In situ observation of directional solidification in Ga-In alloy under a transverse DC magnetic field

S He¹,², N Shevchenko¹ and S Eckert¹

¹Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany
²State Key Laboratory of Advanced Special Steel & School of Materials Science and Engineering, Shanghai University, 200072 Shanghai, P. R. China
E-mail: n.shevchenko@hzdr.de

Abstract. The directional solidification of a Ga-25wt%In alloy under the effect of a transverse DC magnetic field is investigated by X-ray radiography. The magnetic field pointing parallel to the X-ray beam is generated by two ring-shaped permanent magnets. The magnetic field reaches values up to ~0.19 T in the field of view. The dendritic growth and the flow patterns of Ga-rich plumes migrating along the solidification front are captured and analysed. It shows that the local fluctuations of solute concentration are partially damped by the magnetic field. At the temperature gradient of 1 K/mm, the growth velocities of solidification front and plumes are not affected. In the case of higher temperature gradient (~2 K/mm), the magnetic field causes an increase of the plume velocity in the horizontal direction and a decrease in the vertical direction while the velocity of the solidification front remains constant. Additionally, it is found that the magnetic field damps the fluctuations of tip velocity and refines the primary arm spacing. Above phenomena are discussed based on the thermoelectric magnetic and electromagnetic braking effects. The in situ experimental data are important for verification and improvement of the existing numerical simulations of solidification under the magnetic field.

1. Introduction

Extensive experimental and theoretical investigations have been performed to reveal the influence of magnetic fields on metals solidification, such as phase migration [1], alignment and orientation of crystal/phase [2], microstructure transition [3], solute redistribution [4] and forced/damped flow in liquid [5]. Particularly, the application of X-ray radiography has attracted considerable attention since it can provide the visualization of alloys solidification [6, 7]. Thus, in situ and real-time observations of microstructure evolution under a magnetic field are widely reported. For instance, many studies have observed the motion of dendrite fragments and equiaxed grains for Al-Cu alloys under a steady magnetic field by means of synchrotron radiation providing the experimental evidence of the thermoelectric magnetic force (TEMF) acting on the solid [8-10]. Meanwhile, natural and forced convection during alloys solidification under various magnetic fields have been preliminarily studied. Wang et al. [11] found that an initially tilted solid-liquid interface was changed into a nearly flat shape together with the motion of Cu-rich melt during directional solidification of Al-Cu alloy under a transverse magnetic field, which uncovered the existence of thermoelectric magnetic convection (TEMC). Cao et al. [12] observed the local movement of Sn-rich plumes showing the forced melt flow induced by applying a traveling magnetic field. For Ga-In alloys, Shevchenko et al. [13, 14] studied the microstructure and the pattern
of Ga-rich plume under the impact of an electromagnetic driven convection. These works extended the understanding of alloys solidification under the magnetic field.

In recent years, a large number of interesting phenomena have been found during the directional solidification under the transverse magnetic field, such as deformation of interfacial morphology, enrichment of solute and modified crystal orientation [15-17]. The effect of the magnetic field on the melt convection has been proposed to be responsible for these phenomena in most publications. Nevertheless, the interrelation between convection and dendritic growth is still an open question. In the present work, directional solidification of Ga–25wt%In alloy under a 0.19 T transverse DC magnetic field is experimentally studied using a microfocus X-ray tube. The dendritic growth and melt convection are in situ observed and quantitatively analysed. Further, possible reasons for the variations of plume motion and dendritic growth under the transverse magnetic field are discussed.

