Predictions of zinc selenide single crystal growth rate for the microgravity experiments

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Abstract One predicts the crystal growth rate of ZnSe with a low vapor pressure system in a horizontal configuration based on one dimensional advection-diffusion and two-dimensional diffusion-convection model. The present results show that for the ratios of partial pressures, $s = 0.2$ and 2.9, the growth rate increases with the temperature differences between the source and crystal. As the ratio of partial pressure approaches the stoichiometric value, $s = 2$ from $s = 1.5$ (zinc-deficient case: $s < 2$) and 2.9 (zinc-rich case: $s > 2$), the rate increases sharply. For the ranges from 1.5 to 1.999 (zinc-deficient case: $s < 2$) and from $s = 9$ to 2.9 (zinc-rich case: $s > 2$), the rate are slightly varied. From the viewpoint of the order of magnitude, the one-dimensional model for low vapor pressure system falls within the 2D predictions, which indicates the flow fields would be advective-diffusive. For the effects of gravitational accelerations on the rate, the gravitational constants are varied from 1 g to 10^{-6} g for $\Delta T = 50$ K and $s = 1.5$, the rates remain nearly constant, i.e., 211 mg/hr, which indicates Stefan flow is dominant over convection.

Key words Zinc selenide, Microgravity

1. Introduction

The wide-bandgap II-VI compounds hold promise for blue-light emitting diodes and laser diodes. Zinc selenide (ZnSe) with a bandgap of 2.72 eV is one of the most promising material for laser cathode-ray tube emitting in the blue range and other e-beam pumped semiconductors, so there is growing in much interest and a consistant demand for a photorefractive material for optical data processing [1, 2]. Although polycrystalline ZnSe will continue to be adequate for many applications, single crystals of high quality will be required for use in optoelectronic devices. Laser diodes have been fabricated on GaAs substrates since it is difficult to obtain ZnSe single crystals of high quality for substrates. However, in the epitaxial system ZnSe-GaAs, significant biaxial strain is introduced because of the lattice mismatch and different thermal expansion coefficients. To overcome these difficulties homoepitaxy should be applied with ZnSe substrates [3]. Recent progress in II-VI device technology with the fabrication of blue-green ZnSe-based laser diodes has been based on successful electrical control of the epitaxial layers [4, 5].

ZnSe single crystals have been grown from the melt and from solution [6-10], as well as from the vapor phase by either chemical vapor transport (CVT) [11-13] or physical vapor transport (PVT) [14-19]. In this study the crystal growth method used was PVT in sealed quartz ampoules due to its high melting point (1568 K). PVT has the advantage of requiring lower temperatures than growth from the melt and it thus makes the process more amenable in instances as well as less vulnerable to contamination and the severe strains resulting from growth at very high temperatures. Lower temperature growth also reduces the potential for altering the stoichiometry of the material. Other benefits stem from the inherent purification mechanism in the process due to differences in the vapor pressures of the native elements and impurities, and the enhanced solid-vapor interfacial morphological stability during the growth process, which has been described by Rosenberger [20]. Further, the implementation of physical vapor transport (PVT) growth in closed ampoules affords experimental simplicity with minimal needs for complex process control which makes it an ideal candidate for space investigations in systems where gravity tends to have undesirable effects on the growth process. PVT is less complex than growth by chemical vapor transport (CVT) and it avoids the problem of contamination of the growing crystal by the transport agent.

It has been shown [21-23] that vapor-phase composition during physical vapor transport (PVT) growth of II-VI semiconductors has a significant effect on mass transport and hence allows a control of the growth rate.

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Our interest is to investigate the effects of advective-diffusion and diffusive-convection on the ZnSe crystal growth processes during physical vapor transport (PVT) based on dissociative sublimation-condensation for a parametric range corresponding to actual experimental conditions. In other words, in the PVT system of ZnSe, the molecular species ZnSe is dissociatively sublimed into the gas of the $Zn(g)$ and $Se_2(g)$ in the vapor phase from the crystalline system phase $ZnSe(s)$, and is subsequently transported and re-incorporated into the single crystalline phase (ZnSe) [24]. The influence of a residual gas is included in the two-dimensional model. The simulations show that the Stefan flux dominates the system and subtle gravitational effects can be gauged by subtracting this flux from the calculated flow fields. Up to now the previous works are concerned with the effects of convection on congruent transport. In recent years Zhou et al. [25] and Ramachandran et al. [26] addressed the effect of thermal-solutal convection non-congruent transport by dissociative sublimation-condensation. Zhou et al. [25] reported that the traditional approach of calculating the mass flux assuming one-dimensional flow for low vapor pressure systems such as ZnTe is indeed correct.

