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Directional melting and solidification of gallium in a traveling magnetic field as a model experiment for silicon processes

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ABSTRACT

Small-scale model experiments for directional solidification processes are performed using a gallium volume with a square horizontal cross-section and dimensions of $10 \times 10 \times 7.5\ \text{cm}^3$. A heater at the top and a cooler at the bottom generate a vertical temperature gradient while an external coil system produces a traveling magnetic field (TMF) leading to Lorentz forces in the melt. The position and shape of the phase interface as well as the melt flow during melting and solidification processes are investigated both experimentally and with a coupled 3D numerical model. Uncertainty in various experimental parameters and appropriate methods of calibration are discussed to enable precise validation of numerical simulations. A distinct influence of the melt flow is observed, which results in a concave melting interface with an upward TMF and a convex shape with a downward TMF. In both cases, the corner region demonstrates local deflections in the opposite directions, which illustrates the challenge to obtain a smooth interface shape in silicon solidification processes. These processes can be further investigated using the validated 3D model. Additionally, direct transfer of the results between model experiments and silicon processes using scaling laws is discussed.

1. Introduction

The directional solidification (DS) process is used in the photovoltaic industry to produce large square silicon ingots with a weight up to 1000 kg. Recent developments of the DS process involve monocrystalline or multicrystalline seeds at the crucible bottom that are not melted and thus allow for a better control of the ingot properties [1]. The melting of the feedstock material starts from the top, and the shape and velocity of the melting interface in the final melting stage is crucial for the seeding process because high interface deflection may result in partly failed seeding. In the solidification phase, the crystallization interface may have local or global deflections which influence thermal stresses in the crystal [2] as well as the convective transport of impurities in the melt and their incorporation into the crystal [3]. Consequently, the shapes of solid–liquid interfaces play an important role in the DS process and influence both the yield of the process as well as the quality of the ingots.

In the silicon melt, buoyancy and Marangoni forces as well as Lorentz forces from external magnetic fields generate a flow, which determines the convective heat transport and hence also interacts with the solid–liquid interface. This interaction has a complex character and may lead to asymmetric flow patterns and highly deflected interface shapes [3]. Numerical simulation is an efficient tool for the understanding and optimization of such complex phenomena especially because in-situ flow measurements are very limited in the case of liquid silicon due to its high melting point. The validation of numerical models can be performed using model experiments with low-melting-point liquid metals. Small-scale setups with square-shaped GaInSn melts up to $10 \times 10 \times 10\ \text{cm}^3$ have been developed for the isothermal case [4] and with a vertical temperature gradient [5]. Various flow patterns generated by a traveling magnetic field (TMF) were observed. Geza et al. [6] developed a larger setup with melt dimensions of $42 \times 42 \times 24\ \text{cm}^3$ using Wood's alloy (Bi–Pb–Sn–Cd) and analyzed the properties of the turbulent flow influenced by alternating magnetic field and vertical temperature gradient. Currently, there are no model experiments for the DS process with direct measurements of the interaction between the melt flow and the solid–liquid interface.

In situ measurements of the solid–liquid interface with a distinct influence of melt convection have been carried out in various low-melting-point metallic alloys, e.g., GaInSn [7], GaZnSn [8,9], GaIn [10], Wood's alloy [11], SnPb [12]. However, a smooth and well-defined solid–liquid interface as in pure silicon cannot be obtained in such systems. Additional effects related to thermosolutal convection and component segregation may also occur as reviewed in [13]. Similar experiments with pure metals such as Sn [14–17] or Al [18] are often

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related to higher working temperatures exceeding 200 °C, which limits the applicable measurement methods. An exception is pure gallium with a melting point of 29.8 °C. It has been used in model experiments to investigate flows driven by buoyancy forces [19,20] and/or by external static [21–23] or time-dependent [22,24–26] magnetic fields. Phase change in gallium has been investigated in the well-known experiment with a temperature gradient between two vertical walls generating an intense buoyant flow [27–29]. Campbell et al. [30] considered a very similar system and carried out real-time interface shape measurements with radioscopy methods, while Ben-David et al. [31] measured the interface position and flow velocity using ultrasonic probes. The influence of a static magnetic field on the melting rate and interface shape has been demonstrated by Zhang et al. [32] who proposed such experiments to investigate melting in low-gravity environments. Brito et al. [33] solidified a small cubic gallium volume and correlated growth conditions with the resulting crystal anisotropy.

This contribution focuses on small-scale model experiments for the DS process considering also the interaction between the melt flow and the solid–liquid interface, for the first time, with gallium as the model substance. The setup described in [5] is developed further to allow directional melting and solidification of gallium in a traveling magnetic field. The melt flow and interface shape for various TMF parameters are directly measured and compared to 3D numerical simulations.

