Experimental and Numerical Investigations of the Multi-scale Thermoelectromagnetic Convection on the Microstructure during Directionally Solidified Sn-5wt%Pb Alloys

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In this paper, the effect of multi-scale thermoelectromagnetic convection (TEMC) on the microstructure in directionally solidified Sn-5wt%Pb alloys under a transverse magnetic field was studied experimentally and numerically. The experiments are conducted within sample diameters ranging from 0.8 to 12 mm and with various magnetic field intensities. Experimental results show that the transverse magnetic field tilts solid/liquid interface shape and causes the channel segregation. The sloping degree first increases to a maximum value at a critical magnetic field intensity ($B_{\text{max}}$), and then decreases with the increase of the magnetic field intensity. The critical magnetic field intensity ($B_{\text{max}}$) decreases with the increase of sample diameter. Finite-element modeling is performed to simulate the multi-scale TEMC by using COMSOL software. Numerical results indicate that the value of the TEMC increases to a maximum and then decreases with the increase of the magnetic field intensity. The tendency of the simulated TEMC agrees with the evolution process of solid/liquid interface morphology by experimental results. The inter-dendritic TEMC increases monotonically with the increasing of magnetic field intensity in the present study ($B \leq 2$ T). The modification of the solid/liquid interface and the channel segregation under the magnetic field should be attributed to the TEMC at the sample and inter-dendritic scales, respectively.

KEY WORDS: transverse magnetic field; directional solidification; thermoelectromagnetic convection; Sn–Pb alloys; numerical simulation.

1. Introduction

TEMC is based on the principle that thermoelectric current in a fluid can create a driving force responsible for convection in the presence of an applied magnetic field crossing these current.1) Due to fact that the magnetic field is capable of damping the melt convection, the effect of the magnetic field on solidification structure has been investigated for several decades.2–4) However, Boettinger et al.5) found that no influence on microstructure was detected during the directional solidification of the Pb-57wt%Sn alloy under a transverse or an axial magnetic field of 0.1 T. Tewari et al.6) studied the effect of a higher transverse magnetic field of 0.45 T on cellular array in the directionally solidified hypo-eutectic Pb-17.7wt%Sn alloy and the results showed that the cellular array was severely distorted at very low growth speeds (less than 1 μm/s), showing “stripes” of freckles perpendicular to the magnetic field direction. They argued that this phenomenon could be due to the braking of one of the components of the natural thermosolutal convection by the field. Alboussiere7) and Laskar8) systematically studied the effect of the magnetic field on solidifying Bi-60wt%Sn and Cu-45wt%Ag alloys, and suggested that convection could arise during directional solidification in the presence of the magnetic field. They proposed that the convection was produced by the interaction between the magnetic field and thermoelectric (TE) effects. Subsequently, Lehmann et al.9) gave quantitative experiments on the thermoelectric magnetic convection (TEMC) effect. In our previous works, the influence of a static axial magnetic field on liquid/solid interface shape and cellular/dendrite morphology has been investigated, and the experimental results showed that a moderate magnetic field ($B \leq 0.5$ T) caused the formation of the ring-like structure. Theoretical analysis indicates that the TEMC increases to a maximum value and then decreases with the increase of the magnetic field intensity. Later on, the influence of a transverse magnetic field on the macrosegregation and the dendritic growth in the directionally solidified Sn-20wt%Pb alloys11) and Al-7wt%Si alloys12) were investigated. It was found that the transverse magnetic field has modified the solid/liquid interface, and decreased the dendrite spacing and the mushy zone length. However, little attention has been paid to the effect of TEMC at various scales (i.e., sample scales and inter-dendritic scale) during dendrite/cell growth. In this work, the effect of a transverse...
magnetic field on solidification structure at various scales has been investigated experimentally and numerically. Results show that the magnetic field has caused the deflection of the liquid/solid interfaces, segregation channel in the mushy zone. Further, numerical simulation was performed to reveal the flow features of TEMC at various scales. The excellent agreement between TEMC velocities and those obtained from numerical simulated TEMC confirms that TEMC is responsible for the interface shape modification and the formation of channels. This proves the existence of TEMC and their impact during solidification. The purpose of the present work is to deepen the understanding of the effect of magnetic field and convection on the solidification structures during directional solidification by means of solidification experiments and numerical modeling. The experiment is described in Section 2. The results are presented and discussed in Section 3. Some conclusions are drawn in Section 4.

