Numerical And Experimental Study Of Macrosegregation During The Casting Of Al-Cu-Mg Alloys

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Abstract
Using a continuum mixture model to simulate the solidification of ternary alloys and a series of casting experiments, the macrosegregation and microsegregation of Al rich Al-Cu-Mg alloy is studied. Calculations were performed for Al-4wt%Cu-1wt%Mg alloy in a square 0.192m x 0.275m domain, actively cooled on one vertical boundary. Considering the diffusion on micro level, three cases have been performed: no diffusion (Scheil case), infinitive diffusion (Equilibrium solidification) and limited diffusion case. Casting of Al-4wt%Cu-1wt%Mg alloy has been performed in order to do comparisons with calculations for both mixture compositions of Cu and Mg and temperature during the casting as well. Temperature of the boundaries of the rectangular mold is recorded at the nine points and is used as boundary conditions in calculations that are assumed as 2-D geometry. Five measurements of temperatures in ingot during the casting were performed and compared with temperatures given by numerical calculations.
1 Introduction

The phase change phenomena, which occurs during the melting and solidification of different alloys is one of the most representative in technical applications. The importance of knowing those phenomena is their high participation in primary energy consumption in the world generally. Therefore, strong demands to get proper quality of alloys structure, makes tasks to the research for better knowledge of processes which occurs during the casting and solidification as well.

Macrosegregation of alloying elements during the solidifications is caused by fluid motion in early stage of solidification. The fluid motion in closed molds is usually caused by thermal and solutial contraction of fluid that occurs due the temperature and compositional gradients in liquid alloy. Once when solidification started alloying elements, which can’t be dissolved in solid was depleted in liquid and increases its composition. This depleted elements of alloy was “advected” by fluid motion in other part of ingot, which is crucial point of macrosegregation. On the other hand, microsegregation occurs on the level of primary and secondary dendrites, and it’s caused by limited diffusion of alloying elements in solid phase. The new solidified layers have different compositions from previous, so the gradients of compositions establish as solidification continues. First attempts in describing of macrosegregation were related with Flemings & Mehrabian works [1]. Their model of microsegregation assumes equilibrium solidification, neglecting the solidification in invariant points and considering only primary and secondary solidification. Using the well-known continuum model [2], developed in last decade of 20th Century, many analysis of two component systems both numerical and experimental was performed, in order to describe phase change liquid-solid process, which occurs with no constant temperature. The energy equation was enthalpy based, so the calculation of temperature was based on modeling of particular phase diagram that is considered. In initial stage of research was focused on two component systems and their simply described phase diagram. The solidification was assumed to be equilibrium, which is far from reality knowing the cooling rates that are applied. But many of alloys which uses in technical applications has more than one alloying element, as steels, so-called super alloys on the nickel (Ni) base, as well as aluminum alloys with cooper (Cu) and magnesium (Mg) as alloying elements. As one of the current research that was performed in recent years, Schneider and Beckermann’s works [3] can be mentioned to. Their model predicts fluid flow, temperature distribution, as well as redistribution of alloying elements for two, five and ten component alloys. They extent basic model developed by Beckermann and Viskanta [4] in order to include more elements in their analysis. Krane and Incropera [5] extended existing continuum model [2] to include third component in analysis. The new model for describing ternary phase diagram was described by different sets of equations for primary, secondary and ternary solidification respectively. Equilibrium solidifications were assumed in calculations of solid phase compositions. The performed results of model were compared with particular casting experiments done with Pb-Sb-Sn alloys.

2 Physical model and experimental rig

Numerical simulation, which has been performed for above, mentioned alloy was based on geometry and conditions that are determinate with experimental rig. Alloy was cast in rectangular mold with 1% of vertical slope, and they dimensions are as follows: L x H w W = 0.192 x 0.275 x 0.122m. Rectangular mold is welded steel construction with diameter holes for boundary temperature measurements around 10mm. The vertical side view of casting rig is shown on figure 1. Metal mold is surrounded with three electrical heaters with gathered power about 8.5kW. On left vertical side water cooled heat exchanger is placed in order to provide vertical side cooling. Between heat exchanger and
mold, slab of soil is placed to provide constant specific heat flux (W/m²K). Thermal conductivity of this soil is around 1.0 W/mK, so the slab around 1.0 cm provides specific heat flux around 100 W/m²K. The distance between bottom of hole and inner side of mold is 1mm, which is quite close to measure temperature at the boundary between alloy and mold.

