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P20

Growth interface shape control of InGaSb crystals in the Vertical Gradient Freeze method under microgravity conditions and optimization using machine learning methods

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1. Introduction

Semiconductors materials represent the foundation of present-day electronic devices. Their unique conducting properties between insulators and metals have unveiled many practical applications. For instance, InGaSb is an III-V ternary alloy characterized by tunable properties and can be used in the field of energy conversion, as an efficient material for thermoelectric (TE) and thermophotovoltaic (TPV) applications. These approaches provide rigid solutions for global warming and clean energy crisis. However, the growth of such homogeneous InGaSb alloy bulk crystal with uniform compositions on earth is difficult and affected by natural convection. This undesirable effect can be minimized under microgravity, which is an appropriate environment for investigating the growth kinetics and gain deeper insight into the transport phenomena, hence, valuable crystal growth space experiments have been carried out for that purpose. In addition to microgravity growth, it is known that, applying rotation is an effective way to create optimal crystal growth conditions and obtain a convex interface toward the melt, which is essential for preventing polycrystalline growth.

Our group has built a numerical analysis based on the experiments performed at the International Space Station (ISS) by Inatomi et al.[1)], where InGaSb alloy crystals were grown using the vertical temperature gradient method on a GaSb/InSb/GaSb sandwich-structured sample as illustrated in **Fig. 1**. In this method, the sample is subjected to a fixed temperature gradient in the axial direction and the system is heated over the melting point of InSb which melts first. GaSb material then dissolves in the molten InSb (dissolution process) resulting in a mixture of InGaSb (growth solution) that becomes supersaturated under the applied temperature profile. GaSb is fed from the higher temperature feed crystal into the solution and InGaSb crystal grows on the surface of the lower temperature seed crystal (growth process).

In our previous research work [2)], we established a relationship between the diffusion coefficient of GaSb in InSb melt and its concentration based on the microgravity experimental results. The motivation behind it is that, onboard the ISS, the gravity-driven convection in the melt is minimized and diffusion-controlled heat and mass transport conditions are achieved, thus, it is anticipated that the crystal growth rate is strongly affected by the diffusivity of GaSb and its concentration, which has been supported by the numerical results. In the present study, the effects of rotation and temperature gradient change on the interface shapes and concentration distribution are discussed. Simulation results show that, such methods are effective for controlling the growth interface and promote a less concave shape. Bayesian optimization was also used in order to efficiently determine the associated control parameters of InGaSb crystals growth process.

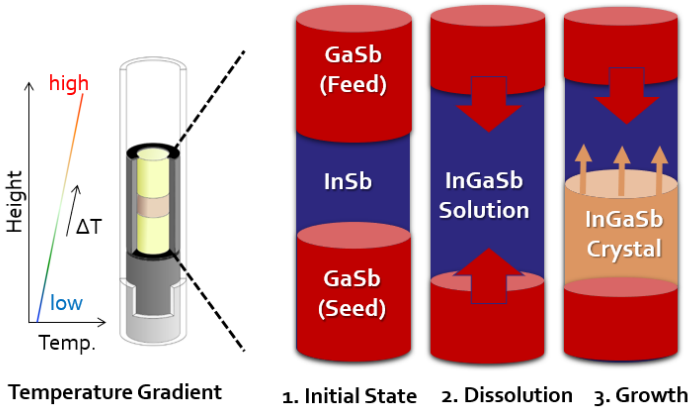

Fig. 1 Schematic diagram of the vertical gradient freezing method [1].

2. Numerical Analysis

2.1 Simulation model

A schematic description of the InGaSb crystal dissolution/growth system used is shown in **Fig. 2**. The GaSb(feed)/InSb Te-doped /GaSb(seed) sandwich sample was stacked in a quartz ampoule and sealed with Boron Nitride (BN) and carbon sheet as seen in **Fig. 2(a)**. The system was subjected to one vertical temperature gradient (**Fig. 2(b)**) with the top being hotter and the bottom being cooler. The whole system was heated at a heating rate of 0.01K/s up to the target temperature and then kept constant. The grid system used for the simulation is also shown in **Fig. 2(c)**.