2. Description of the Experiments

The samples of the Sn-5wt%Pb alloys used in this study were prepared with high-purity Sn (99.99%) and Pb (99.99%) in an induction furnace. The alloys were placed in a high-purity graphite crucible and heated to 873 K, magnetically stirred for half an hour. The homogeneous liquid was cast into 0.8, 3, 5, 12 mm diameter rod with a length of 180 mm. The alumina crucible with inner diameter of 0.8 mm was put in an crucible with inner diameter of 5 mm to get a same temperature gradient and less error on the plane cutting. A schematic view of the directional solidification apparatus in the presence of a transverse magnetic field is shown in Fig. 1. The apparatus consists of a static electromagnet, a Bridgman-Stockbarger type furnace and a growth velocity and temperature controller. The electromagnet could produce a transverse static magnetic field with an adjustable intensity up to 1 T. A water-cooled cylinder containing liquid Ga–In–Sn metal (LMC) was used to cool the specimen from below. The furnace chamber is controlled by NiSi–NiCr thermocouples with the precision of ±1 K. The apparatus is designed such that the specimen moves downward while the furnace remains stationary. After 80 mm of steady state growth, the sample was quenched by rapidly withdrawing the crucible into a water-cooled cylinder containing liquid Ga–In–Sn metal. The samples obtained from the experiments were examined in the etched condition (etched with an acid solution: 30 ml CH₂COOH, 10 ml H₂O₂) by the optical microscope.

3. Results and Discussion

3.1. Experiments

Figure 2 shows the optical micrographs of the longitudinal and transverse solidification structure near the quenched liquid/solid planar interface of Sn-5wt%Pb alloy solidified at the growth rate of 10 μm/s and temperature gradient of 78 K/cm under various transverse magnetic fields for the sample diameter of 3 mm. It can be observed that the microstructure is typically dendritic one and the solid/liquid interface obviously protrudes into the melt without magnetic field. The application of the transverse magnetic field tilted the solid/liquid interface to the left side of the sample. The red arrows in Fig. 2 show the estimated depth of the tilted solid/liquid interface. The depth reaches the maximum...
at a 0.3 T magnetic field, and then decreases with the increase of the magnetic field intensity. Moreover, when the applied magnetic field intensity is higher than 0.3 T, with the decreasing of sloping magnitude, segregated channels appear in the longitudinal section (see blue dotted line in Fig. 2. The transverse structures in Fig. 2 also show that, at a higher magnetic field (B ≥ 0.1 T), the left and right side of the sample morphologies are cellular/dendritic and planar structure, respectively. Figure 3 shows the effect of the transverse magnetic field on the longitudinal structures of Sn-5wt%Pb alloys at various sample diameters. The results indicate that a transverse magnetic field also tilts the solid/liquid interface morphology and the maximum sloping value is smaller for a larger sample diameter. It is worth noticing that the sloping degree of channel segregation which is formed at a 0.8 T magnetic field is nearly the same at various sample diameters.

Furthermore, in order to systematically characterize the sloping value of solid/liquid interface and the sloping factor $\Gamma$ is defined as: $\Gamma = h/D$, where $h$ is the measured depth of the sloping solid/liquid interface, and $D$ is the sample diameter. Figure 4 shows the dependence of the sloping factor $\Gamma$ on the magnetic field intensity for sample diameter of 0.8, 3 and 12 mm, respectively. It can be learned that the sloping factor $\Gamma$ reaches a maximum value at a low magnetic field of 0.05 T and decreases with the increase of magnetic field intensity rapidly at a large sample diameter of 12 mm. However, at a small sample diameter of 0.8 mm, the sloping factor still slightly increases at a 0.8 T magnetic field. Above experimental results show that the transverse magnetic field has modified the solid/liquid interface shape and caused the appearance of segregated channels when a higher magnetic field was applied. To further confirm that TEMC caused the solute lead segregation along a horizontal line, the distribution of the Pb contents in the mushy zone was investigated. Figure 5 shows the measured distribution of the Pb contents in the quenched area at the position of 100 μm above the solid/liquid interface and the measurement line is shown in Fig. 5(a). The analysis of the distribution of the Pb contents under various magnetic fields shows that the content of Pb is higher at the left side of sample with the application of a transverse magnetic field. Moreover, the Pb content at the left side of sample is much higher at a 0.3 T magnetic field than other conditions. The above results indicate that a transverse magnetic field has affected the distribution of the Pb solute at the sample scale. Figure 5(b) clearly shows that the right side of the sample is depleted with respect to the nominal composition even ahead of the columnar front, whereas the left side is enriched. This clearly indicates that there is a solute transport due to TEMC from right to left.