The main purposes of this paper are to discuss the crystal growth rate predicted by one and two-dimensional mass transfer-limited model with idealized boundary conditions and to perform the generic research as ground-based (1 g) studies for future microgravity experiments. Also, we believe the results obtained here will be helpful for proper design of experimental conditions during the PVT of ZnSe by dissociative sublimation.

2. Modeling and Simulations

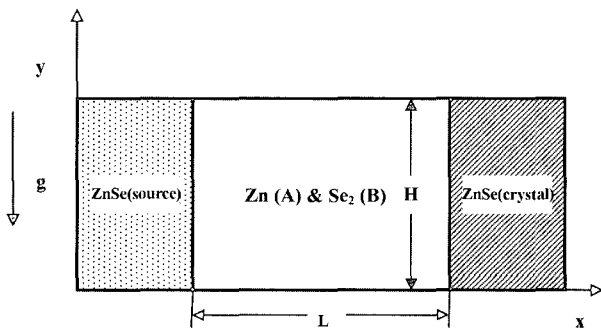One considers our study for the effects of advective-

Fig. 1. Schematic representation of the PVT growth reactor in a two-dimensional rectangular system.

diffusion and/or convection on the crystal growth rate of ZnSe and its distribution across an interface. In this numerical study, a two-dimensional model is used for the analysis of the PVT processes during vapor-growth of ZnSe single crystals in horizontally oriented, closed ampoules in a two-zone furnace. Consider a rectangular enclosure of height H and transport length L, shown in Fig. 1. The source is maintained at a temperature T_s, while the growing crystal is at a temperature T_c, with $T_s > T_c$. The source material ZnSe is sublimated at x = 0 to form a single crystal at x = L. Since we consider the growth of crystals by dissociative sublimation, the binary compound ZnSe is assumed to obey the following reaction [24]:

$$ZnSe(s) = Zn(g) + \frac{1}{2}Se_2(g). \tag{1}$$
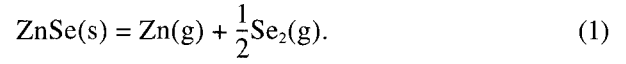
Thermodynamic equilibrium conditions at interfaces are as follows:

$$P_{Zn}(0)P_{Se_2}^{1/2}(0) = K_p(T_s) \quad \text{at } x = 0. \tag{2}$$

$$P_{Zn}(L)P_{Se_2}^{1/2}(L) = K_p(T_c) \quad \text{at } x = L. \tag{3}$$

The equilibrium constant $Kp = P_{Zn}P_{Se_2}^{1/2}$ [26] for reaction (1) at temperature T is

$$\log_{10}P_{Zn}P_{Se_2}^{1/2}(T) = -18900/T + 10.003 \text{ (atm)}. \tag{4}$$

The Kp can be expressed as RT $\ln(P_{Zn}P_{Se_2}^{1/2})$
$$= -361807 + 191.485T \text{ (J/mol)} \tag{5}$$

Also, the ratio of the partial pressures, s of Zn and Se_2 at the source is assumed not equal to the stoichiometric value of 2,

$$s = \frac{P_{Zn}(0)}{P_{Se_2}(0)} \neq 2. \tag{6}$$

Therefore, with s specified, the partial pressures $P_{Zn}(0)$ and $P_{Se2}(0)$ can be found by solving (2), (4) and (6) and with the total pressure assumed nearly constant, we have

$$P_t = P_{Zn}(0) + P_{Se_2}(0) \approx P_{Zn}(L) + P_{Se_2}(L). \tag{7}$$

The transport of fluid within a rectangular PVT crystal growth reactor is governed by a system of elliptic, coupled conservation equations for mass (continuity), momentum, energy and species (diffusion) with their appropriate boundary conditions. Let u_x, u_y denote the velocity components along the x- and y-coordinates in the x, y rectangular coordinate, and let T, ω_A, p denote the temperature, mass fraction of species A, Zn(g) and pressure, respectively.

The dimensionless variables scaled are shown in Ref.