2. Experimental method

2.1. Setup

Fig. 1 shows the present experimental setup – a photo of the gallium container and a cross-sectional sketch. The inner horizontal dimensions of the container are 100 × 100 mm^2, and the sidewalls of the container consist of 15 mm thick polycarbonate (tradename: *Makrolon*®) plates bolted with several screws in the corners. Makrolon is a good thermal insulator with a thermal conductivity of about 0.2 W/mK. In the temperature range below 50 °C, which is relevant for the model experiments, lateral heat losses are negligible so that an almost vertical heat flux can be established across the container.

The bottom of the container is formed by an aluminum cooler with a height of 23 mm, which contains a cylindrical cavity of Ø80 × 8 mm. The heating element is made of copper. It has a total height of 11 mm and contains a squared cavity of 90 × 90 × 4 mm^3. The heater is mounted at a distance of 40 mm from a Makrolon cap, which is fixed by several springs on the top of the container. Thus the container is separated into a growth chamber between heater and cooler and a free volume above the heater, which can be flushed with argon to reduce the oxidation of gallium. The cavities in the cooling/heating elements are connected to *LAUDA* thermostats to control the temperatures on their surfaces. Special connector configurations were developed to achieve uniform temperature fields across the surfaces. For the cooler, two water inlets are located at two opposite sides and two outlets at the other two sides, whereas the heater is equipped with four inlets in the corners and two outlets at side centers. Typical water flow rates of the heater and cooler are 3.3 l/min and 2.3 l/min and typical powers are 3.5 kW (heating) and 1.5 kW (heating)/ 0.18 kW (cooling), respectively. The surfaces of heater and cooler are electrically insulated using a *Thermiflex* foil of 0.07 mm thickness.

The temperature in gallium is measured through the sidewalls of the container with two *type K* thermocouples (diameter of 1 mm) located 2 mm from the heater in the center and 2 mm from the cooler at the side wall. The commercial UDV (ultrasonic Doppler velocimetry) measurement system *DOP2000* from *Signal Processing SA* with an 8 MHz probe (outer diameter of 8 mm) is used for measurements of the axial velocity profile and the position of the phase interface through a central opening in the heater. This opening is also used to detect the interface position by mechanical touching with a quartz rod. The interface shape is obtained with several quartz rods simultaneously after stopping an experiment and removing the heater for a few minutes. In some experiments, the melt is removed for a visual inspection and detailed measurements of the interface shape.

The magnetic field is generated using six concentric, equidistant, circular coils fed from an AC current supply providing a constant phase shift of 60° between adjacent coils. This setup gives rise to a magnetic field traveling vertically upward (TMF up) or downward (TMF down), which is described in detail in [34]. The gallium container was placed in the center of the coil system as indicated in Fig. 1(b). The magnetic field of the coil system was measured using the commercial Gaussmeter *Model 460* from *LakeShore*.

2.2. Gallium as a model substance

Gallium material with 6N purity is used in this study. Despite of flushing with argon gas, gallium showed a strong tendency to oxidation, which resulted in a thin oxide film on the melt surface. This film was periodically removed by mechanical means (without applying any chemicals). This approach ensured an optimal density of scattering particles in the bulk of the melt and continuous UDV measurements up to several hours. It should be also noted that the gallium melt partly showed wetting behavior at the container walls. If the crystal sticks to the container walls, local

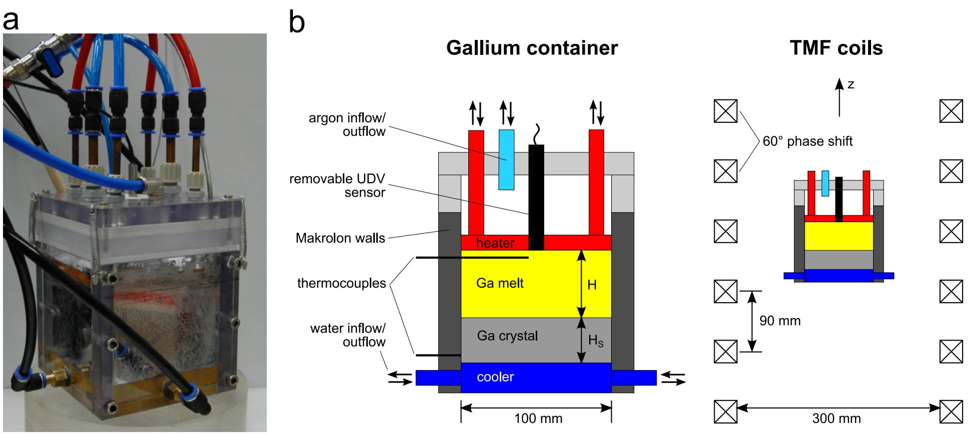

Fig. 1. Photograph of the experimental setup without TMF coils (a); sketch of the entire setup (b).