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Numerical Simulation of Solid–Liquid Interface of GaInSb Crystal Growth with Travelling Heater Method

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Abstract: The heat transfer and liquid phase convection during GaInSb crystal growth via the traveling heater method (with a seed) were investigated using numerical simulation to optimize the process parameters and shorten the experimental period in order to produce a high-quality crystal widely used to make various optoelectronic devices. There will be a phenomenon of “thermal impermeability” with an increase in crystal radii for the same furnace temperature profile. The maximum furnace temperature should display an increase of at least 1030 K to 1060 K in order to ensure the successful introduction of the seed with an increase of the crystal radius from 0.01 m to 0.03 m. The interface bending of the solid–liquid interface significantly increases with an increase of the crystal radius from 0.01 m to 0.02 m by about 50%, 67%, and 140%, corresponding to the maximum furnace temperatures 1030 K, 1040 K, and 1050 K, respectively. However, it decreases significantly when the maximum temperature increases from 1030 K to 1050 K, from 0.16 to 0.05, 0.2 to 0.105, and 0.24 to 0.12, corresponding to the crystal radii 0.01 m, 0.015 m, and 0.02 m, respectively. The maximum flow velocity of melt increases slightly with the furnace maximum temperature for the same radius, less than about 6%. However, it increases significantly with the increase of the radius from 0.01 m to 0.02 m, more than 68%.

Keywords: GaInSb crystal; traveling heater method; numerical simulation; temperature field; flow field



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1. Introduction

As both the band gap and the lattice parameters can be regulated by the x value in $\text{Ga}_{1-x}\text{In}_x\text{Sb}$, GaInSb crystal can be widely used as substrate material to make various optoelectronic devices, such as detectors [1,2], lasers [3], satellite navigation [4], and so on. However, it is still quite difficult to prepare a high-quality $\text{Ga}_{1-x}\text{In}_x\text{Sb}$ single crystal. The first challenge is the serious indium element segregation and the second is the large dislocation density, both of which are strongly affected by the temperature field and the convection field during crystal growth.

In recent years, a few works on the growth of GaInSb crystals have been published, in most of which the numerical simulation played an important role. Hayakawa et al. [5] investigated the effect of gravity on both the solid–liquid interface and solute distribution of GaInSb with numerical simulation. The results show that there was a compositional difference in the melting zone, so the distribution of GaSb at the liquid phase and solid–liquid interface were significantly affected by the solute convection. At normal gravity, a large amount of gallium in the upper region of the melting zone moved upward, and the width of the dissolution zone increased in the direction of gravity. Okano Y et al. [6] investigated the effect of natural convection on the melting process of a GaSb–InSb–GaSb system with

numerical simulation. The results show that the contribution of Marangoni convection caused by the change of the temperature to the flow field was greater than that caused by the change of the solute with the influence of gravity. Kimura T et al. [7] investigated the effects of temperature gradient, heating rate, and gravity on the solute dissolution process during the growth of an InGaSb single crystal. The results show that with the increase of the temperature gradient, the content of GaSb dissolved in InSb would decrease, and the shape of the solid–liquid interface changed from asymmetrical to symmetrical. It was easier to realize the stable distribution of both the interface temperature and the solute at a high heating rate in a short time; the dissolved quantity of GaSb decreased with the increase of the heating rate, and the shape of the solid–liquid interface bended seriously at the gravity field. However, with the decrease of gravity level, the shape of the solid–liquid interface gradually flattened, and the distribution of the solute became uniform. Strelov et al. [8] simulated mathematically the heat and mass transfer processes during directed crystallization under terrestrial and space conditions, showing that the necessary conditions for the growth of homogeneous semiconductor crystals were the absence of Marangoni convection, a melt-free surface, and optimization of the temperature gradients. Stelian et al. [9] simulated the solute segregation of GaInSb crystals under natural convection. The results show that the solute segregation and the curvature of the solid–liquid interface could be reduced three times by a graphite baffle in the gravity field. The quartz crucible had lower thermal conductivity compared with that of the boron nitride crucible, so it could effectively improve the chemical uniformity of the GaInSb crystal. Nobeoka et al. [10] simulated the mass transfer during the growth of $\text{Ga}_{0.5}\text{In}_{0.5}\text{Sb}$ in a GaSb–InSb–GaSb interlayer system. The simulation results show that solute diffusion played a major role in the crystal growth and the natural convection existed in the solute at the gravity field, so the dissolution rate of GaSb was enhanced. Mirsandi et al. [11] simulated the growth of GaInSb crystals in a GaSb–InSb–GaSb interlayer system. The simulation results show that the solid–liquid interface shape was affected by the orientation of the GaSb seed crystal, which lead to the difference between the growth rate of the InGaSb crystal and the dissolution rate of the GaSb crystal in the interlayer system. Danilewsky et al. [12] proposed a consistent formulation from micro to macro segregation, revealing the dopant segregation in III-V compound semiconductor crystals grown from melt. Liu et al. [13] applied a rotating magnetic field (RMF) to the vertical Bridgman process for GaInSb crystal growth, significantly reducing both the radial and axial segregations of indium in the GaInSb crystal.

The traveling heater method (THM) has a few merits, and has been a very competitive technique to prepare high-quality crystals of binary or ternary compounds. McCoy et al. [14] compared the advantages and disadvantages of the THM and vertical Bridgman (VB) methods for CdZnTe crystal growth, and attempted to combine their advantages. Zheng et al. [15] prepared CdZnTe crystals with high quality via unseeded THM. Fiederle et al. [16] improved the quality of CdZnTe crystals grown by the THM with microgravity. Derby et al. [17] presented a comprehensive THM model including detailed species transport and phase-change thermodynamics in order to optimize the growth parameters for CdZnTe crystals. Compared with the THM without a seed, the THM with a seed (seed-THM) can significantly improve the single crystal yield of a single ingot, showing greater advantages. However, it requires a more precisely controlled temperature field. If the temperature is too low, the crystal seed cannot be completely melted on the cross section at the initial growth stage, i.e., “thermal impermeability”. While the seed crystal may melt completely if the temperature is too high; the action of crystal seed introduction cannot be achieved in both cases. Moreover, a temperature that is too low may also cause the increase of both the convexity of the solid–liquid interface and the solute segregation in the process of crystal growth. Therefore, it is necessary to accurately control the temperature of the furnace chamber and its temperature gradient to obtain high-quality crystals. It was found that there is very little literature on GaInSb crystal growth using vertical seed-THM after a careful literature search.

In order to optimize the parameters to grow a high-quality GaInSb single crystal, the heat transfer and liquid phase convection of GaInSb crystal seed-THM growth was simulated with the finite element method. The relationship between the crystal radius and the required furnace temperature field, and their effects on the temperature field and the melt convection field, were investigated.

2. Experimental

2.1. Physical Model

The crystal growth system and its geometric data were partially simplified and modified according to the needs of the simulation experiment, which is schematically shown as Figure 1a. GaInSb crystal seed-THM growth is depicted as follows: the seed crystal and the polycrystalline ingot are placed into a high-purity quartz crucible, vacuumed and sealed, and then fixed on a ceramic support in a THM furnace, which can produce an appropriate temperature curve by multiple heaters, as shown in Figure 1b. At the beginning, the top surface of the seed crystal, i.e., the bottom surface of the polycrystalline ingot, is located at the highest temperature point of the furnace; the temperature for the crucible and the seed crystal and the polycrystalline ingot is set to room temperature, and the temperature of each point in the furnace chamber (the temperature profile) remains unchanged. When the heat transfer reaches an equilibrium (for example, the temperature field in the seed crystal domain does not change within 60 min) the melting state of the seed crystal in both the radial and axial directions is observed. After the seed crystal is successfully introduced, the temperature field begins to move upward at a rate of 0.5 mm/h.