It is well known that, in any metallic material, a tempera-
ture gradient $\vec{V}T$ produces a Seebeck electromotive force $\vec{V}T$, where $S$ is the thermoelectric power of the material. Thus, for a cell/dendrite grown by Bridgman furnace, the interaction between the magnetic field and the TE current produces a thermoelectric magnetic force. Furthermore, this force will generate a stress on the solid phase and the TECM in the vicinity.

In the directionally solidified Sn-5wt.%Pb alloys, with the melt above the crystal, due to the thermal-solute convection, the crystal-melt interface is far more protruding than the local isothermal surfaces. Accordingly, the TECM in the mushy zone can be separated to two cases: on the one hand, the macro-scale TECM in the liquid around the dendrite cluster; on the other hand, the micro-scale TECM in the inter-dendritic liquid. Due to these two anisotropy TECMC rolls in the liquid in the mushy zone, the concentration of the solute Pb is transported from one side of the sample to the other side and the sloping solid-liquid interface will be formed.

Based on the results from previous analyses,$^{1,10}$ estimates of the velocity of TECM, $u_l$, can be obtained. For example, by balancing the TEM forces and inertia one may obtain the following estimate valid for moderate magnetic field:

$$u_l = \left( \sigma G S \beta / \rho \right)^{1/2} \quad \text{(1)}$$

where $\sigma$, $G$, $\beta$, and $\rho$ denotes the electrical conductivity, temperature gradient, magnetic field, typical length scale and density, respectively. Equation (1) put forth the thermoelectric power $S$ of the materials. Actually, as far as the Seebeck-Peltier effect near the interface is concerned the relevant parameter is the difference between the thermoelectric powers of the liquid and of the solid $S_{SL} = S_L - S_S$, $L$ and $S$ denoting respectively the liquid phase and the solid one. The value of the TECM increases to a maximum when the magnetic field reaches a critical value and then decreases as the magnetic field still increases. The corresponding magnetic field intensity $B_{max}$ can be derived by equating the braking force to thermoelectric magnetic force (TEMF):

$$B_{max} = \left( \rho G S / \beta \sigma \right)^{1/3} \quad \text{(2)}$$

from which $u_{max}$ is obtained:

$$u_{max} = \left( \left( S G \beta \right)^2 \lambda \sigma / \rho \right)^{1/3} \quad \text{(3)}$$

Below $B_{max}$, the magnitude of TECM increases with increasing magnetic field intensity; above $B_{max}$, TECM decreases as the magnetic field intensity is increased. The units and properties of above physical parameters can be found in Table 1. Table 2 provides a comparison between the numerical values of the maximum magnetic field $B_{max}$ provided by (2), the experimental ones and the computed TEM velocity amplitude (see Fig. 10 below). It is noticeable that the orders of magnitude are in good agreement.

Notably, the thermoelectric power difference ($S_{SL}$) between the solid and melt during directional solidification of the Sn-5wt.%Pb alloys were measured in-situ by Mephisto furnace. The observed Seebeck signal ($E_S$) between the solid and melt during directional solidification can be directly expressed by the equation:

$$E_S = S_{SL} \Delta T = S_{SL} G_L L \quad \text{(4)}$$

| Table 1. Physical properties of the Sn-5wt%Pb alloys used in the evaluation. $^{15)}$ |
|---|---|---|---|---|
| Properties | Magnitude |
| Electrical conductivity of $\beta$-Sn ($\sigma, \Omega^{-1} m^{-1}$) | $4 \times 10^4$ |
| Electrical conductivity of liquid ($\sigma, \Omega^{-1} m^{-1}$) | $2 \times 10^6$ |
| Thermoelectric power between liquid and solid ($\Delta S, V/K$) | $1 \times 10^{-4}$ |
| Temperature gradient ($G, K m^{-1}$) | $7.8 \times 10^3$ |
| Kinematic viscosity ($\nu, m^2 s^{-1}$) | $1.34 \times 10^{-7}$ |
| Density ($\rho, kg m^{-3}$) | $7.0 \times 10^3$ |
| Density ($\rho, kg m^{-3}$) | $7.7 \times 10^3$ |
| Thermal conductivity ($\lambda_s, W/m K$) | 55 |
| Thermal conductivity ($\lambda_s, W/m K$) | 30 |

where $S_{SL}$ is the thermoelectric power difference between the solid and melt, $G_L$ is the temperature gradient, $\Delta T$ is the average undercooling and $L$ is the length of mushy zone. At a given growth velocity, the Seebeck signal should be measurable directly as a function of time. One can learn from Fig. 6 that three different regimes can clearly be identified and correlated to the position of the interface as given by the electrical resistance of the sample. After an initial transient corresponding to the buildup of the solute boundary layer, a steady state occurs with a constant drift. Then, when stopping the interface movement, a final transient is visible allowing the solute boundary layer to re-homogenize in the bulk liquid. The basic principle of the interfacial thermo-electric measurement using the Seebeck technique was schematically presented in Fig. 6(a) and more details of the principle can be found in Refs. 15, 16). Figure 6(b) shows the value of the $E_S$ at the liquid/solid interface as a function of the growth speed. One can learn that the value of the $S_{SL}$ increases as the increase of growth speeds.