2.2 Governing equations

In the present numerical simulation model, the following assumptions were made:

(1) Liquid phase is an incompressible Newtonian fluid;

(2) Densities of the solid and liquid phases are constant, thus the associated volume changes (shrinkage and expansion) during phase changes are negligible;

(3) Changes in physical properties (because of compositional variations) during the dissolution and growth processes are not significant,

Under these assumptions, the governing equations of the crystal phase, namely continuity, momentum conservation, energy conservation and mass transport equations are given as:

$$\nabla . \boldsymbol{v} = 0 \tag{1}$$

$$\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v} . \nabla)\boldsymbol{v} = -\frac{1}{\rho}\nabla p + \nu\nabla^2 \boldsymbol{v} + \boldsymbol{g}(\beta_T \Delta T + \beta_C \Delta C) + \frac{\boldsymbol{F_r}}{\rho} \tag{2}$$

$$\frac{\partial T}{\partial t} + (\boldsymbol{v} . \nabla)T = \alpha\nabla^2 T \tag{3}$$

$$\frac{\partial C}{\partial t} + (\boldsymbol{v} . \nabla)C = \nabla^2 (D \cdot C) \tag{4}$$

Where $\boldsymbol{v}$ is the velocity, ρ density, p pressure, ν kinematic viscosity, $\boldsymbol{g}$ gravitational acceleration, β_T thermal expansion coefficient, β_C solutal expansion coefficient, F_r centrifugal force, T temperature, α thermal diffusivity, C solute concentration and D is the diffusion coefficient of GaSb in InSb melt defined as follows [2]:

$$D = 2.7 \times 10^{-8} \times e^{(-3 \times C)} \tag{5}$$

In the solid phases (BN and Quartz), only the energy balance is considered:

$$\frac{\partial T}{\partial t} = \alpha_i \nabla^2 T \qquad \text{(i subscript standing for BN and Quartz)} \tag{6}$$

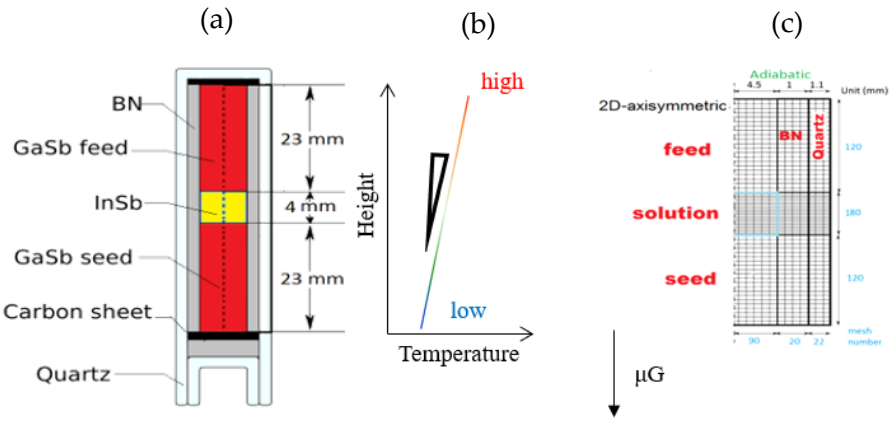

Fig. 2 Schematics of the growth ampoule (a), the applied temperature profile (b) and the grid system of the simulation (c) [3].

2.3 Boundary conditions and discretization

The boundary conditions for the top and bottom walls are adiabatic for the temperature field, no-slip conditions for the velocity field, and no flux condition (in the normal direction to the ampoule walls) for the concentration field. In addition, on the boundaries between the feed and seed crystals, the walls and the seals (BN and Quartz) are assumed no-slip condition for the velocity field. The same along the normal direction to the wall and seal boundaries; no mass and heat fluxes are allowed. On the outside of the ampoule along the outer walls, the temperature profile varies according to the selected temperature gradients.