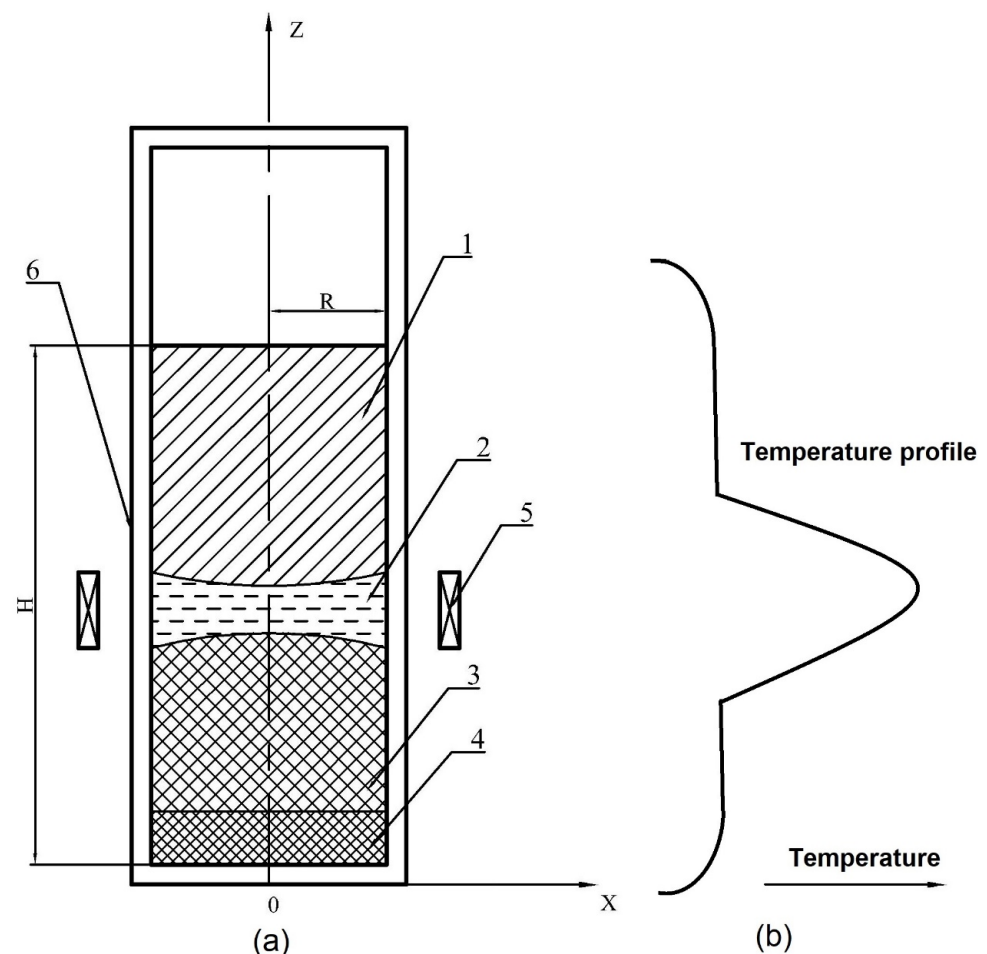


Figure 1. Diagram of the crystal growth system (a) and the furnace chamber temperature curve (b): 1, polycrystalline ingot; 2, melting zone; 3, crystal; 4, initial seed crystal section; 5, heater; 6, crucible.

The process of GaInSb crystal growth via seed-THM is very complex. For example, when the resistance wire is used to heat the quartz crucible, the electric energy transforming to heat energy should be considered; thermal radiation and air convection exists between the furnace and quartz crucible, there are heat transfers between crucible and polycrystalline materials, latent heat of crystallization will be released, etc.

To construct an appropriate numerical simulation model, it is required to perform a few reasonable simplifications. Below are our main assumptions and simplifications:

- (1) Both the bottom and the top THM furnace are sealed very well, and the gap between the crucible and the furnace chamber is small enough that the air convection can be ignored. There is only a radiant heat transfer. The top and bottom of the quartz crucible transfer heat with the furnace chamber only via radiation. In this work, we ignore the air convection heat transfer and air heat conduction between the inner wall of the furnace and the crucible, which will certainly bring a certain degree of error. We will establish a more complete calculation model including air convection heat transfer in our future work.
- (2) All materials' physical properties are isotropic; some properties are dependent on the temperature.
- (3) The THM system is axisymmetric; thus, a two-dimensional model is reasonable and practical. Figure 1 shows the coordinate system; only the right half is taken as the simulation domain.
- (4) The solid–liquid interface is presented with the melting point isotherm.

2.2. Control Equation

2.2.1. Energy-Governing Equation

The energy conservation equation is as follows:

$$\frac{\partial q_i}{\partial \chi_i} + Q - \rho c \frac{\partial T}{\partial t} = 0 \quad (1)$$

where Q is the latent heat per unit mass and per unit time; T stands for the temperature; q_i for the heat flux vector component; and c , ρ , t , for the specific heat, material density, and time, respectively.

The temperature gradient and the heat flux density satisfy the formula:

$$q_i = -\lambda_{ij} \frac{\partial q_i}{\partial \chi_i} \quad (2)$$

where λ_{ij} is the heat transfer sensor component in a specified direction. For isotropic materials, the values of thermal conductivity in different directions are consistent.

Substituting Equations (1) into (2), the heat transfer equation in a volume unit becomes:

$$\frac{\partial}{\partial \chi_i} (\lambda_{ij} \frac{\partial q_i}{\partial \chi_i}) + Q - \rho c \frac{\partial T}{\partial t} = 0 \quad (3)$$

The heat transfers between the melt, crystal, or crucible during crystal growth obey Equations (4)–(6), respectively:

$$\rho_m C_{p,m} \frac{\partial T}{\partial t} + \rho_m C_{p,m} (\vec{u} \cdot \nabla) T = \nabla (k_m \nabla T) \quad (4)$$

$$\rho_c C_{c,m} \frac{\partial T}{\partial t} = \nabla (k_c \nabla T) \quad (5)$$

$$\rho_a C_{a,m} \frac{\partial T}{\partial t} = \nabla (k_a \nabla T) \quad (6)$$

where c stands for the specific heat, k for the thermal conductivity, μ for the velocity vector, and ρ for the material density. The subscripts “ c ”, “ m ”, and “ a ” indicate the crystal, the melt, and the ampoule, respectively.

$$\nabla = \frac{1}{r} \frac{\partial}{\partial r} i + \frac{\partial}{\partial z} j \quad (7)$$

The temperature field boundary condition: the left boundary is the crucible symmetry axis $r = 0$, where $\partial T / \partial r = 0$. The other three boundaries of the calculation domain are the right, upper, and bottom of the crucible, respectively, all of which display radiant heat transfers with the furnace if we ignore the heat conduction between the ceramic support and the crucible bottom. Therefore, we can express the heat flux intensity as the following:

$$q_{12} = \varepsilon \sigma F_{12} (T_1^4 - T_2^4) \quad (8)$$

where ε stands for the complex emissivity; σ for Boltzmann constant; T_1 for the crucible surface temperature and T_2 for the furnace inner surface temperature; and F_{12} stands for the viewing angle coefficient, dependent on the relative position of two surface units:

$$F_{12} = \frac{1}{A} \int A \int A_1 \int A_2 \frac{\cos \varphi_1 \cos \varphi_2}{\pi r^2} dA_2 dA_1 \quad (9)$$

2.2.2. Momentum Governing Equation

The conservation condition of momentum in the process of fluid flow is as follows:

$$\Delta F_{va} = F_{in} - F_{out} + \Delta F_f \quad (10)$$

where ΔF_{va} stands for the momentum variation per unit time of a differential unit, F_{in} for the momentum flowing into the unit, F_{out} for the momentum flowing out the unit, and F_f for the work done on the unit by the external force.

