Observation of dendritic growth under the influence of forced convection

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Abstract. The directional solidification of Ga–25wt%In alloys within a Hele-Shaw cell was visualized by X-ray radioscopy. The investigations are focused on the impact of melt convection on the dendritic growth. Natural convection occurs during a bottom up solidification because lighter solute is rejected during crystallization. Forced convection was produced by a specific electromagnetic pump. The direction of forced melt flow is almost horizontal at the solidification front. Melt flow induces various effects on grain morphology primarily caused by convective transport of solute, such as a facilitation of the growth of primary trunks or lateral branches, dendrite remelting, fragmentation or freckle formation depending on the dendrite orientation, the flow direction and intensity. Forced flow eliminates solutal plumes and damps local fluctuations of solute. A preferential growth of the secondary arms occurs at the upstream side of the dendrites, whereas high solute concentration at the downstream side inhibits the formation of secondary branches.

1. Introduction
Electromagnetic stirring by rotating or travelling magnetic fields can be used as an effective tool for flow control in solidifying metal alloys [1, 2]. Recent publications show that the application of such methods during solidification can lead to homogeneous, fine-grained, globular structures without macrosegregation [2, 3]. Nevertheless, the interaction between solidification and melt flow is rather complex and still needs to be fully understood. A better founded knowledge requires new diagnostic tools which allow in situ observation of the process and provide high resolution data of the flow field, the concentration field and the solidified structure. Moreover, access to different length scales, as for example dendrite morphology and flow structure, is of particular interest. There exist plenty of ex-situ investigations considering the melt flow effects on the dendritic growth [3,4]. Moreover, many in situ experiments with transparent materials as model liquids have been carried out using diverse optical diagnostic techniques [5-8]. However, these model liquids can be extrapolated to metal alloys only to a certain extent due to significant differences in thermo-physical properties.

During the last decade in situ X-ray radioscopy became an important method for visualization of solidification processes in metallic alloys [9,10]. Koster et al. used this method for the investigation of melt convection effects in liquid Gallium - Indium alloys and they were able to reproduce qualitative flow patterns [10]. Shevchenko et al. [11] have shown that the forced melt flow damps local fluctuations of solute concentration. Also a preferential growth of the secondary arms at the upstream side of the primary dendrite trunks was observed. The present investigation is focused on the visualization of the dendritic growth during the bottom-up solidification of a Ga-25wt%In alloy under
the influence of forced convection of different strength. We study the effect of fluid flow on the concentration profiles, the dendrite morphology and growth velocities.

2. Experimental setup

The solidification experiments were carried out at Helmholtz-Zentrum Dresden-Rossendorf. The detailed description of the experimental setup can be found elsewhere [11-15]. Ga–25wt%In alloy (\(T_{\text{Liquidus}}=25 \, ^\circ\text{C}, \ T_{\text{Solidus}}=15.3 \, ^\circ\text{C}\)) used in our experiments was prepared from high-purity Ga (99.99%) and In (99.99%). Than it was melted in a furnace and filled into a 30x30 mm\(^2\) Hele-Shaw quartz cell with a 150 \(\mu\text{m}\) gap parallel to the direction of the X-ray beam (see Figure 1b). A linear array of Peltier elements on the bottom part of the solidification cell cools the alloy and an electric heater mounted at the upper part of the solidification cell heats up the alloy. The temperature gradient across the cell and the cooling rate are controlled by a simultaneous regulation of Peltier cooler and electric heater. A set of miniature K-type thermocouples were contacted to the surface of the cell to monitor the temperature. The cooling rate was 0.01 K/s and the temperature gradient was kept ~ 0.9±0.1 K/mm along the whole experiment. The electromagnetically driven flow was produced by a specific pump, which represents a rotating wheel consisting of two parallel discs with a set of permanent magnets (NdFeB) with alternating polarization at their inner sides. The top part of the solidification cell was placed between the disks. The rotation speeds of the magnetic wheel of 30 and 80 revolutions per minute (rpm) were chosen.

