\theta} = \frac{1}{2} \sigma \omega B^2 r \left(1 - \frac{u_{\theta}}{\omega R} \right) \bar{e}$$
⁽⁵⁾

where σ is the electrical conductivity of the melt, $\omega = 2\pi f$ is angular frequency (f = 50 Hz), B (= 10 mT) is the magnetic induction, r and R (= d/2) is a radial coordinate and radius of the sample, u_{θ} is the azimuthal velocity of the melt at a radial coordinate r, and \bar{e} is the tangential unit vector. The material properties and other parameters used in this study are referred to Zhang *et al.* [14].



Figure 1. Geometry configuration of the simulation domain and applied boundary conditions.

4. Simulation results and discussions

4.1. Modelling results of solidification process

Before the start of the RMF, the thermo-solutal convection is very weak and the alloy solidifies in columnar dendritic structure. Once the RMF is activated at 210 s, some fragments form and develop into equiaxed crystals. Simulation results under the RMF at t = 350 s are shown in figure 2. To assist in the result explanation, the solidification and melt flow pattern are schematically drawn in figure 2(a). Due to the axisymmetry, only the modeling results on the left half of the sample are shown. The pink dotted line represents the axis.

During the solidification process, the solute element (Si) is rejected into the interdendritic melt region, resulting in an increase of solute concentration in the deep mushy region (figure 2(b)). Under the electromagnetic stirring of the RMF, the buoyancy effect is negligible. At the solidification front, the so-called Ekman effect [15] leads to a meridional circulation with the velocity magnitude of 0.014 m/s (vectors in figure 2(c)). The flow pattern in the mushy zone is similar to that in the bulk liquid, but the flow intensity is $2\sim4$ orders of magnitude weaker [14]. This kind of flow transports the solute-enriched interdendritic liquid (figure 2(b)) from the periphery of the sample to the centre, causing the formation of a central segregation channel which is filled with solute-enriched liquid (figure 2(h)).

Figure 2(d) shows the regions where fragments mostly form. Maximum formation rate of fragments reaches a value of 10^9 (1/m³/s) at the solidification front. In the side-arms of central segregation channel,

some fragments also form, but the formation rate is relatively low $(10^5 \sim 10^7 (1/m^3/s))$. The meridional motion of equiaxed crystals (vectors) and the number density of equiaxed crystals (isolines) are shown in figure 2(e). In the bulk liquid region, equiaxed crystals move almost with the same velocity as the liquid.



Figure 2. Solidification process of the sample under the RMF. (a) Schematic of the solidification process and melt flow pattern. (b) Contour of liquid concentration (c_{ℓ}) . (c) Meridional flow of liquid (\bar{u}_{ℓ}) overlaid with isolines of volume fraction of columnar phase ($f_{\rm c}$). (d) Formation rate of fragments ($N_{\rm e}$) overlaid with $f_{\rm c}$. (e) Meridional motion of equiaxed crystals $(\bar{u}_{\rm e})$ overlaid with isolines of number density of equiaxed crystals $(n_{\rm e})$. (f) Contour of mass transfer rate from the liquid phase to the equiaxed phase ($M_{\ell \rm e}$). (g) Contour of volume fraction of equiaxed phase ($f_{\rm e}$). (h) Contour of mixture concentration ($c_{\rm mix}$). To better display the velocity of liquid and equiaxed phase both in bulk liquid region and in mushy zone, length of vectors in (c) and (e) has been scaled by a log function. Contours in (b), (d), (f)-(h) are also shown in colour scale with red for maximum and blue for minimum values.

Near the solidification front, the velocity of equiaxed crystals is rapidly reduced. When the columnar phase reaches a critical volume fraction (assumption 0.2), the equiaxed crystals are captured by columnar dendrites. There is no motion of equiaxed crystals seen in lower left corner of figure 2(e). From the

isolines, it can be seen that most of the fragments $(10^9 \sim 10^{10} \text{ 1/m}^3)$ are captured by the columnar dendrites near the sample centre. The equiaxed crystals, as transported to the sample centre, lead to an accumulation of equiaxed crystals ($\sim 10^{10} \text{ 1/m}^3$) initially; and they are then further transported into bulk liquid region and remelted there. The same fate of remelting occurs to those fragments which form in the side-arm regions of central segregation channel.

If the captured fragments survive, they can continue to grow and interact with the columnar dendrites during the subsequent solidification. The mass transfer (solidification) rate from the liquid phase to the equiaxed phase ($M_{\ell e}$) is shown in figure 2(f). The maximum $M_{\ell e}$ is observed near the solidification front with a value of 3.5 kg/m³/s. From figure 2(e), large number of equiaxed crystals are observed near the sample centre, and $M_{\ell e}$ is also relatively large there.

The final distribution of equiaxed phase fraction (f_e) is shown in figure 2(g). Based on the current simulation results, equiaxed crystals with a volume fraction of approximate 0.05 form near the sample centre, whereas there are almost no equiaxed crystals at the periphery of the sample. Figure 2(h) exhibits the distribution of mixture concentration (c_{mix}). A strong positive segregation channel forms along the axis of the sample.

4.2. Model validation

The effect of the RMF on as-solidified structure and macrosegregation can been seen in figure 3. The distribution of the eutectic phase fraction (f_{eut}) is shown in figure 3(a). Before the RMF is activated, the eutectic phase uniformly distributes between columnar dendrites. After the activation of the RMF, an accumulation of the eutectic phase in the sample centre forms a shape of "fishtail". It is then followed by a tube-shape. Figure 3(b) demonstrates the mixture concentration (c_{mix}), which takes the same distribution as the eutectic phase. At the solidification front, the RMF induced flow sweeps the solute-enriched melt to the sample centre, leading to the formation of this kind of segregation pattern. Figure 3(c) displays the distribution of equiaxed phase fraction (f_e). With the effect of the RMF, some equiaxed crystals form and then are captured by the columnar dendrites. The maximum value of f_e is equal to 0.05. Figure 3(d) shows the as-solidified microstructure in the experiment. The current numerical study cannot quantitatively reproduce the experimental results. However, both numerical and experimental results agree with each other qualitatively.