The momentum balance expression in the x axis is as follows:

$$\frac{\partial}{\partial \tau} \rho v_x = - \left(\frac{\partial}{\partial x} \rho v_x v_x + \frac{\partial}{\partial y} \rho v_y v_x + \frac{\partial}{\partial z} \rho v_z v_x \right) - \left(\frac{\partial}{\partial x} \tau_{xx} + \frac{\partial}{\partial y} \tau_{yx} + \frac{\partial}{\partial z} \tau_{zx} \right) - \frac{\partial P}{\partial x} + \rho g_x \quad (11)$$

where the first term on the right side of the equation is the momentum form of the fluid flow in the x -axis direction and v_x , v_y , and v_z , are the flow rates of the fluid in the x , y , and z , directions, respectively. The second term on the right side of the equal sign is the work done by the external force to the differential unit of the object. τ_{xx} is the normal stress that is perpendicular to the plane of the x axis, τ_{zx} is the shear stress that is perpendicular to the plane of the z axis, and τ_{yx} is the shear stress that is perpendicular to the plane of the y axis. The third term on the right side of the equal sign is the change of the x axial pressure. The fourth term is the work done by a differential element in the gravity field, in which g_x is the component of gravity acceleration in the x axis direction.

The momentum balance expressions in the y axis direction and the z axis direction are as follows:

$$\frac{\partial}{\partial \tau} \rho v_y = - \left(\frac{\partial}{\partial x} \rho v_x v_y + \frac{\partial}{\partial y} \rho v_y v_y + \frac{\partial}{\partial z} \rho v_z v_y \right) - \left(\frac{\partial}{\partial x} \tau_{xy} + \frac{\partial}{\partial y} \tau_{yy} + \frac{\partial}{\partial z} \tau_{zy} \right) - \frac{\partial P}{\partial y} + \rho g_y \quad (12)$$

$$\frac{\partial}{\partial \tau} \rho v_z = - \left(\frac{\partial}{\partial x} \rho v_x v_z + \frac{\partial}{\partial y} \rho v_y v_z + \frac{\partial}{\partial z} \rho v_z v_z \right) - \left(\frac{\partial}{\partial x} \tau_{xz} + \frac{\partial}{\partial y} \tau_{yz} + \frac{\partial}{\partial z} \tau_{zz} \right) - \frac{\partial P}{\partial z} + \rho g_z \quad (13)$$

The constrained boundary conditions are as follows:

- (1) The interface between crystal and melt (the bottom boundary) and the interface between melt and crucible (the right boundary) are non-slip rigid boundary conditions.

The velocity component that is perpendicular to the interface is zero; the velocity that is parallel to the interface is equal to that of the solid.

$$v_z|_{z \rightarrow 0} = 0 \quad (14)$$

$$v_x|_{z \rightarrow 0} = v_{xS} \quad (15)$$

$$v_y|_{z \rightarrow 0} = v_{yS} \quad (16)$$

where v_{xS} and v_{yS} are the rate of motion components of the solid surface in the x and y directions. If the solid is stationary:

$$v_x|_{z \rightarrow 0} = 0 \quad (17)$$

$$v_y|_{z \rightarrow 0} = 0 \quad (18)$$

- (2) The melt-free surface (the top boundary) is a free slip boundary condition. The shear stress that is parallel to the surface is zero, so that the velocity gradient component is zero, and the boundary conditions of the surface are as follow:

$$\frac{\partial v_x}{\partial z}|_{z \rightarrow l_0} = \frac{\partial v_y}{\partial z}|_{z \rightarrow l_0} = 0 \quad (19)$$

$$v_z|_{z \rightarrow l_0} = 0 \quad (20)$$

2.2.3. Physical Parameters of the Crystal Growth System

Tables 1 and 2 list the material properties and the geometric data used in the numerical simulation, respectively, all of which utilize units belonging to the International System of Units. Table 3 lists the geometric data of the crystal growth system.

Table 1. Physical parameter of $\text{Ga}_{1-x}\text{In}_x\text{Sb}$ [18–20].

Parameter	Symbol	Condition (Temperature Range)	Value
Thermal conductivity	λ ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)	Solid	7.8
		Liquid	17.1
Density	ρ ($\text{kg}\cdot\text{m}^{-3}$)	273 K < T < 1500 K	$5.59 \times 10^3 + 0.47 T$
Specific heat	C ($\text{J}\cdot\text{K}^{-1}\cdot\text{kg}^{-1}$)	Solid	0.3×10^{-3}
		Liquid	0.33×10^{-3}
Heat of fusion	ΔH ($\text{J}\cdot\text{kg}^{-1}$)		3.13531×10^5
GaSb melting point	T (K)		0.98×10^3
Viscosity	μ ($\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}$)	Liquid	$2.3 \times 10^{-3} + 2.76 \times 10^{-4} T - 3.39 \times 10^{-7} T^2$
Electric Conductivity	σ_E ($\Omega^{-1}\cdot\text{m}^{-1}$)	Liquid	1×10^{-6}
		Solid	5×10^{-6}
Thermal expansion coefficient	β_T (K^{-1})	Liquid	1×10^{-4}

Table 2. Physical properties of quartz crucible [21].

Parameter	Symbol	Temperature	Value
Thermal conductivity	λ ($\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)	273 K < T < 1500 K	$1.25 + 1.05 \times 10^{-3} T - 3.95 \times 10^{-7} T^2 + 7.33 \times 10^{-11} T^3$
Density	ρ ($\text{kg}\cdot\text{m}^{-3}$)		2200
Specific heat	C ($\text{J}\cdot\text{K}^{-1}\cdot\text{kg}^{-1}$)	273 K < T < 1500 K	$760 + 0.634 T - 2.39 \times 10^{-4} T^2 + 4.44 \times 10^{-8} T^3$
Emissivity	ε		0.35

Table 3. Geometric parameters of the crystal growth system.

Parameter	Symbol	Value
Seed length	L_s (m)	0.02
Seed radius	R_s (m)	0.01~0.03
Polycrystalline material length	L_r (m)	0.15
Polycrystalline radius	R_r (m)	0.01~0.03
Furnace length	L (m)	1.2
Furnace chamber emissivity	ε	0.7

3. Numerical Results and Analyses

3.1. The Temperature Field at the Initial Stage

In order to achieve seed introduction during GaInSb crystal growth with seed-THM, four numerical samples have been carried out to investigate the relationship between the crystal radius and the required maximum furnace chamber temperature, as shown in Table 4. The melting points of GaSb and InSb are 980 K and 798 K in the binary pseudo-phase diagram of InSb-GaSb, respectively. The melting point of the GaInSb crystal can be adjusted by changing the molar content of indium. Herein, the content of indium is set to zero percent in the temperature field simulation calculation. Therefore, the isotherm line at 980 K is regarded as the solid–liquid interface in the process of crystal growth.