At the growth speed of 10 $\mu m/s$ and a given temperature gradient ($G_L = 5 \times 10^3$ K/m) of the Mephisto furnace, the solid/liquid thermoelectric power difference ($S_{SL}$) is on the order of $1 \times 10^{-6}$ V/K, which equals to the value measured by I. Kaldre et al.$^{17}$ Thus, for the present work, the corresponding $B_{max}$ is about 0.3 T for the sample diameter of 3 mm, the interface shape levels off gradually when the applied magnetic field is higher than 0.3 T, which corresponds to the experiment results given in Fig. 2.

### 3.2. Numerical Modeling

To confirm the above theoretical analysis, 3D numerical simulation was performed by using the finite element software COMSOL. We deal with a heuristic model which consists in calculating TECM near an interface between a
solid medium and a liquid one. The liquid/solid interface is prescribed and solidification is not taken into account. The electric currents both in the solid and in the liquid are described by Ohm’s law

$$\mathbf{j}/\sigma = \dot{E} + \mathbf{u} \cdot \mathbf{B} - \nabla \mathbf{T}$$ .................. (5)

with

$$\nabla \cdot \mathbf{j} = 0$$ .................. (6)

where \( \dot{E} \), \( \sigma \), \( \mathbf{u} \), \( \mathbf{B} \) and \( S \) respectively denote the electric field, electrical conductivity, flow velocity, magnetic field flux intensity and Seebeck coefficient. The physical properties \( S \) \( \sigma \) and \( \mathbf{u} \) takes different values both in liquid and solid.

As in previous works, \( \mathbf{u} \) TEMC velocity is governed by incompressible Navier-Stokes equations as:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = -\nabla p + \rho \mathbf{u} \cdot \nabla \mathbf{u} + \mathbf{j} \times \mathbf{B}$$ ............... (7)

with the continuity equation:

$$\nabla \cdot \mathbf{u} = 0$$ .................. (8)

Where \( \rho \) is the density, \( \mu \) is the dynamic viscosity, and \( p \) is the pressure.

Two kinds of computation have been performed, namely at the scale of the sample (macro-TEMC) and inside the mushy zone (micro-TEMC). Figure 7 provides the numerical simulation of the macro-TEMC for a sample diameter of 3 mm. Figure 7(a) shows the 3D geometry with a certain curved interface to simulate the TEMC at the sample scale. The 3D simulation was performed based on the real solid/liquid interface morphology without magnetic field obtained from Fig. 2. Figure 7(b) shows the direction and magnitude of computed TE current around the solid-liquid interface in y-z plane. It is noted that the TE current flows from the liquid to solid phase at the periphery and flows back in the center of the interface and mainly concentrates near the solid/liquid interface. The maximum of the TE current
density reaches $10^3 \text{A/m}^2$. When a magnetic field parallel to the positive y-axis is applied, the TEMC appears as can be seen from Fig. 7(c). The black arrows in Fig. 7(c) show the flow from negative x-axis to positive at the periphery and go up to form a backflow mainly in the bulk liquid center. It is clearly shown that in y-z plane the magnitude of the TEMC at the periphery is higher than the back flow at the top of the interface. Simulated results show that the TEMC at the sample scale will transport the rejected lead solute to left side of the sample and leads to a localized accumulation of solute, which explains the tilting of the solid/liquid interface.