[27] and the dimensionless governing equations are given by:

$$\nabla^* \bullet V = 0, \tag{8}$$

$$\vec{V} \bullet \nabla^* \vec{V} = -\nabla^* p^* + Pr \cdot Ar \nabla^{*2} \vec{V} \\ -Gr \cdot Pr^2 \cdot Ar^2 T^* \cdot e_g, \tag{9}$$

$$\vec{V} \cdot \nabla^* T^* = Ar \nabla^{*2} T^*, \tag{10}$$

$$\vec{V} \bullet \nabla^* \omega_A^* = \frac{Ar}{Le} \nabla^{*2} \omega_A^*. \tag{11}$$

These nonlinear, coupled sets of equations are numerically integrated with the following boundary conditions:
On the source ($x^* = 0, 0 < y^* < 1$):

$$u(0,y^*) = -\frac{Ar}{Le(v-1)} \frac{1}{\partial x^*} \frac{\partial \omega_A^*}{\partial x^*}, \tag{12}$$

$$v(0, y^*) = 0,$$
$$T^*(0, y^*) = 1,$$
$$\omega_A^*(0, y^*) = 1.$$

On the crystal ($x^* = L/H, 0 < y^* < 1$):

$$u(L/H, y^*) = -\frac{Ar \Delta \omega \partial \omega_A^*}{Le \, C_v \, \partial x^*}, \tag{13}$$

$$v(L/H, y^*) = 0,$$
$$T^*(L/H, y^*) = 0,$$
$$\omega_A^*(L/H, y^*) = 0.$$

Walls ($0 < x^* < L/H$, $y^* = 0$ and 1):

$$u = v = \frac{\partial \omega_A^*}{\partial y^*} = 0, \tag{14}$$

$$T^*(x^*, 0) = T^*(x^*, 1) = -\frac{1}{Ar} \cdot x^* + 1$$

Thermo-physical data are obtained from Refs. [28]. The thermophysical properties of the gas mixture are estimated from gas kinetic theory using Chapman-Enskog's formulas [29]. The nomenclatures related to this study are found in Ref. [27].

3. Results and Discussion

The parametric study is useful for showing trends and generalizing the problem, but many parameters are involved in the problem under consideration, which renders it difficult for a general analysis. One of the purposes for this study is to correlate the growth rate, for a particular material (ZnSe), to process parameters: (a) partial pressure ratio, s, and (b) temperature gradient

across the ampoule. Thus, it is desirable to express some results in terms of dimensional growth rate, however they are also applicable to parameter ranges over which the process varies in the manner given. For this application, ranges of process parameters are typical for PVT processes under ground-based laboratory conditions. The six dimensionless parameters, namely Gr, Ar, Pr, Le, C_v and Pe, are independent and arise naturally from the dimensionless governing equations and boundary conditions. Because the precise values of s that correspond to the experiments are unknown, as a first step we assume values of s which are close to experimental growth conditions. Then one simulates diffusive-convective cases corresponding to actual experiments. The results shown in Figs. 3 through 9 are based on Ar = 6.67, H = 1.5 cm, $T_s = 1160°C$.

3.1. 1-D advective-diffusion model

One examines one-dimensional advective-diffusive case based on Stefan flow in order to understand the basic phenomena in the PVT process, which serves as numerics. The predictions based on 1-D advectivediffusive Stefan model shows a uniform streaming velocity from the source to the crystal [21]. Note that the 1-D case corresponds to the case simulated for the case when the zero-stress condition and Gr = 0 are employed.

Figures 2 and 3 show the crystal growth rates of ZnSe as a function of temperature difference, ΔT (K). One sees that the closer to the stoichiometric case (s = 2), the larger is the growth rate. It means the rate is much
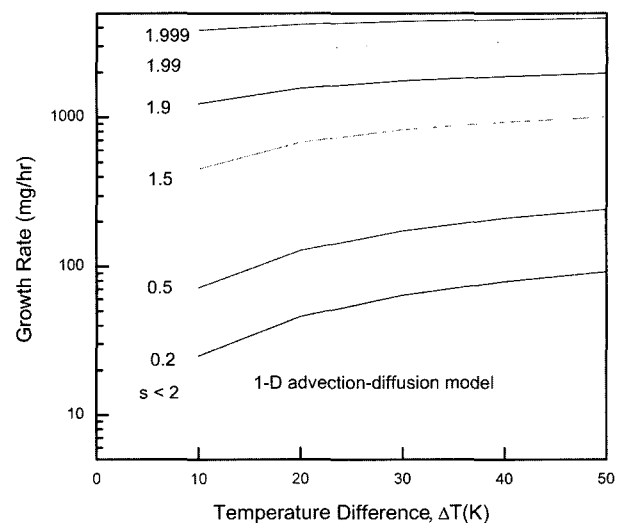