Table 4. The crystal radius and the furnace maximum temperature.

Examples	Crystal Radius R (m)	Furnace Maximum Temperature T_{MAX} (K)
(a)	0.01	1030
(b)	0.02	1030
(c)	0.03	1030
(d)	0.03	1060

The temperature field of the melt zone at the initial stage of crystal growth is simulated as shown in Figure 2. In the initial stage of crystal growth, the upper half of the seed crystal needs to be melted completely in the radial direction, but the lower half should be prevented from melting completely in the axial direction. In this model, the length of the seed crystal is 0.02 m, and the height of the melting zone is 0.04 m. In Figure 2, the colored area is for the melt and the gray area is for the solid. The maximum furnace temperature T_{MAX} is 1030 K in the samples (a), (b), and (c). In the sample (a), the melting zone height on the central axis is 0.032 m. In the sample (b), the melting zone height on the central axis is 0.0263 m. It is clear that the seed or polycrystalline rod fully melted in the radial direction in Figures 2a and 3b while it did not in Figure 2c. The melting zone height of (a) is higher than that of (b); we can conclude that a decrease in the crystal radius can increase the height of the melting zone at the same furnace temperature.

It is clear that there is a “thermal impermeability” in the sample (c), i.e., the upper half of the seed crystal does not melt completely in the radial direction, even if the temperature is kept constant for a long time. In the sample (d), when the maximum temperature of the furnace increases to 1060 K, the “thermal impermeability” disappears, though the melting zone height on the central axis is small, about one third of that on the crucible inner wall, and half of that on the central axis in Figure 2a. This means that the maximum temperature of the furnace should be higher than 1060 K so as to obtain a flatter crystal–melt interface. However, if the maximum temperature of the furnace chamber is too high and remains stable for too long, the seed crystal will all melt completely. In an actual crystal growth process, both the “thermal impermeability” and the “melting disappearance” of the seed crystal should be avoided as much as possible, which can be achieved faster and more accurately with the help of computational simulation.

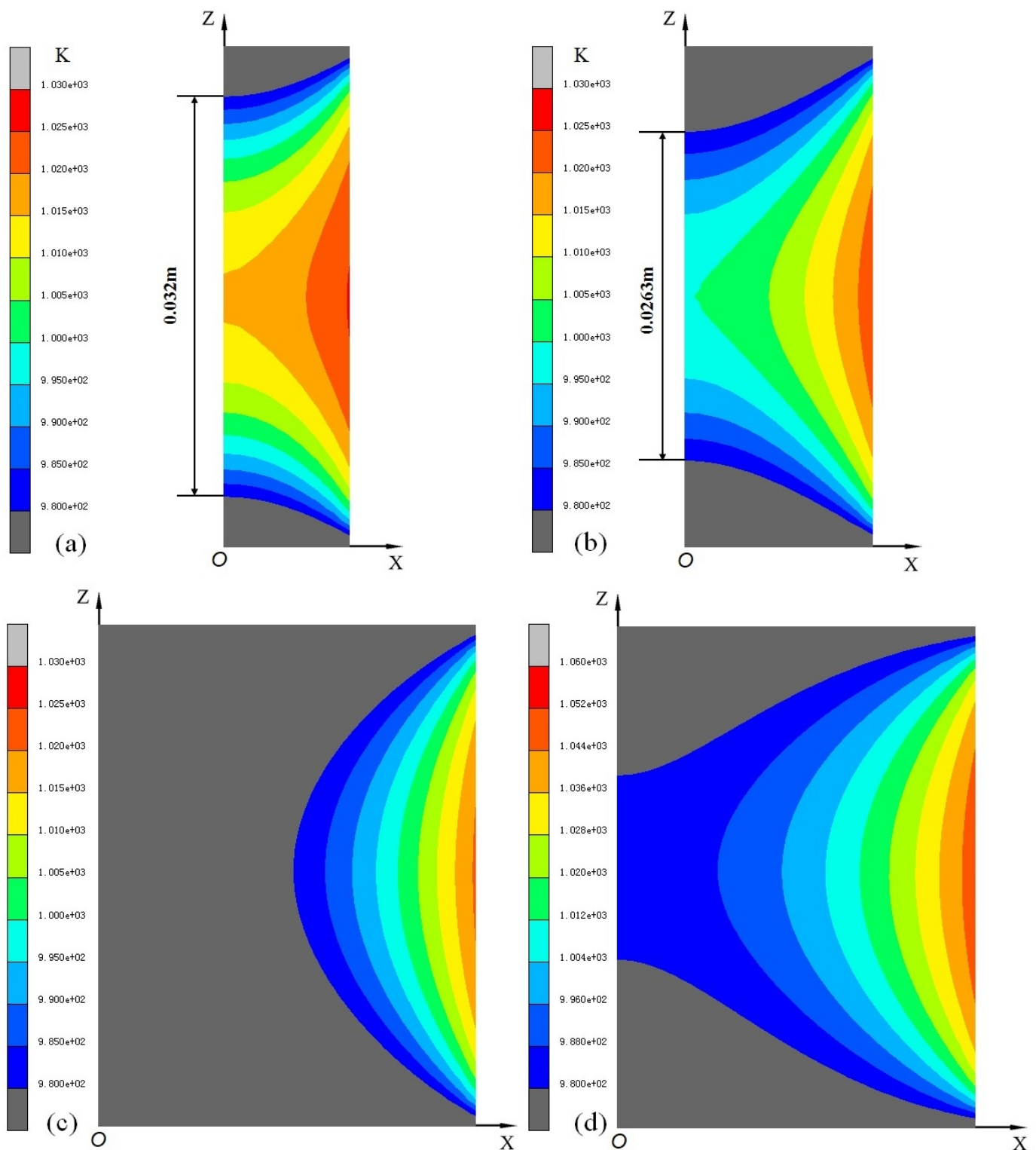


Figure 2. The temperature field at the initial stage of crystal growth: (a) the crystal radius is 0.01 m and the maximum furnace temperature is 1030 K; (b) the crystal radius is 0.02 m and the maximum furnace temperature is 1030 K; (c) the crystal radius is 0.03 m and the maximum furnace temperature is 1030 K; (d) the crystal radius is 0.03 m and the maximum furnace temperature is 1060 K.