Figure 1. Experimental setup: (a) X-ray diagnostic system; (b) square Hele-Shaw solidification cell equipped with electric heater, Peltier elements and magnetic pump for generation of the melt flow.

The solidification experiments were carried out using a micro-focus X-ray tube (XS225D from Phoenix\(\text{X-ray}\)). The observation window was approximately 9x12 mm\(^2\). The images were captured with a scan rate of 50 half frames per second. The images were integrated over a period of 1 s to reduce the noise level of single images. The tube voltage was 63 kV and the electrical current was 140 \(\mu\text{A}\). The solidification cell was positioned between the X-ray tube and detector as demonstrated in Figure 1a.

The analysis of the solidification front velocity and primary and secondary arm spacing follows the algorithm proposed by Boden et al. [12]. The concentration profiles were achieved from the measurements of the local brightness in the images.

3. Results and discussions

The Ga-25wt%In alloy was solidified in vertical direction starting from the bottom of the solidification cell. Prior to the solidification experiment the alloy was additionally heated up to 20 K above the liquidus temperature to avoid segregations in the melt. Figure 2 shows a sequence of images demonstrating a growth process of In dendrites captured at different time steps. Here it should be noticed that no special preparations were made in our experiments to control the nucleation at the bottom of the solidification cell. This in turn results in the development of ensembles of dendrites with different orientations [11].

Figure 2a is captured before the magnetic wheel was switched on. This image illustrates the natural convection. On the corresponding image one can see well developed solutal plumes which are ejected at the solid-liquid interface since their density is lighter than the initial melt composition. The rising plumes are accompanied by a downward flow of In-rich melt in the intermediate regions. This leads to an inhomogeneous horizontal concentration profile along the solidification front (see the black curve
Figure 2. Image sequence showing the solidification of the Ga - 25wt%In alloy under the forced convection captured at different time steps: (a) 155 s, (b) 425 s, (c) 795 s and (d) 875 s. The magnetic wheel was rotated with 30 rpm.

in Figure 3a). Figures 2b and 2c are captured at two different time steps after the magnetic wheel was switched on. In this experiment the magnetic wheel was rotated contra-clockwise with 30 rpm. The direction of the induced flow is almost horizontal with respect to the solidification front and pointing in clockwise direction. The horizontal velocity component reaches values of ~ 0.3 mm/s. For the chosen rotation speed the solutal plumes are strongly damped and the final concentration profile along the solidification front becomes smooth (see the red curve in Figure 3a). Slight increase of the Ga concentration on the left-hand side of the red curve is caused by smeared plumes running along the solidification front from right to the left. The solidification front velocity decreases gradually from ~ 12.9 µm/s down to ~ 4.5 µm/s during exposure to magnetic pump as it can be seen in Figure 3b. This can be explained by an increasing Ga concentration near the solidification front with exposure time (high Ga concentration near the solidification front is especially pronounced in Figure 5c).

Figure 3. a) Concentration profiles along the solidification front 10 s before (black) and 30 s after (red) switching on the magnetic pump across the line highlighted in Figure 2a. b) Solidification front velocity. The magnetic wheel was rotated with 30 rpm.
Another interesting effect can be observed during solidification: uneven growth of primary dendrites at the beginning of the experiment leads to the formation of Ga-rich zones near the solidification front which develop into distinct segregation freckles as it can be seen on the left-hand side of Figures 2b and 2c. Also a competitive growth between secondary branches and primary dendrites leads to an increase of the primary arm spacing. A similar competition effect is observed for higher order arms: for example the secondary arm spacing is increasing. Moreover, preferential growth of the secondary arms is observed at the upstream side of the dendrites (i.e. on the right hand side of the dendrite trunks). In contrast, high solute concentration at the downstream side strongly influences the formation of secondary arms: in case when dendrites are inclined against the upcoming flow the secondary arms at the downstream side are fully eliminated; in case when the dendrites are inclined towards the upcoming flow only few secondary arms develop and create tertiary arms. Furthermore, the inclination angles of the growing dendrites change by several degrees under the influence of the forced flow. After the magnetic pump is switched off the solute plumes re-appear (see Figure 2d) and the solidification velocity increases (see Figure 3b).

