Transient permeability in macrosegregation of static casting in binary alloys: Use of CDF statistical model for analysis

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Abstract. Recently, a statistical method for analyzing macrosegregation predicted by casting simulations or measured in experimental systems has been proposed [1]. The basis of this approach is to collect and order the distribution of nodal concentration values in the final solidified product and thereby create a cumulative distribution function (CDF), presented in a log-log or semi-log format. By considering changes in the shape of the CDF, this statistical tool has been used to study grid convergence and the effect of key physical parameters in 2D casting simulations of the Al-4.5%Cu alloy [2]. These simulations assumed fixed constant representative values for the parameters, in particular a fixed arm dendrite arm spacing in the permeability model for the solid/liquid mushy region. Here the intention is to see how consideration of an arm spacing that coarsens with time (Voller [3]) might influence the shape of the CDF curves. To achieve this, calculations are performed for the recent benchmark experimental study on the Sn-3%Pb alloy presented by Hachani et al [5]. The comparison of the CDF from this experimental benchmark with the one from the simulation, is used to determine the effect of transiently adjusting the permeability during the simulation. Results indicate that an appropriate coarsening model in the permeability calculation of the simulation may improve the comparisons between the measured and simulated CDF. Any level of improvement, however, is on the order of a reasonable expectancy for the resolution of the experimental measurement.

1. Introduction
Macrosegregation, as key defect phenomenon which occurs in many metal casting systems has been widely investigated, both with experiments[4-6], and numerical simulations[7-15]. Recently, Voller and Vušanović [1] proposed a simple statistical tool for comparing experimental and model results. The key is to collect and order the distribution of nodal concentration values in the final solidified product and thereby create a cumulative distribution function (CDF). When it is presented, in a log-log or semi-log form, this CDF curve can be used to determine the volume fraction of the solidified casting above a given level of segregation. In its first application the utility of the method was demonstrated using the experimental measurement data presented in Quillet et al. [6]. The CDF statistical method was subsequently used to study the sensitivity of casting simulation model parameters [2]. In this later work it was shown that the shape and spreading of the CDF curves is, not surprisingly, sensitive to change in the parameters that control the fluid flow in the mushy region. Flow in the mushy region is dominantly driven by thermal and solutal expansions of the liquid phase, while the main resistance to the flow comes from the rigid network of solid dendrites. In most current
simulation technologies the permeability constant ($\kappa_0$) in the Carman Kozeny equation [8] controls the resistance of flow in the mushy region. Typically this is calculated in terms of an assumed fixed secondary arm space (DAS):

$$\kappa_0 = \frac{DAS^2}{180}.$$  \hspace{1cm} (1)

The objective of this paper is to use the CDF tool to investigate the influence of a transient permeability due to the coarsening effects, and thus provide advice for improvements of the mathematical modeling of flow in the mushy region.

2. Physical and mathematical model of benchmark experiment

Recently, researchers [4-6] have performed sets of controlled benchmark casting experiments in a rectangular cavity using binary Sn-Pb and Sn-Bi alloys. These experiments report temperatures, compositions and phase fractions on grid of positions in the final cast. To demonstrate the CDF statistical method we have previously used the Sn-10%Bi alloy measurements [1]. By contrast in this study we use the physical model and casting conditions for benchmark solidification of the Sn-3%Pb alloy performed by Hachani et al. [5]. In this benchmark experiment the alloy was solidified in a rectangular cavity of 100x60x10mm size, using two controlled water cooled heat exchangers. In the simulation model presented here, the material properties of Sn-3%Pb, given in Table 1, were taken from Table E.1 in ref [5]. In addition the imposed boundary temperature conditions used in the simulation matched those reported in [5] for the case of a cooling rate of 0.03Ks\(^{-1}\). The simulation of this system is based on the standard macrosegregation mixture model of mass, momentum, solute and heat conservation introduced by Bennon and Incropera [8]:

$$\frac{\partial \rho}{\partial \tau} + \nabla \times (\rho u) = 0,$$

$$\frac{\partial}{\partial t} (\rho u) + \nabla \times (\rho uu) = \nabla \times (\mu \nabla u) - \frac{\partial \rho}{\partial x} - \mu K^{-1} u,$$

$$\frac{\partial}{\partial t} (\rho v) + \nabla \times (\rho uv) = \nabla \times (\mu \nabla v) - \frac{\partial \rho}{\partial y} - \rho g \left[ \beta_r (T - T^{\text{ref}}) + \beta_L (C_L - C_L^{\text{ref}}) \right] - \mu K^{-1} v,$$

$$\frac{\partial}{\partial t} (\rho C) + \nabla \times (\rho C_L) u = 0,$$

$$\frac{\partial}{\partial t} (\rho H) + \nabla \times (\rho c T + \rho L) u = \nabla \times (k \nabla T),$$