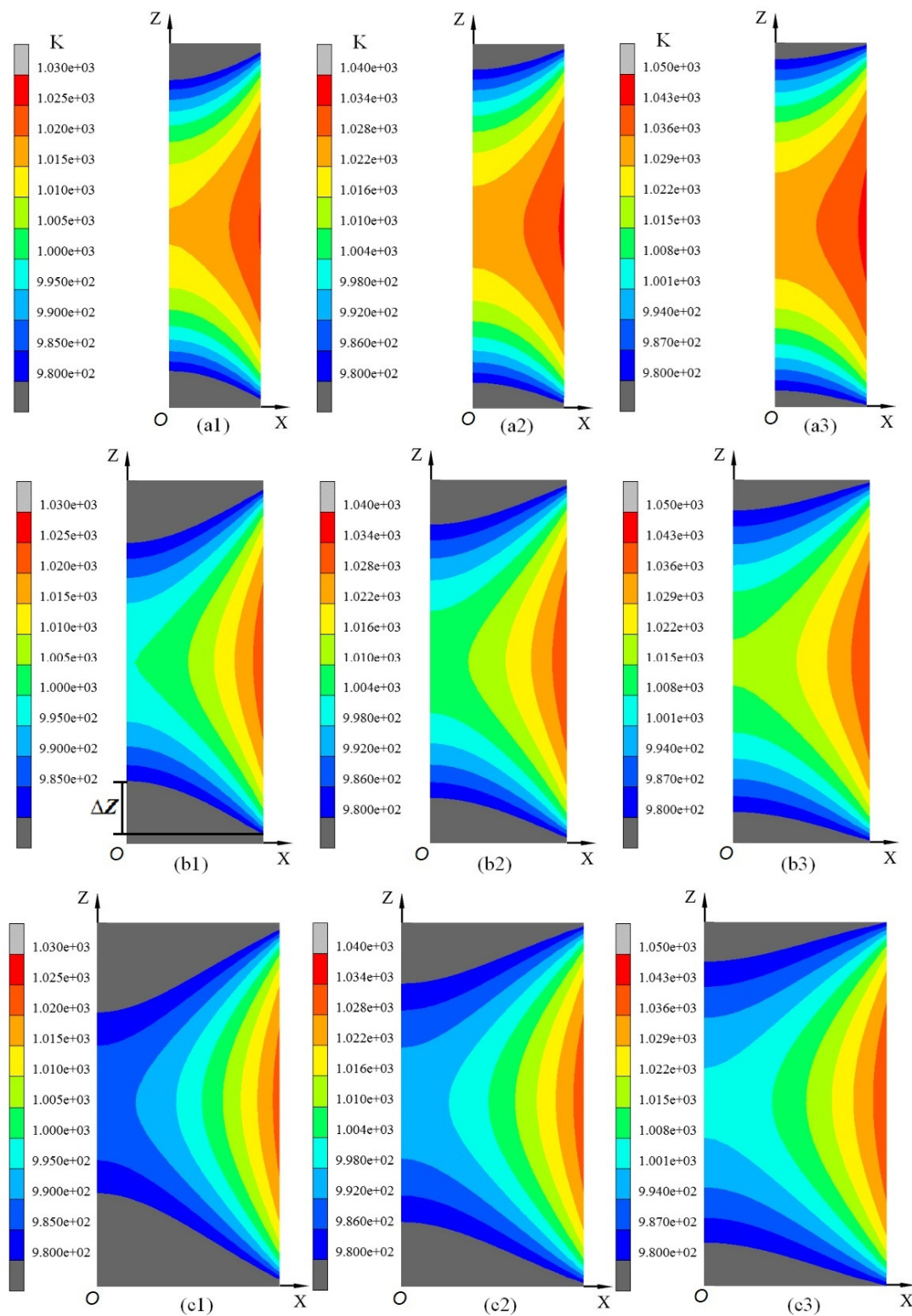


Figure 3. The temperature field when the melting zone was with 0.08–0.12 m away from the bottom of crucible. The unit of the temperature field is K, and the 980 K isotherm line was regarded as the solid–liquid interface. **(a1)** crystal radius 0.01 m, the maximum furnace temperature 1030 K; **(a2)** 0.01 m, 1040 K; **(a3)** 0.01 m, 1050 K; **(b1)** 0.015 m, 1030 K; **(b2)** 0.015 m, 1040 K; **(b3)** 0.015 m, 1050 K; **(c1)** 0.02 m, 1030 K; **(c2)** 0.02 m, 1040 K; **(c3)** 0.02 m, 1050 K.

3.2. Effects of Furnace Temperature and Crystal Radius on Temperature Field

In order to investigate the effects of the furnace temperature and the crystal radius on the temperature field distribution of the crystal, nine numerical samples were studied, as shown in Table 5. When Samples (a1), (a2), (a3) were grouped together, samples (b1), (b2), (b3) together, and samples (c1), (c2), (c3) together, the effect of the maximum furnace temperature on the temperature field could be analyzed at the same radius. While samples (a1), (b1), (c1), (a2), (b2), (c2), and (a3), (b3), (c3) were grouped together, respectively, the effect of the crystal radius on the temperature field at the same furnace temperature could be analyzed. The melting zone was within 0.08–0.12 m away from the bottom of crucible.

Table 5. Relationship between crystal radius and maximum furnace temperature.

Examples	Crystal Radius R (m)	Furnace Maximum Temperature T_{MAX} (K)
(a1)	0.01	1030
(a2)	0.01	1040
(a3)	0.01	1050
(b1)	0.015	1030
(b2)	0.015	1040
(b3)	0.015	1050
(c1)	0.02	1030
(c2)	0.02	1040
(c3)	0.02	1050

The temperature field of the melting zone for every numerical sample is shown in Figure 3. The colored area is the melting part, the gray area below the melting zone is the crystallized GaInSb crystal, and the gray area above the melting zone is the GaInSb polycrystalline rod. The solid–liquid interface below the melting zone is the growth interface and the solid–liquid interface above the melting zone is the melting interface of the polycrystalline rod. It is clear from Figure 3 that the high-temperature region of the melt concentrates on the crucible side wall for each sample. In the sample group of (a1) to (a3), as the furnace temperature increased, the melting zone height along the central axis increased from 0.032 m to 0.0368 m, the convexity of the solid–liquid interface gradually decreased, and both the sample groups of (b1) to (b3) and (c1) to (c3) presented the same conclusion. For the sample group of (a1), (b1), and (c1), the maximum furnace temperature was 1030 K, the height of the melting zone along the central axis decreased from 0.032 m to 0.0208 m as the radius increased, and the convexity of the solid–liquid interface gradually increased. The sample group of (b1), (b2), and (b3) presented the same result, as did the sample group of (c1), (c2), and (c3).

The concept of interface bending is introduced, that is, $\Delta\zeta_r = (Z_C - Z_E)/R_C = \Delta Z/R_C$. Where Z_C is the distance between the center point of the growth solid–liquid interface and the crucible bottom, Z_E is the distance between the edge point of the growth solid–liquid interface and the crucible bottom, and R_C is the radius of the crystal. For the definition of ΔZ , please see Figure 3b1. As can be seen from the above formula, when the $\Delta\zeta_r$ is greater than zero, the solid–liquid interface shape is convex, and when the $\Delta\zeta_r$ is less than zero, the solid–liquid interface shape is concave.

Figure 4 shows the effects of the furnace maximum temperature and the crystal radius on the solid–liquid interface bending (the interface convexity). Firstly, the increase of the crystal radius significantly increases the interface bending of the solid–liquid interface. As the crystal radius increases from 0.01 m to 0.02 m, the interface bending increases by about 50% when the maximum furnace temperature is 1030 K, from 0.16 to 0.24, the interface bending increases by about 67% when the maximum furnace temperature is 1040 K, from 0.12 to 0.20, and the interface bending increases by about 140% when the maximum furnace temperature is 1050 K, from 0.05 to 0.12. Secondly, the increase of the maximum furnace temperature significantly reduces the interface bending of the solid–liquid interface. As the maximum temperature increases from 1030 K to 1050 K, the interface bending decreases from 0.16 to 0.05 when the crystal radius is 0.01 m, from 0.2 to 0.105 when the crystal

radius is 0.015 m, and from 0.24 to 0.12 when the crystal radius is 0.02 m. The higher the maximum temperature, the greater the influence of the crystal radius on the bending of the solid–liquid interface.

