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Estimation of the Diffusion Coefficient of GaSb into InGaSb Melt using Bayesian Optimization Method and the ISS Experimental Results

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1. Introduction

Semiconductor materials have always been playing a crucial role in the development of novel electronic devices. The unique conducting properties of semiconductors between metals and insulators have unveiled many practical applications. Among these materials, III-V compounds have a significant feature of their direct bandgaps which can be efficiently used in photonic and optoelectronic devices. InGaSb, an III-V ternary alloy, belongs to this category and can also 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.

InGaSb has the advantage of tunable properties between its binary compounds InSb and GaSb. However, because of the differences in its constituents' properties, growing high-quality InGaSb crystals is a challenging task, the process is significantly affected by various parameters, primarily, natural convection. As gravity represents the driving force for unsteady convective flows in the crystal growth from the melt, valuable space experiments have been carried out for comprehending the underlying physical phenomena in a convection free environment. But due to the cost and time needed, the number of microgravity experiments that can be performed is extremely limited, hence the importance of adopting a numerical approach.

In our study, we build a numerical analysis based on the experiments performed at the International Space Station (ISS)[1]. InGasb alloy crystal was grown using the vertical temperature gradient method on a GaSb/Te-doped InSb/GaSb sandwich sample as illustrated in **Fig. 1**. During the process, InSb melts first. Then GaSb material (solid) dissolves into the molten InSb (dissolution process) resulting in a mixture of InGaSb (growth solution) which becomes supersaturated under the applied temperature profile and starts accumulating on the GaSb seed crystal at the bottom. Consequently, single crystals of InGaSb alloy begin to grow on the seed (the growth process). The results of previous numerical simulations diverged from the experimental ones by predicting larger growth rates and excessive feed or seed dissolution [2]. It is suggested that diffusion is dominant during the growth process under microgravity and the diffusion coefficient needs to be accurately defined. Therefore, we aim to predict its value using machine learning and Bayesian optimization methods.

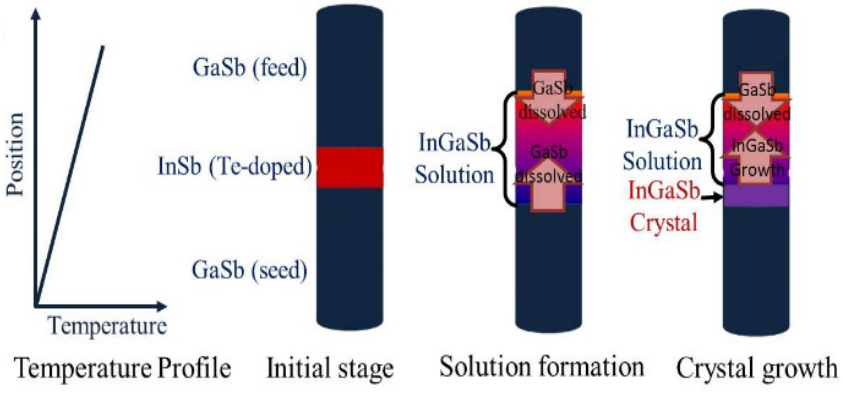

Fig. 1 Schematic diagram of the vertical gradient freezing method [1].

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/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 three vertical temperature gradients (**Fig. 2(b)**) with the top being hotter and the bottom being cooler. The whole system was heated at a heating rate of 0.001K/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:

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

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

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

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

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, T temperature, α thermal diffusivity, and C and D are, respectively, the concentration and diffusion coefficient of GaSb.