Figure 3. Microstructure and macrosegregation in the as-solidified sample near the section of RMF switch-on (a) eutectic phase fraction (f_{eut}), (b) mixture concentration (c_{mix}), (c) equiaxed phase fraction (f_e). (d) Experimentally measured microstructure.

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4.3. Influence of fragmentation on microstructure and macrosegregation

In order to study the influence of crystal fragmentation on the microstructure and macrosegregation, an additional case of simulation is performed by assuming no fragmentation. Two cases are compared: one with the fragmentation and one without fragmentation. As shown in figure 4(a), without consideration of fragmentation there is no formation of equiaxed crystals, and the alloy solidifies as columnar dendrites with rest eutectics embedded in the inter columnar dendritic region and in the sample centre. When fragmentation is considered, some equiaxed phase forms near the sample centre; at the periphery of the sample columnar dendrites are dominant. The macrosegregation distribution patterns of both cases are quite similar, as shown in figure 4(b). A strong positive segregation channel form in the centre of the sample, and mixture concentration (c_{mix}) reaches 12.0, which is close to the eutectic composition (12.6); at the periphery, c_{mix} for both cases is equal to 5.5. The main difference in macrosegregation between these two cases can be seen in the side-arm regions of the central segregation channel. Without consideration of fragmentation, a thinner segregation channel forms in comparison to the case with fragmentation. It seems that the crystal fragmentation promotes the formation of side-arms (oscillations in figure 4(b)). As shown in figure 2(d), some fragments form in the side-arm regions. If these fragments are not captured by columnar dendrites, they can be transported by the liquid flow to the bulk liquid region. This fragmentation phenomenon makes more space for the flow and promote the formation of the side-arms. $c_{\rm mix}$ along the radial direction, as drawn by the dash line, is plotted in figure 4(c). Consideration of fragmentation during solidification increases c_{mix} in the central segregation channel.



Figure 4. Numerical parameter study of the influence of fragmentation event on the assolidified structure and macrosegregation. Left-half figures of (a) and (b) shows the modelling result for the case without considering the fragmentation; right-half figures of them shows the modelling result for the case with the fragmentation. (a) Contour of f_e overlaid with isolines of f_c . (b) Contour of c_{mix} overlaid with isolines of the volume fraction of rest eutectic phase (f_{eut}). (c) c_{mix} along the radial direction of the sample.

4.4. Discussion on the formation mechanism of fragments

Figure 5 shows the simulation results on the fragmentation rate and fragmentation-induced columnar-toequiaxed mass transfer rate at t = 350 s. According to the current model, equation (3), the production rate of fragments (N_{frag}) should be proportional to fragmentation-induced mass transfer rate from the columnar phase to the equiaxed phase (M_{ce}). As shown in figure 5(a) and (b), in the regions where M_{ce} is positive, fragments form. Most fragments form at the solidification front, and a small amount of fragments form in the side-arms of central segregation channel. Based on equation (1), if the direction of \bar{u}_{ℓ} is opposite to

 ∇c_{ℓ} , the interdendritic flow leads to local increase of c_{ℓ} , which promotes the formation of fragments. This can be seen from the zoom-in views of zone A and zone B in figure 5(c). In regions where the angle between two vectors (\vec{u}_{ℓ} and ∇c_{ℓ}) is larger than 90°, M_{ce} is positive and some fragments form.



Figure 5. Analysis of the formation mechanism of fragments by solute-driven remelting due to interdendritic flow (t = 350 s). (a) Fragmentation-induced mass transfer rate from the columnar phase to equiaxed phase (M_{ce}), overlaid by isopleths of liquid volume fraction f_{ℓ} (solid lines) and eutectic temperature (dash line). (b) Contour of production rate of fragments (N_{frag}). (c) Zoom-in views of zone A and zone B in (b) with vectors indicating the directions of liquid velocity \vec{u}_{ℓ} (dark red) and liquid concentration gradient ∇c_{ℓ} (yellow).

It is well known that both heterogeneous nucleation and crystal fragmentation are two main origins of equiaxed crystals during alloy solidification. In current study, we only investigated the influence of crystal fragmentation on the formation of microstructure and macrosegregation during unidirectional solidification under forced convection condition. Although fragmentation is considered to be the most important one for the formation of equiaxed crystals under forced convection condition [16], the role of another mechanism by heterogeneous nucleation in the current experiment is not clear. It needs further investigation.

5. Conclusions

A mixed columnar-equiaxed solidification model was used to study the influence of crystal fragmentation on the formation of microstructure and macrosegregation in a unidirectional solidification experiment under the forced convection condition (RMF: rotating magnetic field). Although a simple formulation [11, 12] which is based on the solute-driven remelting as enhanced by the interdendritic flow was used to estimate the fragmentation rate the modelling results showed a very promising agreement with the experiment. New knowledge was obtained. An inward flow forms above the mush under the RMF, leading to the formation of central segregation channel. Solute-driven remelting leads to fragmentation near the columnar tip front and in the central segregation channel. Some fragments are transported by the fluid flow to the sample centre, while some fragments are captured by columnar dendrites near the sample centre. Two important impacts of the fragmentation on solidification are suggested: (1) it widens the central segregation channel and promotes the formation of side-arms; (2) it leads to the formation of relatively high volume fraction of equiaxed crystals near the sample centre.

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