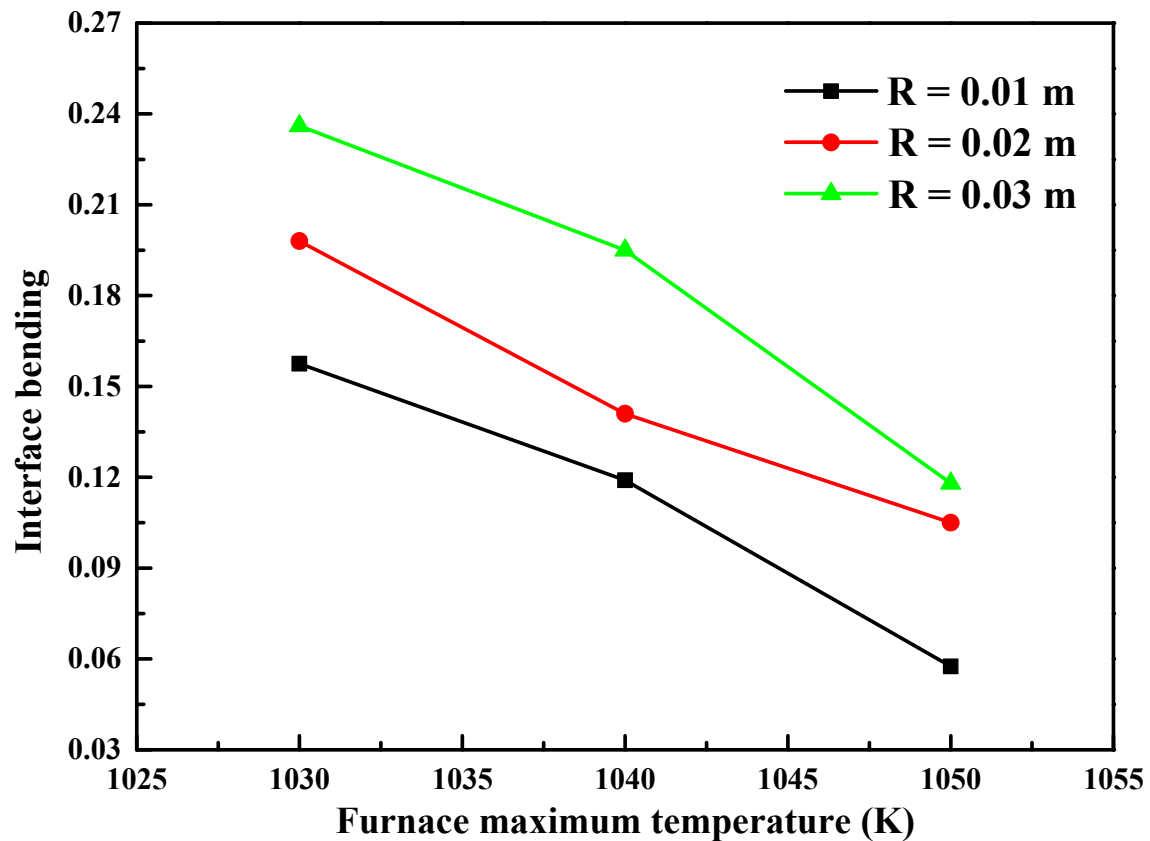


Figure 4. The effects of the furnace maximum temperature and the crystal radius on the growth solid–liquid interface bending when the melting zone is 0.08–0.12 m away from the bottom of the crucible.

Figure 5 shows the temperature gradient distribution curve over the central axis of each crystallized crystal. In the sample (a3), the axial temperature gradient is the highest and the variation of the temperature gradient on the central axis is the largest in the crystallized crystal; the maximum temperature gradient is 5824.77 K/m. In the sample (c1), the axial temperature gradient is the lowest and the variation of the temperature gradient on the central axis is the lowest in the crystallized crystal; the maximum temperature gradient is 2301.84 K/m. Comparing curves a1–a3, b1–b3, and c1–c3, we can find that when the radius is the same, the maximum temperature gradient (the value corresponding to the rightmost point of all curves) increases by about 20% with the maximum temperature increasing from 1030 K to 1050 K via 1040 K. Comparing curves a1–c1, a2–c2, and a3–c3, we can find that when the maximum temperature is the same, the maximum temperature gradient decreases by about 50% with the crystal radius increasing from 0.01 m to 0.02 m.

In summary, the increase of the furnace temperature can greatly reduce the convexity of the solid–liquid interface, but can greatly increase the thermal stress in crystallized crystals. The former could be helpful to decrease component segregation, while the latter would result in a dislocation increase.

3.3. Effect of Furnace Temperature and Crystal Radius on Melt Flow Field

According to the numerical samples shown in Table 5, the flow fields in the melting zone are calculated and simulated as shown in Figure 6. We can see that the velocity of the fluid close to the crucible inner wall is larger, and the maximum velocity appears in the middle of the melt zone, while the fluid velocity in both the upper and the lower parts of

the melt zone is smaller. When the crucible radius is determined, the maximum velocity of the fluid does not change significantly with the increase of the crucible wall temperature. When the temperature of the crucible wall is determined, the maximum velocity of the fluid increases obviously with the increase of the crucible radius.

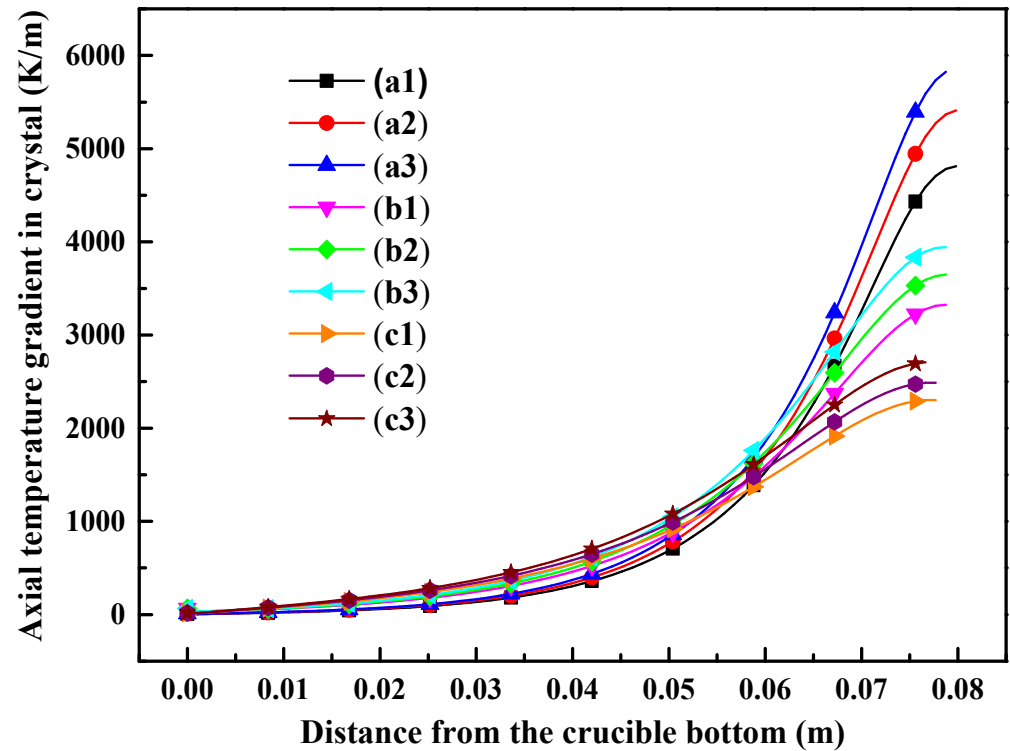


Figure 5. The axial temperature gradient in the crystal when the melting zone is 0.08–0.12 m from the bottom of the crucible. (a1) crystal radius 0.01 m, the maximum furnace temperature 1030 K; (a2) 0.01 m, 1040 K; (a3) 0.01 m, 1050 K; (b1) 0.015 m, 1030 K; (b2) 0.015 m, 1040 K; (b3) 0.015 m, 1050 K; (c1) 0.02 m, 1030 K; (c2) 0.02 m, 1040 K; (c3) 0.02 m, 1050 K.