$$K = \kappa_0 \frac{f^3}{(1-f)^2}.  \hspace{1cm} (7)$$

Where $u_L=(u_L, v_L)$ is the liquid velocity, $u=fu_L$ is the mixture velocity assuming no solid movement ($u_s=0$), $f$ is the liquid fraction, $H=cT+fL$ is the enthalpy, $c$ the specific heat, $k$ thermal conductivity, and the temperature $T$ is related to the liquid solute concentration $C_L$ through $T=T_m + m_L C_L$, where $T_m$ is the fusion temperature of Sn and $m_L$ is the slope of the liquidus line. Numerical solution of equation (2)-(7) is obtained using a square grid of control volumes. The fluid flow equations are solved implicitly (with a SIMPLER method for pressure calculation [18]), while the thermal and solutal equations are solved with the time explicit scheme suggested by Voller et al. [16]. This combination of time schemes may be computationally inefficient, but does not induce numerical noise as we observed in using of fully implicit schemes [19].
Table 1. Thermophysical properties of Sn-3wt%Pb alloy

| Property                              | Value      |
|--------------------------------------|------------|
| Fusion temperature $T_m$ (C)         | 232        |
| Latent heat of fusion $L$ (kJ/kg)    | 57.512     |
| Slope of the liquidus line (K/wt%)  | -1.274     |
| Dynamic viscosity $\mu$ (kg/m s)    | 1.737e-3   |
| Density $\rho$ (kg/m³)              | 6965       |
| Thermal expansion coefficient $\beta_T$ (K⁻¹) | 0.95e-4 |
| Specific heat of liquid $c_L$ (J/kgK) | 243       |
| Liquid solutal expansion coefficient $\beta_S$ | -0.53 |
| Specific heat of solid $c_S$ (J/kgK) | 209       |
| Dendrite arm spacing DAS (µm)       | 90         |
| Thermal conductivity $k_L$ (W/mK)    | 33         |
| Partition coefficient $k_{par}$ (-)  | 0.06562    |
| Thermal conductivity $k_S$ (W/mK)    | 64         |

3. Results and discussions

Fig. 1 shows the CDF's for the 50 measured solute concentration values reported for the CR=0.03 Ks⁻¹ experiment in ref [5]. Creating such a CDF from either experimental or numerical simulation results is as follows [1]. Assuming that both experimental data and numerical results of casting simulation are given in a form of a grid with (N) uniform sized control volumes, the nodal segregation value predictions $M_j=C_j/C_0$, on complete solidification, are arranged in descending order; the entry at $j=1$ being the smallest nodal value predicted and the entry at $j=N$ being the largest. Each entry is then associated with an inverse Weibul “plotting position” determined as:

$$F_j = 100 \times \frac{j}{N+1}, \quad j = 1, 2..N.$$  \hspace{1cm} (8)

The y axis of the CDF (in log scale) is the value $100 \times (1-F_j)$ which represents percentage (%) volume fraction greater than a given normalized segregation factor $M=C/C_0$, where $C$ is the value of computed nodal mixture composition (equation [5]), or measured solute, while $C_0$ is the nominal composition.

The x axis is the value of $M$ in a linear scale. Also to indicate that the measurements will not be perfect we have added as dot/dashed lines +/- 5% of the reported measurement. It is emphasized that these are not meant to represent the actual error in the experimental measurements but rather to indicate that errors are present.

![Figure 1. CDF profiles of experimental results [5] and baseline case](image)

Variation of model parameters is shown in Table 2. The first line in this table is our baseline case which matches the case with cooling rate of CR=0.03 Ks⁻¹ reported in [5]. In the first two cases (1 and 2) we vary the permeability constant ($k_0$) through the dendrite arm spacing (DAS) as is given in equation (1). The consistency of this baseline case is established by solving at a finer grid (see case 3 in Table 2). In our previous research [2] we conclude that the liquid flow (driven by thermal solutal contractions) in the mushy region is a key control on the formation of macrosegregation. To
demonstrate this here we conduct a number of cases that differ from out baseline, see Table 2. In tests Case 1 and Case 3 we vary the permeability of the mushy region through changing the fixed value of the dendrite arm spacing (DAS) (see equation (1)). In case 4 we investigate the impact of increasing the thermal solutal expansion coefficients. As we conclude in previous research [2], flow in the liquid and mush is a key parameter that mostly influence on macrosegregation, and it was driven with thermal and solutal contractions of liquid phase. The last case in Table 2 is an attempt to investigate how the use of a variable DAS in the permeability equation (5) might alter the shape of the CDF.

As a model for the coarsening of the secondary arm space here we use:

\[ DAS = a + b \times (1 - f)^C, \]  

This is based on coarsening model originally proposed by Kirkwood [21], where the secondary arm space scales as time \( t^{1/3} \), i.e., their coarsen as time \( t^{-2/3} \). Assuming that solid fraction \( f_s = 1 - f \) is square root of time [21], this would require exponent \( C \) in equation (8) to be \( C=2/3 \).

| Case | Permeability | Micro model | Grid size | \( \beta_T/\beta_{T,\text{nom}} \) (K\(^{-1}\)) |
|------|--------------|-------------|-----------|-------------------------------------------|
| Baseline | DAS=90 µm | Scheil | 100x60 | 1.0 |
| 1 | DAS=60 µm | Scheil | 100x60 | 1.0 |
| 2 | DAS=150 µm | Scheil | 100x60 | 1.0 |
| 3 | DAS=90 µm | Scheil | 200x120 | 1.0 |
| 4 | DAS=90 µm | Scheil | 100x60 | 1.2 |
| 5 | DAS=f(f_s) | Scheil | 100x60 | 1.0 |

Macrosegregation pattern for baseline case with increased grid size is shown on Fig. 2. Cooling is imposed from the right vertical side (see L10 – L30 in ref. [5]) and flow in the cavity has a clockwise direction, with a first formed solid on the right side. Enriched liquid accumulates on the left while the solidification proceeds in time, and the solidification on the left side starts when the temperature of the left side of cavity falls below the liquidus temperature which is now decreased due to the segregation from the right side.