2. Experimental procedures

The schematic diagram of experimental apparatus is shown in figure 1(a). The microfocus X-ray tube (XS225D, Phoenix) produces a horizontally aligned divergent beam. In this work, a voltage of 63 kV and an electric current of 50 μA are chosen. The X-ray image intensifier with CCD camera captures the attenuated X-ray beam passing through the sample and then converts the beam into images with a scan rate of 50 half frames per second and the images are integrated over a period of 1 s to reduce the noise level of single image. More details of the X-ray radiography setup can be found in reference [18]. The transverse DC magnetic field is generated by two ring-shaped permanent magnets (Nd-Fe-B) with an internal intensity of 1.17 T. The inner diameter of the magnet is 10 mm, the outer diameter is 20 mm, the height is 6 mm. An iron core is designed to constrain the magnetic flux to the sample volume. The magnets are fixed on the iron core and the sample is located in between, as shown in figure 1(b). Based on this configuration, both the direction and the distribution of the magnetic field within the field of view (⌀10mm) are numerically simulated using the commercial finite element software COMSOL Multiphysics. The simulation shows that the direction of the magnetic field is approximately perpendicular to the sample surface, as displayed in figure 1(c). Figure 1(d) compares the magnetic field intensities obtained from the simulation and the measurements by a Gauss meter. The good agreement gives rise to the assumption that the distribution of magnetic field is almost uniform within the field of view and the magnetic field intensity B in the sample is approximately equal to 0.19 T.

The Ga–25wt%In alloy prepared with high-purity Gallium (99.99 %) and Indium (99.99 %) is selected because its low melting temperature ($T_{\text{Liquidus}}=25.7$ °C) enables the experiments to be implemented efficiently and flexibly. The alloy is melted and fully mixed in a heating furnace and then filled into a 30×30 mm$^2$ Hele-Shaw quartz cell with a gap of 150 μm for subsequent directional solidification experiments. The heating/cooling system contains two sets of independently controlled Peltier units which are contacted with the top and bottom edges of the cell, respectively. Miniature K-type thermocouples are used to monitor the temperature. The heating/cooling process and temperature gradients setting are controlled by adjusting the temperatures at both ends. During the experiments, the cell is firstly heated to 55 °C at a constant rate of 0.1 K/s, the temperature gradients $G$ are set to 1 K/mm and 2 K/mm. Then, the cell is solidified at a cooling rate of 0.01 K/s. The entire process is recorded by the microfocus X-ray system. To ensure the statistics of the data, each experiment has been repeated three times. After experiments, the image processing procedure follows the algorithm proposed by Boden et al. [18], from which the velocities of solidification front, plume, tip growth and primary dendritic arm spacing can be obtained.
3. Results and discussion
The Ga-25wt%In alloy is directionally solidified from the bottom upwards applying two temperature gradients with and without a transverse magnetic field of 0.19 T. During the directional solidification, the primary Indium dendrites grow while Ga is rejected at the solid-liquid interface into the melt. Since the density of Ga is much lower than that of the initial melt, Ga-rich melt is driven to move upward by buoyancy force and thus solute plumes form. The motion of plumes uncovers the convection pattern in the melt. The morphologies of solidification microstructure and Ga-rich plumes for directionally solidified Ga-25wt%In alloy under various conditions are characterized using X-ray radiography, as shown in figure 2. The colour level represents the solute composition. Red colour refers to the Ga-rich area while blue colour indicates the Indium dendrites. From figure 2, it is obvious that typical columnar dendrites and upward-moving Ga-rich plumes appear for all parameter sets considered here. Moreover, the pattern of Ga-rich plume also exhibits some discrepancies comparing the different experimental conditions. For the experiments at the temperature gradient of 1 K/mm, the effect of the magnetic field on the plumes is imperceptible, most of the plumes show large-scale dimensions and extend particularly in vertical direction, as shown in figure 2(a)-(b). In the case of a temperature gradient of 2 K/mm, the typical patterns of natural convection with applied magnetic field appear much smaller and are limited to a smaller region in front of the solid-liquid interface, as shown in figure 2(c)-(d). It suggests that the transverse magnetic field modifies the pattern of natural convection during directional solidification.
Figure 2. The morphologies of dendritic structure and Ga-rich plumes of directionally solidified Ga-25wt%In alloy at different temperature gradients with and without the transverse magnetic field: (a) $G=1$ K/mm, $B=0$ T; (b) $G=1$ K/mm, $B=0.19$ T; (c) $G=2$ K/mm, $B=0$ T; (d) $G=2$ K/mm, $B=0.19$ T.