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

$$\frac{\partial T}{\partial t} = \alpha_i \nabla^2 T \qquad (i \text{ subscript standing for BN and Quartz}) \tag{5}$$

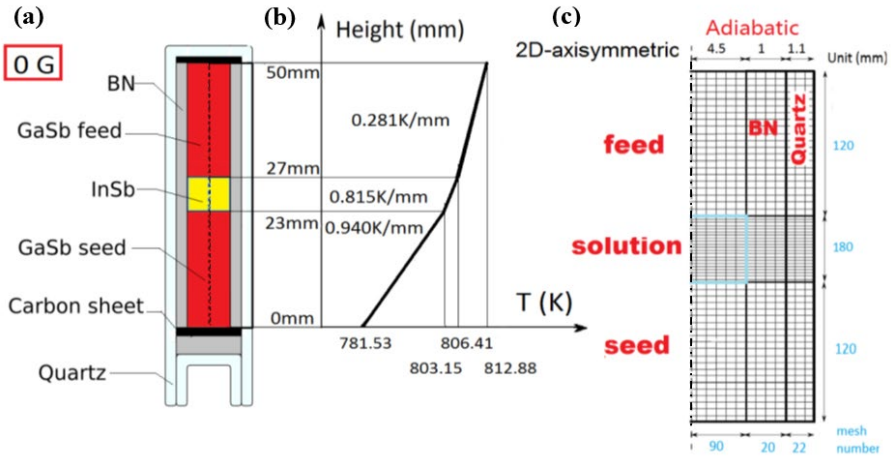

Fig. 2 Schematics of the growth ampoule (a), the applied temperature profile (b) and the grid system of the simulation (c) [2].

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 has been determined from the measured values from the ISS experiment [1].

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 (gravity fluctuation) on the ISS was not considered. The gravity direction was aligned with the axis of the sandwich system and was directed towards the seed crystal. As for the initial conditions, the complete molten state of InSb was regarded as the initial state in the simulation 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 [3].

3. Results

3.1 Initial estimation: constant diffusion coefficient

Figure 3(a) illustrates the growth rate variation along the growth direction for the experimental case (black dots) and initial numerical calculations (blue curve). As it can be seen, the calculated growth rate was found to be much higher than that of the experiments. This latter was determined by measuring the grown length between the striations and reached a maximum of about 0.15 mm/h. While the predicted growth rate surpasses this value and attains 0.6 mm/h.

As the convection effect was suppressed under microgravity, only diffusion phenomenon is present and dominant in GaSb transport. It is anticipated that the crystal growth rate is strongly affected by the diffusion coefficient of GaSb in the InGaSb melt (proportional to it). Initially, the value used is taken from the literature as $D = 1.20 \times 10^{-8}\, m^2/s$ [4]. it is assumed to be too large, hence, we optimized this value and found that, using a diffusion coefficient of $D = 2.20 \times 10^{-9}\, m^2/s$ reproduced considerably better results. Overall, the calculated growth rate has the same tendency as the experimental one. It reaches a value of about 0.14 mm/h at 6mm of crystal grown length and 0.124 mm/h at the end of growth, with the final crystal length being 13 mm.

Furthermore, we notice that for both the experiment and simulation, the growth rate at the initial stage of growth is relatively low and then gradually increases. This could be explained by the melt low saturation during the early stages of growth as shown in **Fig. 3(b)**, where the saturation ratio along the melt is plotted in 4 different stages of growth: beginning of growth (i), middle stages (ii), (iii) corresponding to a crystal length of 2mm and 7mm after a growth time of 16h and 50h respectively, final stage (iv), that is after 105h of growth.

It is deduced that the amount of GaSb supplied to the melt was not yet sufficient during the first stages of growth (steps i to ii), but then the melt progressively becomes saturated due to the feed crystal dissolution and the growth rate increases reaching the steady-state growth (steps ii to iv). Consequently, we proceed to investigate the effect of solute concentration on the diffusion coefficient and the crystal growth process.