Fig. 2. The crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $0.2 \le s \le 1.999$, based the 1-D advection-diffusion (Stefan flow) model.
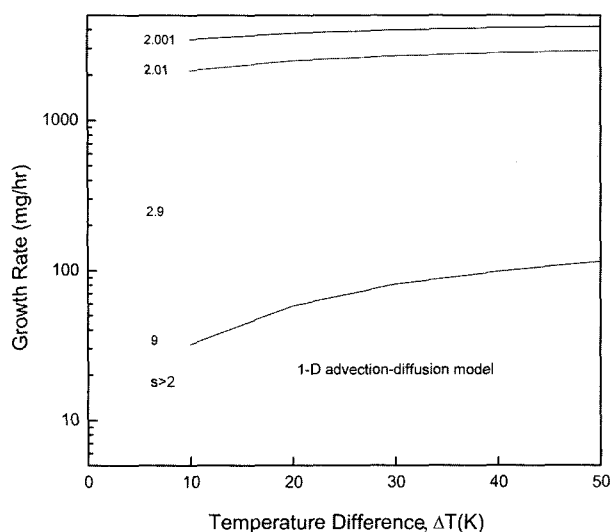
Fig. 3. The crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $2.001 \leq s \leq 9$, based on the 1-D advection-diffusion (Stefan flow) model.

sensitive to the variation of s. One sees in Fig. 2 that for $10 K \leq \Delta T = 50 K$, the rates for $1.9 \geq s$ have three order of magnitude mg/hr, but for $0.5 \leq s < 1.9$, the rates have two order of magnitude. For $s < 0.5$, the rates are of one order of magnitude. At $\Delta T = 10 K$, the rate with $s = 1.5$ is greater than $s = 0.2$ by a factor of 2.56. On the other hand, at $\Delta T = 50 K$, the former is greater than the latter by a factor of 1.14. It reflects that when ΔT is smaller, the larger is an increase in the rate with the ratio s. For the temperature differences closer to 50 K and the ratios of $1.5 < s < 1.999$, the slopes of growth rate versus ΔT are relatively small. The rate sharply increases with increasing ΔT until ΔT reaches 20 K, and after the value, it increases much slowly with ΔT up to 50 K. Therefore, a variation of the ratio s is a parameter more important than that of ΔT when ΔT is larger than 20 K. At $s = 1.999$, the rate is little sensitive to ΔT. Similarly, at $s = 2.001$, the same results are obtained, as shown in Fig. 3. Note that as $s \rightarrow 2$ the mechanism of PVT switches from dissociative sublimation to congruent sublimation. As seen in Fig. 3, the rates for $2.001 \leq s \leq 2.01$ have three order of magnitude and at $s = 2.9$, two order of magnitude, and at $s = 9$, one order of magnitude. For $\Delta T = 10 K$, the rate with $s = 9$ is greater than $s = 2.9$ by a factor of 6.56. On the other hand, at $\Delta T = 50 K$, the former is greater than the latter by a factor of 5.01. Compared with the zinc-deficient case, $s < 2$, at $\Delta T = 50 K$, the rate with $s = 0.5$ is greater than $s = 1.5$ by a factor of 4.15. Therefore, the rate is linearly proportional to the value of s for the cases of zinc-deficient and -rich. For $20 K \leq \Delta T = 50 K$, the differences

between the rates of $s = 2.9$ and 9 are nearly variant. As the partial pressure ratio s approaches 2, the Stefan velocity approaches infinity [25]. This can be seen from boundary condition (15) since $C_v \rightarrow 1$ as $s \rightarrow 2$ and hence u (dimensionless x-component velocity) $\rightarrow \infty$. It must be pointed out that this limiting behavior only serves to show a mathematical trend. Even though the velocity increases dramatically, it is bounded. In order to operate near this singular case, we consider two kinds of values of s where the transport rate is limited by stichiometric variations that is either $s > 2$ (Zn-rich case) or $s < 2$ (Zn-deficient case). Evidently, the ratio s of the components in the vapor phase at the source material has a dramatic effect on the rate of transport. As seen in Figs. 2 and 3, the closer to the stoichiometric case, the larger is the rate. Note that these figures are similar to the plots obtained using the 2D model, as shown later.

3.2. 2-D diffusive-convection model

The cylinder horizontally oriented has a diameter $H = 1.5 cm$ and transport length $L = 10 cm$. The source temperature T_s is $1160°C$. The temperature profile along the side of cylinder is experimentally measured. A Zn-deficient case $(s < 2)$ is considered to mimic the experimental conditions. One calculates the total pressure in the ampoule to be about 20 Torr. The physical properties and system parameters for the 2-D diffusive-convection numerical computations with $s = 0.2$ are listed in Table 1. Typical calculations were performed for a stoichiometry parameter at the source $s = 0.2$ which is close to experimental growth conditions (Zn-deficient vapor phase).