Figure 3. Evolution of Ga-rich plumes during the directional solidification of Ga-25wt%In alloy: (a) $G=1$ K/mm, $B=0$ T; (b) $G=1$ K/mm, $B=0.19$ T; (c) $G=2$ K/mm, $B=0$ T; (d) $G=2$ K/mm, $B=0.19$ T, and (e) the horizontal velocities of plumes at different conditions, (f) the vertical velocities of plumes at different conditions.
Figure 3 (a)-(d) show typical X-ray images at successive times during the directional solidification of the Ga-25wt%In alloy under various conditions, in which mobile plumes can be observed. Generally, the movement of plume consists of horizontal and vertical components. Thus, the plume velocities in both of directions are quantitatively analysed, as indicated in figure 3(e)-(f). It is found that the plume velocities in both directions are not influenced by the transverse magnetic field at the temperature gradient of 1 K/mm. In contrast, the application of the transverse magnetic field promotes the horizontal velocity and reduces the vertical velocity when the temperature gradient is 2 K/mm.

Above results demonstrate that the melt convection is changed under the action of the transverse magnetic field. It results in an essential influence on the migration and redistribution of Ga-rich melt in front of the solid-liquid interface. It is well-known that the magnetic field causes an electromagnetic braking (EMB) effect on the melt convection, which has been widely used to suppress the thermal fluctuation and solute segregation during the crystal growth of alloys and semiconductors [19]. In this work, the directions of the plume motion and the magnetic field are perpendicular. As a result, an eddy current will be generated in the conductive melt according to Faraday's law. The interaction between the eddy current and the magnetic field produces a Lorentz force whose direction is opposite to the plume’s motion. This Lorentz force will decelerate the movement of the plumes. Thus, it can be deduced that the EMB effect should be responsible for the decrease in vertical velocity, while the increase in horizontal velocity should be attributed to another reason.

As mentioned in previous publications [5, 11, 20], the existence of the thermoelectric magnetic (TEM) effect near dendritic tip under the magnetic field has been confirmed. It originates from the interaction between the magnetic field and the thermoelectric current (TEC) induced by a temperature gradient and a difference in thermoelectric power $S$ between the liquid and solid phases at the interface. During the directional solidification, the TEM effect generates a forced convection (TEMC) in the melt. The TEMC possibly modifies the movement of the plumes since many experimental and theoretical studies have found that the TEMC induced by the transverse magnetic field can promote the solute transportation in radial direction and finally enhance segregation during directional solidification [11, 21, 22]. Here, due to $S_{\text{liquid}} < S_{\text{solid}}$ [23], it is reasonable to infer that the TEC flows from the melt into the dendritic tip driving a TEMC from right to left (figure 4). This effect explains the migration of Ga-rich plumes along the interface and the enhancement of the horizontal velocity of the plumes. In the more general case of larger samples, the kinetics of plumes will also be influenced by the flow field in neighbouring regions. However, such effects cannot be investigated in this study owing to the geometrical limitations of the solidification cell.

![Figure 4](image1.png)

**Figure 4.** Schematic illustration of flow effects near the dendritic tip under the transverse magnetic field.

![Figure 5](image2.png)

