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Effect of rotational vibrations on directional solidification of high-temperature binary SiGe alloys

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ABSTRACT

Numerical investigation of flows and heat and mass transfer during the directional solidification of high temperature SiGe alloys in the presence of rotational vibrations has been performed. It has been shown that the structure of a vibration-induced flow has the form of a vortex localized near the crystallization front and the direction of this flow is such that the fluid moves along the front from the symmetry axis to the wall of the crucible. With the increase of the vibration parameter the intensity of the vortex grows, and the concentration gradient on the crystallization front near the crucible wall increases. This causes an increase in the crystallization temperature in this region, the temperature of the melt in this area becomes lower compared to the crystallization temperature, and hence the melt becomes overcooled. For the germanium-silicon system, changes in the gravity force lead to the changes in the flow and heat and mass transfer regimes, which are accompanied by a hysteresis. The analysis of the obtained numerical results has revealed that rotational vibrations are responsible for the disappearance of the zone of non-uniqueness.

1. Introduction

Production of high-quality semi-conductor crystalline substrates is nowadays one of the most important and widely used technological process. The vertical Bridgman method is a popular way of producing semiconductor crystals. The principle of this technique is the directional solidification by pulling the crucible containing the melt out of the non-uniformly heated furnace. Convective flows arising in crystal growth processes have a strong influence on the quality of grown crystals. Controlling these flows makes it possible to manage heat and mass transfer in the melt and, hence, to affect the quality of crystals. Melts flows can improve or disimprove the quality of grown crystals. On the one hand, the flows, which promote dopant mixing in the melt, improve the homogeneity of its distribution and, on the other hand, they transport dopant and may therefore disturb the homogeneity of dopant distribution in the grown crystal, generating the regions of local excess or lack of the dopant. From this point of view, the flows are undesirable and should be suppressed. Convective flows during crystal growth by the vertical Bridgman method were studied in a great number of works. In particular, the crystallization of silicon doped germanium, the subject of our investigation, was studied in [1–4]. The temperature of binary alloy crystallization does not remain a constant value; it depends on solute concentration on the crystallization front. This dependence is represented by a liquidus line in the plane $T_m - C$, where T_m is the crystallization temperature, and C is the solute concentration. At low solute concentrations, the liquidus line is well described by the formula $T_m = T_{m0} + mC$, where T_{m0} is the crystallization temperature of a pure material, and m is the empirical coefficient, which characterizes the liquidus line slope.

As the crystallization front moves, excess solute is formed ahead of it. This solute does not have time to transform into the crystal, and therefore a high concentration gradient may appear ahead of the front. As a result, the crystallization temperature increases with a distance from the front. If the velocity of the front motion and hence the concentration gradient are sufficiently high, then the crystallization temperature increases faster than that of the melt. Due to that at a short distance from the front the melt appears to be overcooled and the small perturbations of the front begin to grow. Instability, known as the morphological instability, thus arises [5]. The melt overcooling can be characterized by the parameter $\Delta G = G_T - G_L$, where G_T is the temperature gradient, G_L is defined as $G_L = mG_C$ (G_C - solute concentration gradient). In this case, $\Delta G < 0$ corresponds to the supercooled melt and the

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threshold for the onset of morphological instability is determined from the condition $\Delta G = 0$.

In [6], the onset of morphological instability in a binary system with a low phase transition temperature (succinonitrile-ethanol, SCN-EtOH) was modeled numerically using unsteady quasi-stationary axisymmetric approach. The critical pulling rate at which the morphological instability arises was determined, and the formation of a pit on the symmetry axis of the system was explored. Numerical simulation of flows and heat and mass transfer during the high-temperature semiconductor melt growth was performed in [7].

As shown in [8], the convective motions, which occur in the melt, affect the onset of morphological instability and the solute distribution in the grown crystal. Therefore, a search for better ways to control convective motions during the process of melt solidification is currently an urgent problem. Vibration action was suggested as one of these techniques. For instance, the influence of high-frequency vibrations on the morphological instability of an infinite plane crystallization front that moves with constant velocity was investigated in [9]. It has been found there that, unlike the case of an interface between two immiscible liquids, high-frequency normal vibrations exert a destabilizing effect and the tangential vibrations play a stabilizing role.

Based on these results it was predicted that rotational vibrations about the crucible axis can stabilize the morphological instability of the front during the directional solidification of binary alloys. A theoretical model for flows and heat and mass transfer in the case of the directional solidification of binary systems subjected to high-frequency small-amplitude rotational vibrations was proposed in [10]. The numerical modeling of flows and heat and mass transfer during the directional solidification of transparent binary systems in the presence of high-frequency small-amplitude rotational vibrations in the framework of this model has indicated that such vibrations are able to generate the flow localized near the crystallization front, and the direction of the vibration-induced flow is opposite to that of the gravitational convective flow. Due to the interaction between vibration-induced and gravity-induced convective flows, the gravitational-convective vortex is pushed away from the crystallization front.