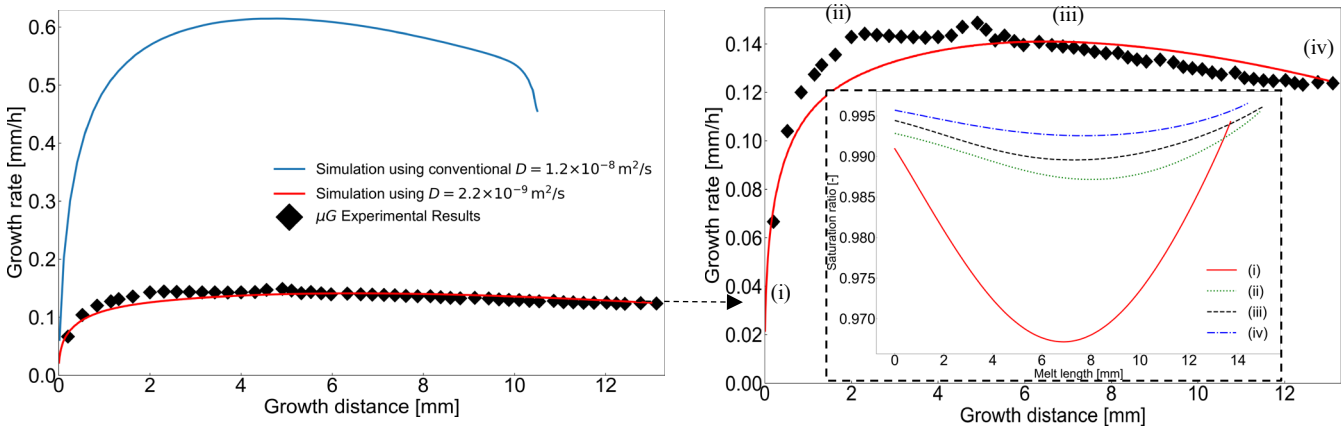

Fig. 3 Comparison between crystal growth rates of the experiments and simulations (a) and melt saturation ratio during the growth process (b).

3.2 Optimization: concentration dependent diffusion coefficient

For this part of the research, after initial trials, we have adopted the following equation as a model equation to express the concentration dependence of the diffusion coefficient: $D = D_0 \times e^{(-b \times C)}$

Where C is GaSb concentration in the melt, and D_0 and b represent the parameters to be determined.

As attributing random combinations of these two 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 diffusion coefficient is optimized as follows:

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

Figure 4 compares between the calculated crystal growth rates using the new equation and the experimental ones for different GaSb crystal orientations (111A, 111B and 110). It is clear that a concentration dependent diffusion coefficient predicted more accurate results, especially during the early stages of the growth process when the saturation ratio of the melt was relatively low. A similar order to the space experiments was also followed (Case111B > Case110 > Case111A).

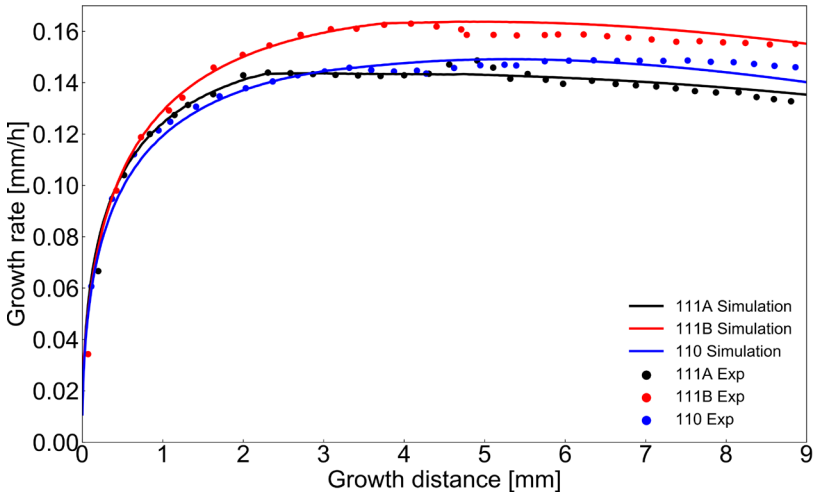

Fig. 4 Growth rates comparison between experimental and simulation results using newly estimated *D* for different crystal orientations.

4. Conclusion

In this study, a relationship between the diffusion coefficient of GaSb into InGaSb melt and its concentration was established based on the ISS experimental results and Bayesian optimization methods. It is concluded that the growth rate, feed/seed dissolutions lengths and grown crystal length are strongly affected by the diffusion coefficient and the GaSb concentration in the melt. The modified numerical model could be implemented in further calculations to identify optimization possibilities using various methods, such as applying to the system external fields, magnetic field, vibration or rotation so as to reach uniform crystal growth and high growth rate.

Acknowledgement

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