The first experiment started when alloy is formed and melt at the around 700°C. The water is put in heat exchanger Thermocouple L1 measures air temperature above the liquid metal, while the rest of it (L2, L6) measure temperature of alloy during the casting in exactly particular points at the their ends. The zone at the left bottom part solidified last as was predicted with code developed from mathematical model [7], so the positions for liquid measurements are based knowing this. When solidification ends heaters was turned on until the alloy is melt again.

![Figure 1: Vertical side view of experimental rig](image)

![Figure 2: Experimental boundary temperatures at the vertical cooled side (a) and bottom and opposite side of the mold (b)](image)
After alloy became a melt, thermocouples L2 to L6 was removed, and solidification is repeated again
to final cast. Temperatures on the mold boundaries and in liquid metal are shown on figures 2 and 3
respectively. The solidification starts at around 1700 sec. from the beginning, and it’s marked in slight
changing of slope when some so called re-calescence is present, due to the extract of heat of phase
change. After this short period negative temperature gradient rises up until remain almost constant at
the end of solidification. At the left cooled boundary (temperatures marked 1–4) re-calescence is not
quite well visible due to the strong influence of heat flux sink. Al temperatures are measured using
“K” type of thermocouples and HP 3852A data acquisition system.

3 Mathematical model

Mathematical model for macrosegregation used in this analysis is similar to model proposed by Krane
and Incropera [5]. It’s developed for rigid and stationary solid, which means zero velocities for every
piece of solid wherever is made. Body forces in energy and compositional equations calculate using
the simple Boussinesq approximation, while the solid and liquid density assumes equal. Darcy law [6]
is assumed for fluid flow in so-called “mushy” zone with solid and liquid. Permeability of mushy zone
was calculated using the one of the wide spread model, based on so-called dendrite arm spacing (DAS)
[5]. The main equations of model are as follows:

\[
\frac{\partial p}{\partial t} + \nabla \left( \rho \nabla \right) = 0, \quad (1)
\]

\[
\frac{\partial}{\partial t} \left( \rho \nabla u \right) = \nabla \mu \nabla u - \frac{\mu_1}{K_x} u + \rho \nabla v - \frac{\partial p}{\partial x}, \quad (2)
\]

\[
\frac{\partial}{\partial t} \left( \rho \nabla v \right) = \nabla \mu \nabla v - \frac{\mu_1}{K_y} v + \rho \nabla w - \frac{\partial p}{\partial y}, \quad (3)
\]

\[
\frac{\partial}{\partial t} \left( \rho \nabla h \right) = \nabla \left( \frac{\lambda}{c_s} \nabla h \right) + \nabla \left( \frac{\lambda}{c_s} \nabla (h_s - h) \right) - \nabla \rho \left( \nabla \cdot \left( f_i \rho \left( \nabla V \cdot \nabla V \right) \cdot \left( h_s - h_s \right) \right) \right), \quad (4)
\]

\[
\frac{\partial}{\partial t} \left( \rho \nabla f_{Cu} \right) + \nabla \left( \rho \nabla f_{Cu} \right) = \nabla \left( \rho \nabla f_{Cu} \right) + \nabla \rho \nabla f_{Cu} - \rho \nabla \left( f_{Cu} \cdot f_{Cu} - f_{Cu} \cdot f_{Cu} \right) - \nabla \rho \left( \nabla \cdot \left( f_i \rho \left( \nabla V \cdot \nabla V \right) \cdot \left( f_{Cu} - f_{Cu} \right) \right) \right), \quad (5)
\]

\[
\frac{\partial}{\partial t} \left( \rho \nabla f_{Mg} \right) + \nabla \left( \rho \nabla f_{Mg} \right) = \nabla \left( \rho \nabla f_{Mg} \right) + \nabla \rho \nabla f_{Mg} - \rho \nabla \left( f_{Mg} \cdot f_{Mg} - f_{Mg} \cdot f_{Mg} \right) - \nabla \rho \left( \nabla \cdot \left( f_i \rho \left( \nabla V \cdot \nabla V \right) \cdot \left( f_{Mg} - f_{Mg} \right) \right) \right), \quad (6)
\]