The simulation was performed under the microgravity level of 10^{-4} G that corresponds to a typical average gravity level observed on the ISS. The g-jitter effect on the ISS was not considered and the gravity direction was aligned with the axis of the sandwich system and directed towards the seed crystal. As for the initial conditions, the complete molten state of InSb was regarded as the initial state and therefore the initial GaSb concentration in the solution was taken zero.

The governing equations together with the boundary conditions were discretized by the finite volume method and the pressure-velocity coupling was handled by the Pressure Implicit with Splitting of Operators (PISO) algorithm and then solved using the open-source CFD code: OpenFOAM and the new volume-average continuum model. This model utilizes volume fractions of the solid and liquid phases determined by using the applied temperature and concentration profiles, as well as the phase diagram of the binary system GaSb-InSb [4].

3. Results and discussion

In this numerical study, the interface shape is investigated from the calculation results of the concentration distribution as shown in **Fig. 3**. In order to quantitively examine the growth interface shape change, the deformation degree (ΔY_d) is introduced and defined as the difference between the growth interface positions at the periphery (Y_p) and at the center axis (Y_c):

$$\Delta Y_d = Y_p - Y_c \tag{7}$$

Hence, a large positive value indicates an undesirable highly concave interface shape, a value closer to zero indicates a flat one and a negative value is an indicator for a favorable convex growth interface shape. Accordingly, we aim to minimize the deformation degree by means of rotation and the use of different thermal boundary conditions. The temperature gradient $\frac{\Delta T}{\Delta Y}$ and the rotation speed ω represent then the control parameters to be determined for controlling the growth interface shape of InGaSb crystals.

As attributing random combinations of these parameters is implausible due to the significant number of numerical simulations that have to be performed, we employ machine learning along with Bayesian optimization algorithms to ensure computational time and cost efficiency. After multiple iterations, the optimal growth conditions for a more desirable interface shape were obtained as follows:

$$\left(\frac{\Delta T}{\Delta Y}\right)_1 = 0.23 \text{ K/mm} \qquad \omega = 60 \text{ rpm}$$
$$\left(\frac{\Delta T}{\Delta Y}\right)_2 = 0.81 \text{ K/mm}$$

Initial numerical calculations suggested that, subjecting the system to two different temperature gradients (with a smaller ΔT near the feed crystal) promotes a less concave growth interface shape, thus, $\left(\frac{\Delta T}{\Delta Y}\right)_1$ and $\left(\frac{\Delta T}{\Delta Y}\right)_2$ were selected as

the temperature gradients for the feed ($Y \geq 25$ mm) and seed ($Y < 25$ mm) regions, respectively.