To study the influence of the intensity of the forced flow on the solidification pattern in the next experiment the magnetic wheel was rotated clockwise with 80 rpm. Figure 4 shows a sequence of images captured at different time steps.

![Figure 4](image.jpg)

**Figure 4.** Image sequence showing the solidification of the Ga - 25wt%In alloy under the forced convection captured at different time steps: (a) 260 s, (b) 400 s, (c) 890 s and (d) 960 s. The magnetic wheel was rotated with 80 rpm.

Figure 4a demonstrates the time moment before the magnetic wheel was switched on. The dendritic structure and the flow field are similar to those already shown Figure 2a. As in the previous experiment the plumes are completely suppressed by the induced forced flow after switching on the magnetic wheel (see Figures 4b and 4c). Figure 5a shows corresponding Ga concentration profiles before (black) and after (red) the magnetic pump was switched on. It can be seen that the forced flow induced by higher rotation speed of the magnetic pump flattens the Ga concentration along the solidification front. The solidification front velocity decreases exponentially from ~ 14.2 µm/s down to ~ 2.5 µm/s (see Figure 5b) when the magnetic pump is switched on. Such a fast and strong decrease can be explained by the accumulation of Ga-rich solute with time near the solidification front. In comparison to previous case a pronounced Ga-rich layer develops near the solidification front (see Figure 4c).

Also Ga-rich zones are formed near the solidification front due to uneven growth of dendrites. They evolve with time into distinct segregation freckles (see the right-hand side part of Figure 4b and
The induced strong flow acts like a carrier of In material for growing dendrites. As a result, secondary arms grow preferably at the upstream side of the dendrites (i.e. on the left hand side of the dendrite trunks); while at the downstream side due to high solute concentration only some of the secondary arms develop or are completely of the primary trunk towards the upcoming flow (see the discussion in the first experiment). Increasing arm spacing due to competitive growth between secondary and primary arms is also observed. Moreover, the solute plumes re-appear again after the magnetic pump is switched off (see Figure 4d).

Despite multiple similarities certain differences between the two experiments can be clearly identified: first, at higher rotation speed (at 80 rpm) regions with high In concentration are built up (see Figures 2c and 4c for comparison). The average In concentration was measured in both experiments in the most In-rich areas of 0.52x0.52 mm² size (the corresponding areas are marked as squares in Figures 2c and 4c). A value of ~ 47.5wt% was found for 30 rpm, while a value of ~ 63.6wt% occurs in the second experiment at 80 rpm (the integration regions are marked in Figures 2c and 4c). Moreover, the forced convection reduces the solidification front velocity much stronger in the second experiment. Also the concentration profiles along the solidification front are getting more flat.

4. Conclusions

The bottom-up solidification of Ga-25wt%In alloys within a Hele-Shaw cell was investigated by the X-ray radioscopy. This method allows in situ observation of the flow pattern, concentration field and dendrite growth. We study the effect of forced convection on the growth behavior of the Indium dendrites. The application of the electromagnetically driven flow displaces the typical pattern of natural convection: depending on the intensity of the induced forced flow the local fluctuations of solute concentration are partially or completely damped. The forced flow induces different effects on grain morphology, such as the uneven growth of primary trunks or lateral branches, remelting of single dendrites and also of larger dendrite ensembles, freckle formation, changes the inclination angle of the dendrites and leads to an increasing arm spacing. These effects are primarily governed by the convective redistribution of solute. A more detailed analysis of the melt flow effects is the subject of ongoing work.

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