Presented model calculates mixture enthalpy and mixture compositions from energy equation (4) and
two compositional equations (5, 6). The temperature (T), and liquid’s compositions of Cu and Mg (f_{Cu},
f_{Mg}) should be known in order to get closed system of equations, suitable for solving. Those unknowns
calculate from constitutive correlation that is in this case represented by sets of equation that describe
ternary phase diagram of Al-Cu-Mg alloy. A solid that forms during the casting assumes to have zero
velocity, and it form so-called staggered rigid solid structure. This assumption should be taken into
account with caution, because some free floating dendrites can exist in early stage of solidification. In
that case, small solid particles with depleted composition can be advected far from heat sink. More
detailed explanation about modeling of free floating dendrites during the casting of Al alloys can be
found elsewhere [7,8], as well as it occurs during the so-called DC (direct chill) casting. On the right
hand side of momentum equations (2, 3) additional pressure drop caused by solid particles in liquid
flow, is described using well-known Darcy law for pseudo-porous media [6]. Permeability, which is
built in $K_x$ and $K_y$ coefficients, is described using one of the well-known models based on dendrite arm spacing (DAS) approach [5,9]. The dendrite arm spacing was assumed 50µm as in [9]. The second and third term on right hand sides of energy and species equations as well, represents diffusion and advection like source terms. Advection like source terms represents net energy and species exchanging over the control volume and are main responsible for Cu and Mg mixture composition changing. The discretization procedure for diffusion and advection like terms are fully described in [10], and it’s applied in this presented model.

For calculations of temperature and solid’s and liquid’s compositions of Cu and Mg the new model is developed [11], and it takes into account so called open system that allow considering microsegregation with changing of mixture compositions. The changing of liquid’s compositions of Cu and Mg can cause accelerated freezing or re-melting, depending on gradient of changing. Those phenomena (accelerated freezing, re-melting) are well known and recognized in many papers that deal with casting both numerically and experimentally. The more detailed explanation of microsegregation model can be found in [11] and it’s not necessary to elaborate in this paper.

4 Results and discussion

As it mentioned above, numerical simulation of casting process of Al-4wt%Cu-1wt%Mg alloys has been performed, using temperature profiles exposed on figures 1 and 2, as boundary conditions. It was assumed that molten alloy has zero velocity and uniform Cu and Mg compositions at the beginning. Initial temperature of process is around 700°C which is quite above the liquid’s temperature and enough to develop significant flow before solidification.

On figure 3 mixture compositions patterns for Cu and Mg are exposed for solidified alloy. Due to the strong flow against the clockwise depleted zone is formed on the left cooled side with solute rich zone on the bottom right side of ingot. Light green color channels on left side was formed due to the strong remelting in the early stage of solidification when the rich solute liquid dissolves already solidified particles. This phenomenon is well known in open literature related with macrosegregation research [3,4,5]. Some depleted zone is formed at the top right hand side of the mold, and it’s probably caused by low boundary temperature on the top right side of ingot, comparing with middle and bottom zone. The highest compositions of Cu and Mg are detected at the bottom line near the cold surface. The reason for this can be found in fact that richest downflow solute comes from the top in this zone, and became fast solidified, so there is no enough time to allow more advection on the right part of mold. Alloying elements are structured in three phases that precipitates during the solidification process, but dominantly in primary Al “α” phase according the low content of Cu and Mg.

Compositional gradients of Cu and Mg in primary “α” phase are pretty high, so the differences between equilibrium and average values of compositions of Cu and Mg in primary phase are significant. In the following figures (Figures 4 to 6) compositions of Cu and Mg in primary phase are shown at the three different levels (bottom, middle and top of mold) for three different models concerning microsegregation: no diffusion case, equilibrium solidification which assumes infinite diffusion in primary solid phase and limited diffusion with 1-D diffusion model. Above mentioned fact is clearly confirmed on the all three following plots.

As can be observed from figures 4-6 much higher differences between no diffusion case and equilibrium solidification is present for Cu, while the Mg remains lower offset between the two extreme cases? Also, diffusion in primary solid phase shouldn’t be neglected due to the long time of solidification and relatively high temperatures of solid phase as well. Differences between cases for Mg is smaller due to the lower compositional gradients of Mg in primary phase, and higher Mg diffusivity in primary Al comparing with Cu. Binary Al-Mg phase diagram shows strong Mg segregation which is opposite with case with ternary Al-Cu-Mg alloy. Therefore, compiling two
respective microsegregation models for binary alloys (Al-Cu, and Al-Mg) to get microsegregation behavior of ternary alloy can lead to wrong conclusions.