In order to have a clue on the TEMC inside the mushy zone, we have simulated the TEMC passing through a cluster of cells on the inter-dendritic scale under a 0.5 T transverse magnetic field. The cells mimic the dendrite envelopes. Figure 8 shows the geometry as well as the numerical results. For 3D numerical simulation, 21 cells are symmetrically distributed in the cylinder with diameter of 800 µm and length of 2 000 µm (see Fig. 8(a)). For simplicity, the surfaces of cell shapes are defined as Bézier curves rotated around z-axis. The length of the cells is 800 µm, and the distance between two cells is 100 µm. The geometry of typical cell spacing is obtained from previous and present works. Figure 8(a) also shows that the TE current emanates from the tips and passes down to the root. Figure 8(a) shows the TEMC in the mushy zone. In the x-z plane, the liquid in the mushy zone flows from the right side to the left one, parallel to the x direction in the mushy zone and goes up to form a backflow mainly in the bulk liquid in front of mushy zone. The corresponding distribution and direction of TEMC at various positions in the mushy zone is depicted in Fig. 8(c) in order to understand the flow formation under the application of transverse magnetic field. It is obvious that the liquid flows around the cells from negative x-axis to positive in the mushy zone and flow back above the mushy zone. To further display the TEMC in the presence of magnetic field, the magnitude of TEMC at x component from bottom of mushy zone to the top of domain in the x-z plane are plotted in Fig. 8(d). For line B, the magnitude of TEMC increases to a maximum value near the top of mushy zone and then decreases with the increase of z position. For other lines, the tendency regarding the magnitude of TEMC is the same to the line B in the mushy zone. However, it increases to another extremum value and then decreases above the mushy zone with the increase of z position (see Fig. 8(d)). As well as the macro-TEMC, the micro-TEMC is able to transport the solute rejected in the mushy zone from

![Fig. 8. Computed thermoelectromagnetic convection (TEMC) for the inter-dendritic scale during directionally solidified Sn-5wt%Pb alloys under a 0.5 T transverse magnetic field: (a) Mesh for numerical simulation and the thermoelectric current; (b) TEMC in the center of mushy zone in x-z plane; (c) TEMC in x-y plane at various z position: (c1) z = 1 150 µm, (c2) z = 700 µm, (c3) z = 350 µm; (d) TEMC of x component in x-z plane for various positions located in the x-y plane as shown in Fig. 8(c3) (colored surfaces present their magnitudes in µm/s). (Online version in color.)](image-url)
the right side of the sample to the left one. Figure 9 shows the computed TEMC velocity at the transverse plane for a protruding solid/liquid interface (i.e., x-y plane) in the center of mushy zone for the sample diameter of 3 mm. It can be seen that with the increasing of the magnetic field intensity, the velocity of TEMC increases to a maximum value at a 0.3 T magnetic field and then decreases. Further, the TEMC at various sample scales and magnetic field intensities was investigated. The maximum values of TEMC velocity under various conditions is shown in Fig. 10. The maximum values of TEMC obtained by numerical simulation match well with the sloping factor shown by the experimental results. This means that the solute accumulation in the left part of the sample should be attributed both to the micro- and macro-TEMC at the sample scale.

Moreover, the experimental results also revealed that the application of a 0.8 T magnetic field tends to cause the lead-enriched channels at the left side of sample, and the channels show the same slope (see blue arrows in Figs. 2 and 3). This may be attributed to coupled effect between the inter-dendritic TEMC (i.e., micro-TEMC) and draining of lead solute driven by density differences. This concurrent flow will transport solute-rich liquid to the left side of crucible (see Fig. 11). Figure 11(a) illustrates the arrangement of a secondary dendrite attached to a primary dendrite and solute enrichment at the root of the dendrite. E. Liotti et al.\textsuperscript{20} gives the explanation of the local liquid concentration in the dendrite root which will promote the dendrite remelting and breaking. Therefore, the concurrent flow also causes the Pb-rich solute washing the root of dendrite. As the lead solute is transported by the unidirectional inter-dendritic TEMC in the mushy zone, the lead concentration is higher in the left part of sample than in the right one. Then, Pb-rich channel will be formed at the left side of the sample and the cell/dendrite structure will exist in the left side of sample. Meanwhile, the value of inter-dendritic TEMC increases with the applied magnetic field. Thus, the channel will be formed at the left side of sample with a same slope under various sample sizes at a higher magnetic field (B ≥ 0.5 T).
4. Conclusions

The influence of a transverse magnetic field \((B \leq 0.8 \, \text{T})\) on the liquid-solid interface morphology and the solidified microstructures has been investigated during directionally solidified Sn-5wt\%Pb alloy. Experimental results show that the application of a transverse magnetic field has tilted the solid/liquid interface and caused the Pb-rich channel in the mushy zone. We have shown that the sloping degree first increases to a maximum value and then decreases with the increase of the magnetic field intensity in accordance with the theoretical estimates. The critical magnetic field intensity decreases with the increase of sample diameter. Further, 3D numerical simulation of TEMC of various scales was performed based on simple model geometry. The amplitude of the tilted solid/liquid interface under the magnetic field is in good agreement with the values of the TEMC. This proves that both macro- and micro-TEMC which acts in the same direction on the liquid near the solid/liquid interface should be responsible for the modification of the interface. The results provided by numerical simulation also show that the inter-dendritic TEMC increases with the applied magnetic field intensity. The inter-dendritic flow contributes to the formation of channel via the re-melting of dendrite caused by the inter-dendritic TEMC.

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