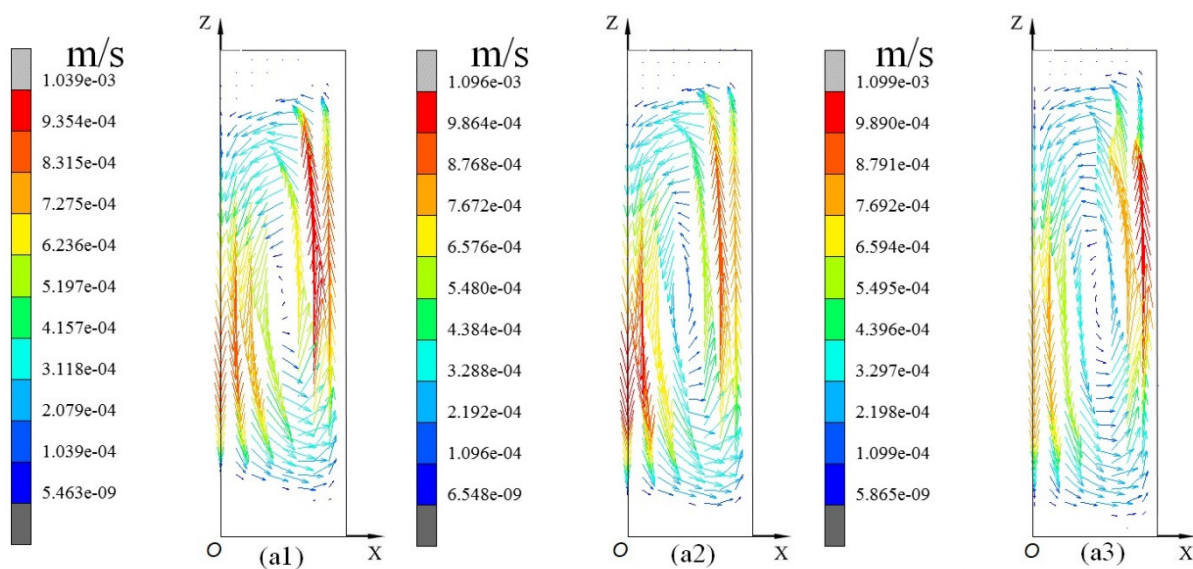


Figure 6. Cont.

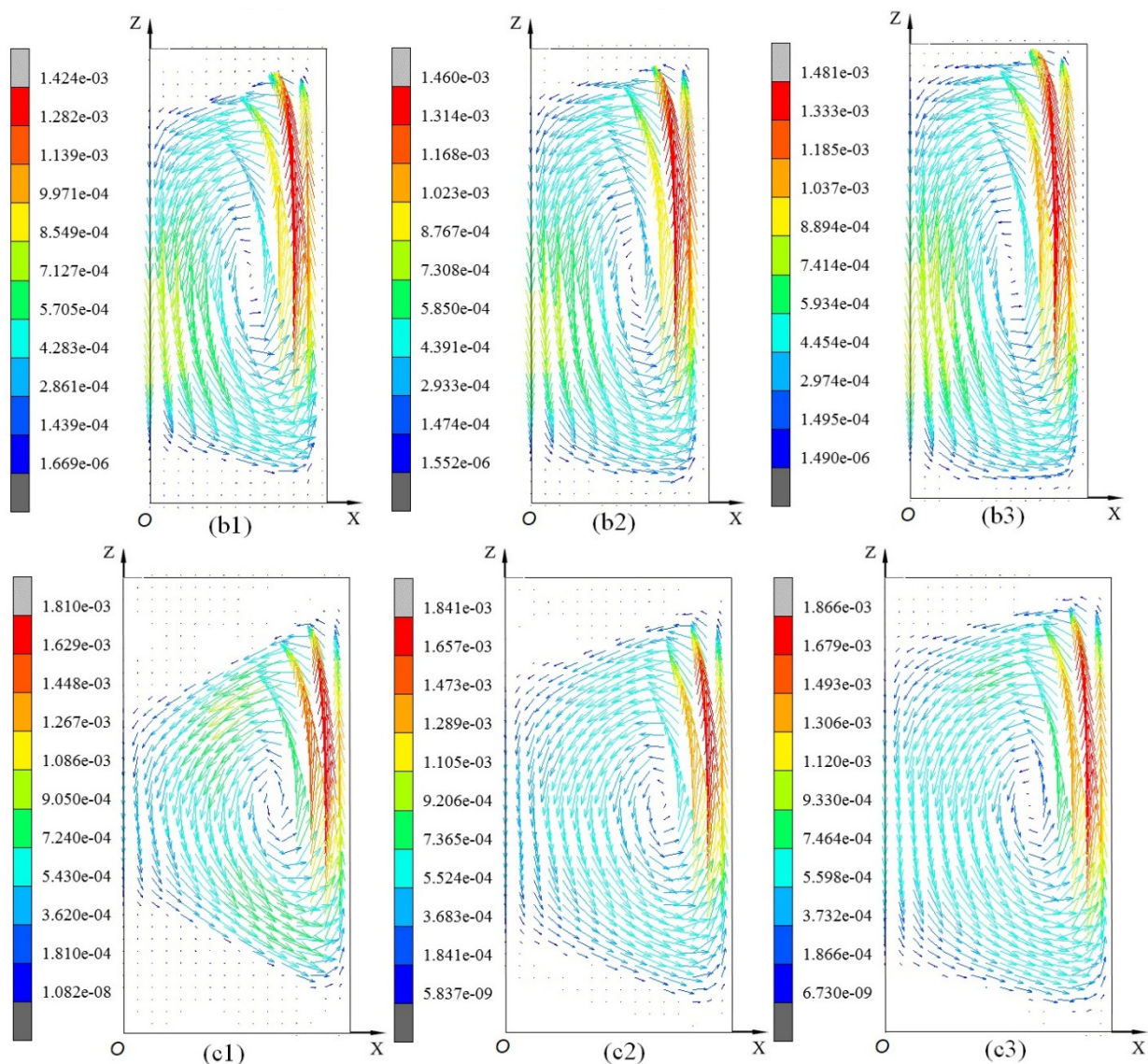


Figure 6. The vector graphs of fluid flow when the melting zone is 0.08–0.12 m from the bottom of the crucible. (a1) crystal radius 0.01 m, the maximum furnace temperature 1030 K; (a2) 0.01 m, 1040 K; (a3) 0.01 m, 1050 K; (b1) 0.015 m, 1030 K; (b2) 0.015 m, 1040 K; (b3) 0.015 m, 1050 K; (c1) 0.02 m, 1030 K; (c2) 0.02 m, 1040 K; (c3) 0.02 m, 1050 K.