![Figure 3](image1.png)

**Figure 3.** Mixture compositions patterns for (a) Cu, and b (Mg) at the end of solidification

![Figure 4](image2.png)

**Figure 4.** Compositions in primary phase on the bottom of ingot (a) Cu, (b) Mg

Next figure (Figure 7) shows both experimental and calculated cross horizontal values of volume fraction of secondary “θ” phase (Al₂Cu), which precipitates in some part of ingot despite high Al content in presented alloy. Values performed by calculations are significantly lower than experimental dots, especially for bottom line of ingot. The reasons for these differences can be found in several facts. The first might be to high coefficient, which reflects permeability of so called “mushy” zone that can cause no advection with even small fraction of solid. The new calculation with several different values of dendrite arm spacing then is taken from [9] can lead to closer agreement between
calculations and experiment. Other important assumption made in model which can has influence on exposed differences can be assumption of rigid stationary solid during the all stage of solidification. It can be found in published papers [7,8] some research related with applying of different approaches in solidification describing. In early stage of solidification when the solid fraction is low, some solid particles can be advected in lower zone of ingot, gather with enriched liquid. Free floating particles in early stage of solidification will allow more rich solute to flow in lower zone of ingot, than in case with rigid stationary solid. The reason more to confirm above exposed explanation is much better agreement between experiments and calculation in upper zone of mold. The more comprehensive experimental results and their agreement with calculations will be shown in papers that will come soon as we finished experiments with composition measurements.

![Figure 5. Compositions in primary phase on the middle plane of ingot (a) Cu, (b) Mg](image)

![Figure 6. Compositions in primary phase on the top of ingot (a) Cu, (b) Mg](image)

As it known from published papers that deal with macro and micro segregation, macrosegregation models and codes can be successfully implemented in case that mixture composition is demanded, without details about phase compositions [5]. One of the confirmations of this can be profiles shown in the following, where both experimental and calculated profiles for L5 and L6 thermocouples for two diffusion cases are exposed. Practically there are no differences in temperature histories between two diffusion cases, except in later stage of solidification that is caused by numerical instability. Note that
temperature calculates from liquid’s surface equation knowing liquid’s compositions of Cu and Mg. Therefore, agreement between temperatures means at the same time agreement between liquid’s paths during the solidification for two different cases of solidification. The time of solidification both experimental and numerical is well agree too and can be seen from temperature plots.

Figure 7. Experimental and calculated volume fractions of secondary “θ” phase

Figure 8. Temperature profiles at the positions in ingot during the solidification (a) L5, (b) L6

5 Conclusion

The new model for macrosegregation of Al rich Al-Cu-Mg alloys is developed. Model is developed as so-called single domain model, and as enthalpy model which consider mixture enthalpy instead of temperature in energy balance equation. Fluid flow in mushy zone is described using well known Darcy law for pseudo porous media with determinated permeability. Conservation equations of species (Cu and Mg) are written for mixture compositions, and equations contains terms on the right hand side who sucessfully describes interaction between solid and liquid phase. The Al-4wt%Cu-1wt%Mg alloy was casted experimentaly with controled cooling on the left vertical side. Temperatures has been
recorded on four boundaries and it was used as boundary conditions in calculation procedure. The calculations has been performed for three diffusion cases. The main differences between cases is in compositions in primary phase, due to the strong gradients of alloying elements in primary aluminum. Using the equilibrium microsegregation approach to calculate phase compositions can lead to serious errors, while the equilibrium approach can be used for mixture composition calculations successfully. It’s clearly visible from temperatures histories in casted alloy, where practically there is no significant differences between three cases. On figure 7 comparaison between experiments and calculations has been shown for volume fraction of secondary phase. Higher values was obtained by experiment and higher values was obtained on the bottom for both experiment and calculations. The reason for discrepancies can be found in rigid stationary solid approach, which is applied in this case. Maybe some appropriate model with combination of free floating model and rigid stationary model can give much more appropriate results.

References