The accelerated crucible rotation technique (ACRT) is also efficient for increasing the uniformity of solute distribution on the crystallization front. This modification of the vertical Bridgman method was first suggested in [11].

Numerical and experimental studies of flows and heat and mass transfer during the directional solidification of model transparent binary systems in the presence of high-frequency small-amplitude rotational vibrations [12–17] have shown that these vibrations can generate the flow, which is localized in the neighbourhood of the crystallization front and directed opposite to the gravitational-convective flow; the interaction between vibration-induced and gravity-induced convective flows results in pushing the gravitational-convective vortex away from the crystallization front.

In [18], the problem of suppression of natural thermal convection during the Bridgman crystal growth by translational high-frequency vibrations under terrestrial conditions was discussed, different modifications of the Bridgman method, the vertical Bridgman method and the horizontal configuration of the Bridgman method were considered, and the results illustrating the efficiency of fluid motion suppression in the presence of magnetic fields and vibrations were compared. It has been found that in the case of the vertical Bridgman method the high-frequency horizontal vibrations can significantly suppress the flows nearby the crucible bottom.

In this paper, numerical modeling has been performed to study the influence of high-frequency rotational vibrations on the morphological instability of the front and the solute segregation during the growth of silicon-doped germanium crystals by the vertical Bridgman method. The melt consists mainly of germanium and contains only small amounts of silicon (1%).

2. Problem configuration and governing equations and boundary conditions in the absence of vibrations

We consider the process of directional solidification of a binary melt by the vertical Bridgman method. In this technique, the melt and the crystal are located inside the crucible, which is slowly moved out of the furnace, such that the upper part of the crucible is occupied by the melt and its lower part contains the crystal (Fig. 1). The solidification front remains practically immovable with respect to the heater.

Since the length of the crucible part filled with the melt gradually decreases, the solidification process is non-stationary. However, in most cases the calculations are carried out on the basis of the so-called quasi-static approach, where the pulling rate of the crucible is assumed to be low and the change in the melt region length is ignored. In this case the flows and heat and mass transfer during crystallization are described by the equations of thermosolutal convection. In the reference frame of the heater, these equations have the form:

$$(\vec{v}\cdot\nabla)\,\vec{v} = -\frac{1}{\rho_l}\nabla p + \nu\Delta\vec{v} + g(\beta_T T_l + \beta_C C)\,\vec{\gamma}, \quad \mathrm{div}\,\vec{v} = 0, \tag{1}$$

$$\vec{v}\cdot\nabla T_l = \chi_l \Delta T_l; \quad \vec{v}\cdot\nabla C = D\Delta C, \tag{2}$$

where ρ_l is the melt density, β_T, β_C are the thermal and solutal coefficients of volume expansion, ν is the kinematic viscosity, χ_l is the thermal diffusivity coefficient of the melt, and D is the diffusion coefficient of the solute in the melt. The concentration is defined as the molar fraction.

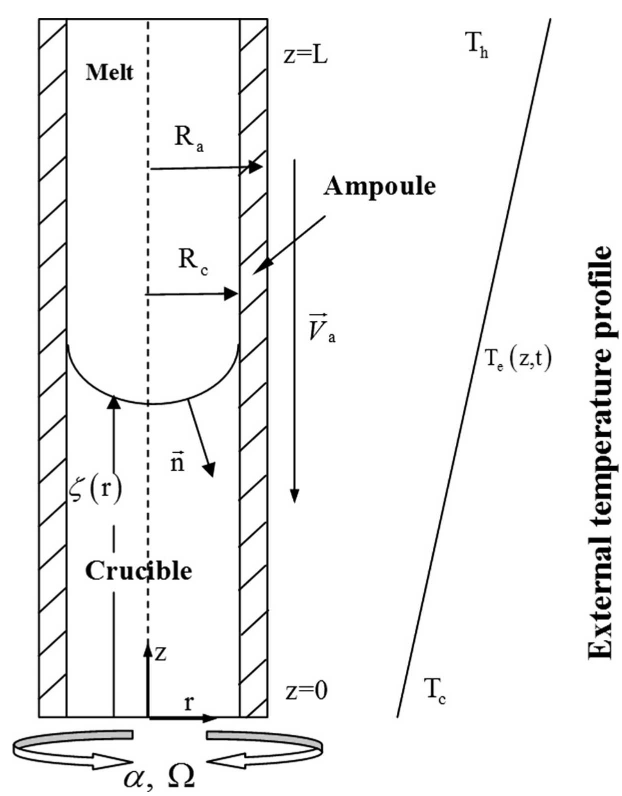

Fig. 1. Problem configuration.