**Figure 5.** The solidification front velocities of Ga-25wt%In alloy directionally solidified at various temperature gradients with and without transverse magnetic field.
In a next step, the behaviour of dendritic growth under the transverse magnetic field is investigated. The mean velocities of the solidification front in different conditions are determined, as shown in figure 5. The results indicate that the solidification front advances faster at the lower temperature gradient. It is known that the relationship between the temperature gradient ($G$), cooling rate ($R$) and solidification front velocity ($V_{\text{Front}}$) can be written as $V_{\text{Front}}=R/G$. Thus, for a given cooling rate a decrease in $G$ will lead to an increase in $V_{\text{Front}}$. In addition, the application of the transverse magnetic field has no significant impact on the solidification front velocity at either of the temperature gradients. Figure 6 shows the variations of the tip growth velocity with solidification time for selected single dendrites. It reveals that the dendritic tip growth velocity always fluctuates with time, while the amplitude of the fluctuation is considerably reduced by the transverse magnetic field. This means, the transverse magnetic field stabilizes the dendritic growth. From the X-ray images, it is obvious that the concentration pattern near the tip becomes uniform by applying the magnetic field. The reduction of local gradients of the solute concentration leads to a smooth tip velocity curve.

**Figure 6.** The relationship between tip growth velocity and solidification time at the temperature gradient of 2 K/mm with and without the transverse magnetic field.

Further, the final primary dendritic arm spacings of Ga-25wt%In alloy directionally solidified at temperature gradient of 2 K/mm with and without the transverse magnetic field are measured, which are 196.44±2.87 μm and 231.55±6.12 μm respectively. The statistic results show that the transverse magnetic field causes the reduction of primary dendritic arm spacing by 15.2 %. The refinement of primary dendritic arm spacing under a magnetic field has been also found in previous publications [24-26]. Based on Kurz’s calculation [27], the primary dendritic arm spacing $\lambda_{i}$ can be written as:

$$\lambda_{i}=4.3(\Delta T_{0}-\Delta T^{*})^{1/2}\left(\frac{D_{\text{eff}}}{\Delta T_{\text{ph}}}\right)^{1/4}V^{1/4}G^{1/2}$$

(1)

where $D$ is the liquid diffusion coefficient, $\Gamma$ is the Gibbs Thompson parameter, $\Delta T_{0}$ is the difference between liquidus and eutectic temperature, $\Delta T^{*}$ is the tip undercooling, $k$ is the equilibrium partition coefficient, $V$ is the growth rate of interface. Although the liquid flow is not considered in Kurz’s calculation, this model still can reflect the general variation of the primary dendritic arm spacing during solidification [28]. In this work, $V$ is found to keep constant (figure 5). $G$ is adjusted by the Peltier elements. The values of $\Gamma$, $\Delta T_{0}$ and $k$ are not affected by the transverse magnetic field [29, 30]. Therefore, the modification of primary dendritic arm spacing may be attributed to the changes of the liquid diffusion coefficient and/or the tip undercooling by the transverse magnetic field. On one hand, the magnetic field accelerates the melt flow in front of the solid-liquid interface and enhances the liquid diffusivity. However, an increase in $D$ would lead to an enlargement of $\lambda_{i}$, which is inconsistent with the
observations. On the other hand, it can be observed that the Ga-rich plumes migrate along the interface under the magnetic field which causes a depletion of Ga concentration near the tip resulting in a higher liquidus temperature and an increase in tip undercooling. According to equation (1), it can be concluded that the increasing $\Delta T^*$ will reduce $\lambda_i$. In addition, due to the low resolution of the images, it is not possible to obtain accurate results of the secondary dendritic arm spacing (SDAS). Thus, the effect of magnetic field on the SDAS is not discussed here.

4. Conclusion
The directional solidification of Ga-25wt%In alloy with and without a 0.19 T transverse magnetic field is experimentally investigated by means of X-ray radiography. In situ observations of Ga-rich plumes show the convection pattern in front of the solid-liquid interface. For $G$=1 K/mm, the plumes are not affected by the transverse magnetic field. For $G$=2 K/mm, the plumes are shifted from right to left and their fluctuation are suppressed to some extent compared with those developing without magnetic field. The magnetic field increases the horizontal velocity of plumes and decreases the vertical velocity. The TEM and EMB effects are deduced to be responsible for the modifications respectively. Further, it is found that the transverse magnetic field refines the primary dendritic spacing and stabilizes the tip growth velocity at $G$=2 K/mm, which should be attributed to the variation of Ga concentration in front of the interface.

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