![Figure 2. Macrosegregation pattern for Sn-3%Pb (baseline case + 200x120 grid size)](image)

The last part of domain to solidify has a tree shape 2cm from the left boundary wall and with some channels on the same side, due to the enhanced heat flux (see Fig. 4 in Ref. [5]) around \( t=6300 \) sec, close to the latest stage of solidification. The wide light blue bay near the right top side of casting is
caused by flow reversing and strong re-melting occurring in the early stage of solidification, while the bottom part on the right side remains negative, with less influence of re-melting. It was noted both in similar numerical study of this benchmark experiment performed by Carrozzani et al. [20] that segregation channels are observed in experiment but not in their numerical simulations. As a reason for that they suspect on anisotropy of the mushy zone regarding permeability, which is good driving point to explore this issues in some future study. Our experience with channels is that they can be formed in zone with low heat flux and with increased mixture concentration [19], but the hypothesis of anisotropy is worthwhile and deserve attention.

The CDF curve covering 99% of the casting for the baseline case (DAS=90 µm) together with experimental data taken from Ref. [5] is shown in Fig. 3a. We note that the CDF of the baseline case predictions is a reasonable match to the CDF from the experimental measurement. Our only concern with the results in Fig. 3 is that, in the range $1<M<1.3$, the simulation CDF has a slightly flatter slope when compared with the experimental CDF. In Fig. 3b predicted CDF curves for different and constant permeability values are shown. These results clearly indicate that changes in the permeability have a significant effect on the shape of the CDF, in particular this parameter controls the slope of the CDF in the mid range, $1<M<1.3$. This motivates us to examine if use of the transient arm spacing model of equation (8) in the permeability model, equation (1), might increase the slope of the CDF in the range $1<M<1.3$. The grid convergence of our predictions is confirmed in Fig. 4a which shows that for 98% of the casting the CDF from our chosen grid and one with double the resolution are essentially the same. As we believe that intensity of fluid flow has a key role in macrosegregation formation, we increase for 20% intensity through the increasing of thermal and solutal expansion coefficient ($\beta_T$, $\beta_S$) to test their influence on CDF curve, and results are shown on Fig. 4b. As can be seen, this doesn't help much in negative ($M<1$) and central zone ($M=1$) where we expect a flatter line in the negative zone, followed by narrow gradient in central part. Instead of that we have further extension in positive zone ($M>1.13$) due to the increased flow and therefore macrosegregation.

Figure 3. Statistical CDF profiles for Sn-3%Pb benchmark experiment: influence of different permeability
The results in Figure 5 confirm this, indicating the use of a transient DAS size, consistent with the expected $t^{-2/3}$ coarsening rate, does indeed increase the slope of the CDF in the mid range $1 < M < 1.3$. Indeed it can be argued that the CDF generated by the choice $DAS = 60 + 60(1-f)^{2/3}$ microns is a better match to the experimental CDF.

4. Conclusions

Our intention in this study was to use the recently proposed CDF analysis with the recent reported experimental measurements on an Sn-3%Pb alloy is to investigate how changing the two-dimensional model parameters that control flow in mushy affect the shape and the spread of predicted CDF’s. Our particular interest was to try and understand how modification of the permeability model through control of the secondary arm space would influence simulation predictions. Beyond this we also had
an interest in investigating other flow controlling features such as anisotropic permeability and solid phase motion. In the first place the analysis clearly demonstrates that the chosen size of the secondary arm space in the two-dimensional permeability model can have quite a dramatic influence on the shape and spread of the predicted CDF. And that if an appropriate transient secondary arm spacing is used in the permeability model the predicted CDF more closely follow the CDF of the experimental measurements. It is important to note that these findings are based on a 2-D model and there is some reason to speculate, due to the higher number of degrees of freedom for flow in the mush, that a 3-D model (e.g., see ref [20]) may match experimental observations without the need of a transient arm spacing. In this context, however, it is also important to recognize that, for the particular Sn-3%Pb experimental casting, a basic macrosegregation model, using a constant arm space in a fixed isotropic mushy region, also produces acceptable agreement between simulated and measured CDFs. This has two significant outcomes. First it suggests that the CDF of the Sn-3%Pb macrosegregation measurements represent a first order benchmark against which basic macro segregation simulations codes can be tested. Second it suggests that additional experimental measurements are needed if the CDF tool can produce a signal that will be able to fully test simulation models that incorporate anisotropic moving solid mushy regions.

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