It can also be seen from Figure 6 that the flow in the melt zone is counterclockwise. Since the numerical domain selected in the mode is the right half of the axisymmetric two-dimensional model, the flow in the left half of the melt zone should be clockwise. Because the fluid has the property of thermal expansion and contraction, its density decreases after heating. When the temperature of the crucible wall is higher than that of the melt, the fluid near the crucible wall will be heated, and its density will decrease and float up. The fluid far from the crucible wall has a lower temperature, so it has a higher density and sinks down, forming a convective cell in the melting zone.

The radial velocity distribution and the maximum velocity distribution of melt flow are shown in Figure 7. It can be seen from Figure 7a that for a radius of 0.01 m, see curves (a1), (a2) and (a3), the difference between the velocity of the melt in the center of the melt zone and that near the crucible wall is small; for a radius of 0.015 m, see curves (b1), (b2), and (b3), the latter is about twice as large as the former. For a radius of 0.02 m, see curves (c1), (c2), and (c3), the latter is about three times as large as the former. That is, the greater the radius, the greater the difference. However, the maximum furnace temperature has

only a small effect on this difference. As the crucible radius increases, the temperature difference between the center of the melting zone and the crucible wall increases, and so do both the convection intensity in the melting zone and the difference between the velocity of the fluid in the crucible center and that of the fluid near the crucible wall. It can be seen more intuitively from Figure 7b that the maximum velocity of melt increases slightly with the furnace maximum temperature for the same radius: about 6% for a 0.01-m radius, about 4% for 0.015 m, and about 3% for 0.02 m. However, it increases significantly with the increase of the radius from 0.01 m to 0.02 m, about 74%, 68%, and 70% for the furnace maximum temperatures 1030 K, 1040 K, and 1050 K, respectively.

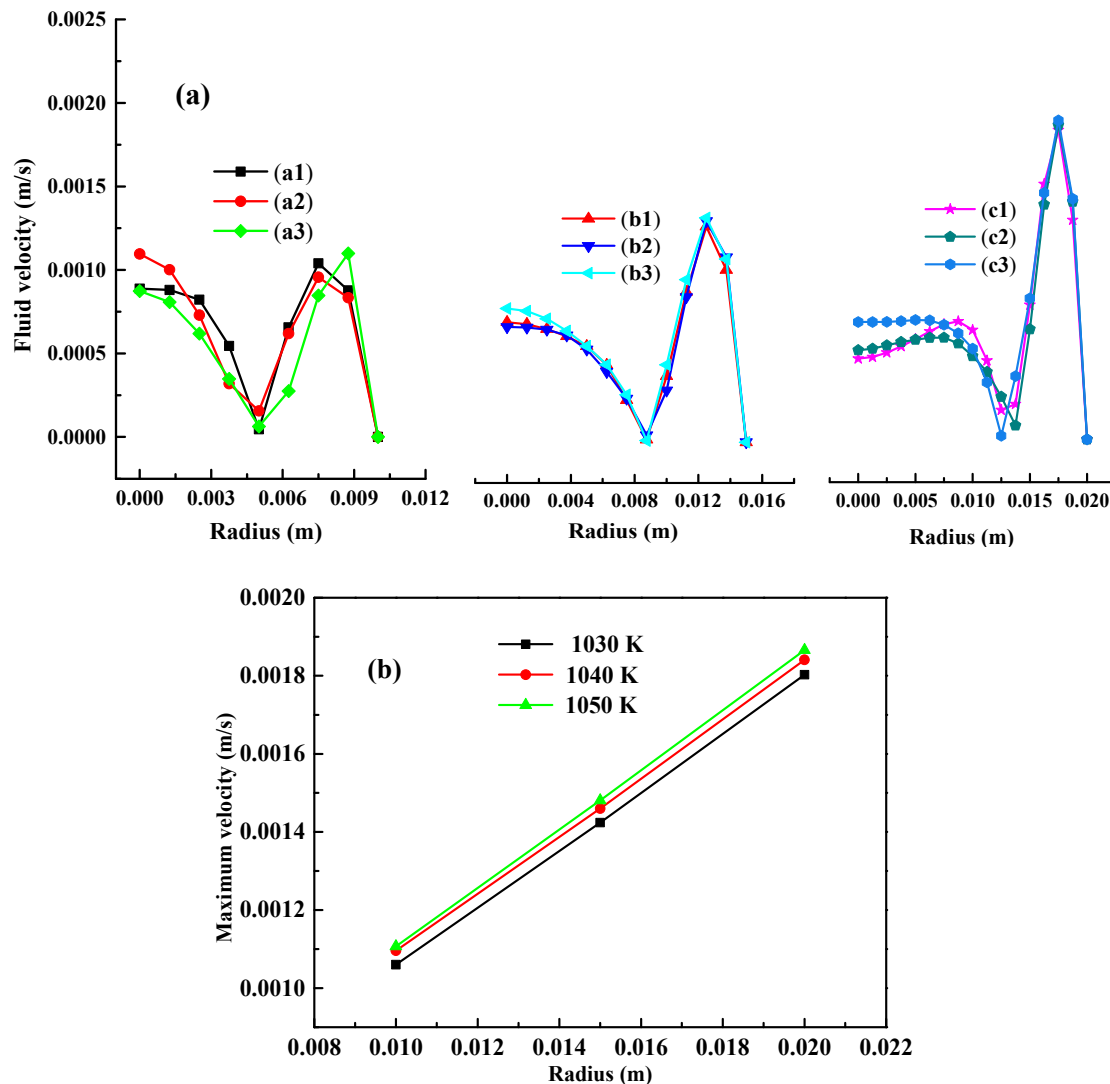


Figure 7. (a) The distribution of flow velocity along the radius of the crucible and (b) the maximum flow velocity variations with the crystal radius and the maximum temperature of the furnace. The melting zone is 0.08–0.12 m away from the crucible bottom.

4. Conclusions

The heat transfer and liquid phase convection during GaInSb crystal growth via the traveling heater method with a seed crystal were investigated using numerical simulation. The results show that:

- (1) At the same furnace temperature profile, there will be a phenomenon of “thermal impermeability” with an increase in the crystal radius. As the crystal radius increases from 0.01 m to 0.03 m, the maximum furnace temperature should increase from 1030 K to 1060 K at least in order to ensure the successful introduction of the seed.

- (2) The increase of the crystal radius significantly increases the interface bending of the solid–liquid interface, which increases by about 50%, 67%, and 140% as the crystal radius increases from 0.01 m to 0.015 to 0.02 m, corresponding to the maximum furnace temperature 1030 K, 1040 K, and 1050 K, respectively. Contrastingly, the increase of the maximum furnace temperature significantly reduces the interface bending of the solid–liquid interface, which decreases as the maximum temperature increases from 1030 K to 1050 K, from 0.16 to 0.05, from 0.2 to 0.105, and from 0.24 to 0.12, corresponding to the crystal radius 0.01 m, 0.015 m, and 0.02 m, respectively. The maximum temperature gradient increases by about 20% with, for the same radius, the maximum temperature increase from 1030 K to 1050 K, while it decreases by about 50% for the same maximum furnace temperature with a crystal radius increase from 0.01 m to 0.02 m.
- (3) The maximum velocity in the melting zone increases slightly with the furnace maximum temperature for the same radius, about 6% for a 0.01-m radius, about 4% for a 0.015-m radius, and about 3% for a 0.02-m radius. However, it increases significantly with the increase of the radius from 0.01 m to 0.02 m, about 74%, 68%, and 70% for the furnace maximum temperatures 1030 K, 1040 K, and 1050 K, respectively.

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