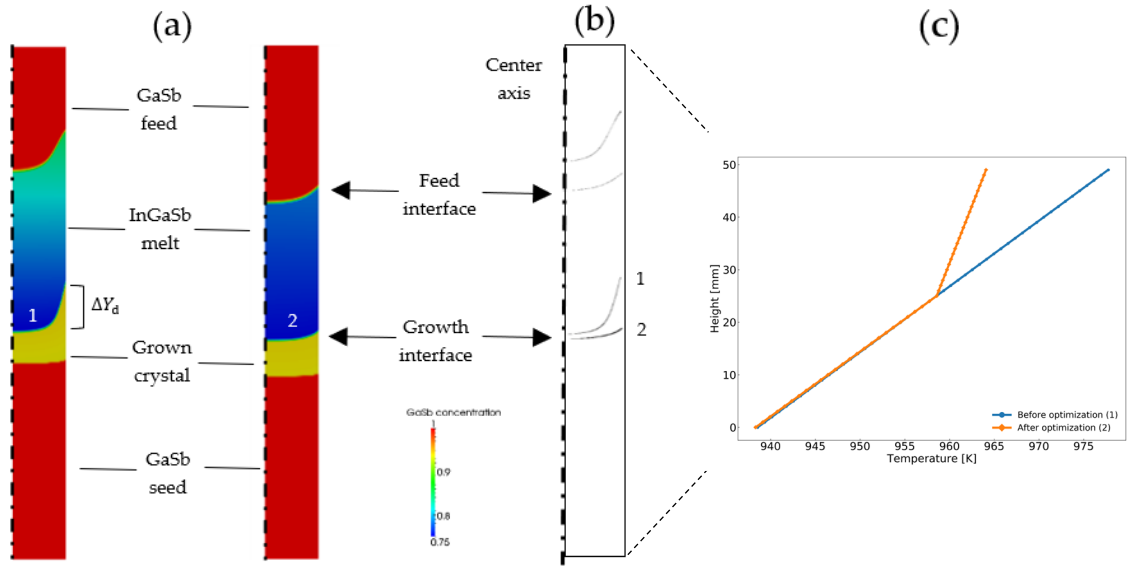

Fig. 3 GaSb concentration distribution (a), interface shapes (b) and temperature gradients (c) of the optimized and non-optimized cases.

Figure 3 illustrates the effects of temperature gradient change and low constant rotation on the concentration distribution (**Fig. 3(a)**) and interface shapes (**Fig. 3(b)**). Initially, both of the growth and feed interfaces are highly concave. After optimization, a flatter growth and feed interfaces shape are obtained and the GaSb concentration in melt is overall more uniform compared to the initial case. By using a smaller temperature gradient near the feed region, as shown in **Fig. 3(c)**, there is a diminishment in the feed dissolution rate, therefore a decrease in the solute concentration in the melt. In addition, under the effect of the centrifugal force, due to its lower melting point, InSb, which represents the heaviest compound in the InGaSb solution, is pushed towards the wall side where it remains liquid at relatively low temperatures, promoting hence a desirable flat interface shape throughout the growth process.

Figure 4 shows the time variation of the deformation degree ΔY_d of the growth interface (melt/InGaSb crystal interface) before and after optimization. From the figure, the degree of deformation of the non-optimized case is seen to rapidly increase with time and reach a large value of about 5.4 mm after 47.5 h of growth time. After optimization, the degree of deformation is significantly reduced and does not exceed the value of 0.8 mm throughout the whole crystal growth process.

From the results above, it is deduced that, subjecting the system to two different temperature gradients (with a smaller temperature gradient near the feed crystal) and a low rotation rate is sufficient to change the solute concentration distribution, reduce the growth interface deformation degree and promote transition to a convex shape.

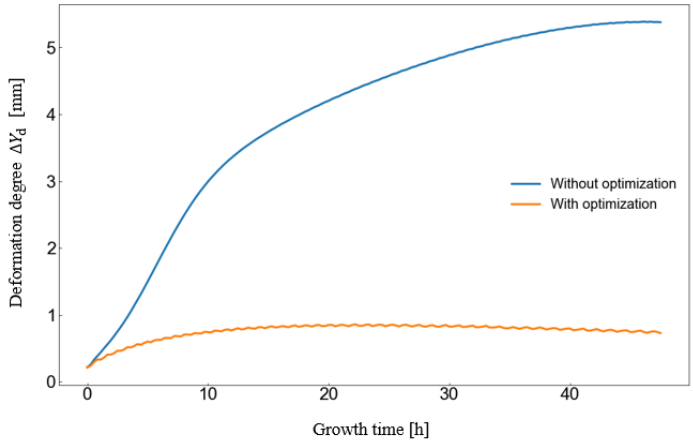

Fig. 4 Comparison between the deformation degree over time of the crystal growth interface before and after optimization.

4. Conclusion

In order to investigate the conditions to grow semiconductor crystal with a flatter solid-liquid interface, a numerical simulation study was carried out for controlling the growth interface shape of InGaSb crystals in the Vertical Gradient Freeze method under microgravity conditions. The simulation results show that, by applying rotation and different thermal conditions, a flatter growth interface can be maintained throughout the growth process. The optimum growth conditions for the desired interface shape were obtained utilizing machine learning methods such as Bayesian optimization to resolve the computational demand issue.

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