Table 1
Typical thermo-physical properties used in simulations ($M_{Zn} = 65.39$, $M_{Se_2} = 157.92$)

| Transport length, L	10 cm
Height, H	1.5 cm
Source temperature, T_s	1160
Crystal temperature, T_c	1130
Density, ρ	3.26×10^{-5} g/cm^3
Dynamic viscosity, μ	0.5×10^{-5} g/cm·sec
Diffusivity, D_{AB}	41.8 cm^2/s
Thermal expansion coefficient, β	6.98×10^{-4} K^{-1}
Prandtl number, Pr	0.76
Schmidt number, Sc	0.48
System stoichiometry (s)	0.2
Total system pressure, P_T	20.08 Torr
Zn partial pressure, source (s = 0.2)	3.35 Torr
Zn partial pressure, crystal (s = 0.2)	16.7 Torr
Se$_2$ partial pressure, source (s = 0.2)	2.17 Torr
Se$_2$ partial pressure, crystal (s = 0.2)	17.9 Torr
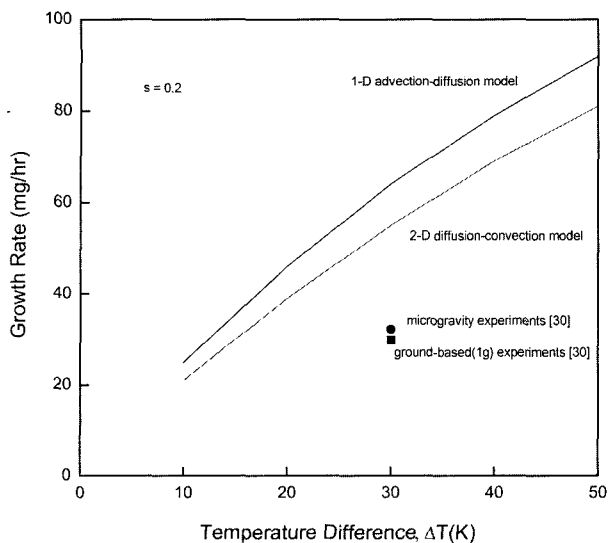
Fig. 4. The crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $s = 0.2$, $0.04 \leq Pe \leq 0.16$, $2.95 \times 10^{-6} \leq Gr_t \leq 0.056$, $4.62 \times 10^{-6} \leq Gr_s \leq 0.01$. For $\Delta T(K) = 10$ K, $Pe = 0.04$, $Gr_t = 0.056$, $Gr_s = 0.01$; in the case of $\Delta T(K) = 10$ K, $Pe = 0.16$, $Gr_t = 2.95 \times 10^{-6}$, $Gr_s = 4.62 \times 10^{-6}$. The data of ● and ■ represent the growth rates obtained under the microgravity and ground-based (1 g) experiments, respectively [30].

Fig. 5. The crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $s = 2.9$, $0.366 \leq Pe \leq 1.00$, $4.65 \times 10^{-3} \leq Gr_t \leq 2.0 \times 10^{-2}$, $-8.5 \times 10^{-2} \leq Gr_s \leq -2.39 \times 10^{-2}$. For $\Delta T(K) = 10$ K, $Pe = 0.366$, $Gr_t = 4.65 \times 10^{-3}$, $Gr_s = -2.39 \times 10^{-2}$; in the case of $\Delta T(K) = 50$ K, $Pe = 1.00$, $Gr_t = 2.0 \times 10^{-2}$, $Gr_s = -8.5 \times 10^{-2}$.

Figure 4 shows the growth rate as a function of temperature difference for two different models, 1-D Stefan and 2-D diffusive-convection model. The results show that for $s = 0.2$, 1-D case is slightly greater than the 2-D case for the temperature differences examined. The 2-D numerical predictions of crystal growth rates fall within the range of 1-D theoretical estimations. This reflects that for the growth conditions of ZnSe the flow fields would be 1-D advective-diffusive. For the temperature ranges of 10 K $\leq \Delta T \leq 50$ K, the variations of growth rate with the temperature differences are nearly same for the two models, which supports the advective-diffusion is dominant over the flow fields within the growth enclosures. For comparisons with others' results [30], the growth rates obtained under the microgravity and ground-based (1 g) experiments are chosen and the corresponding data represent ● and ■, respectively. These experimental results are based on a diameter of 1.1 cm and $\Delta T = 30$ K. For comparisons with ours, the mass per unit hour based on a diameter of 1.5 cm are compensated by a factor of $(1.5/1.1)^2$. The rates predicted by the 1-D advection-diffusion and 2-D diffusion-convection models are of same order of magnitude as those in the microgravity and the ground-based experiments [30]. The former is nearly greater than the latter by a factor of 2. The disparity between experiments and our predictions results point to the fact that under the micrograv-
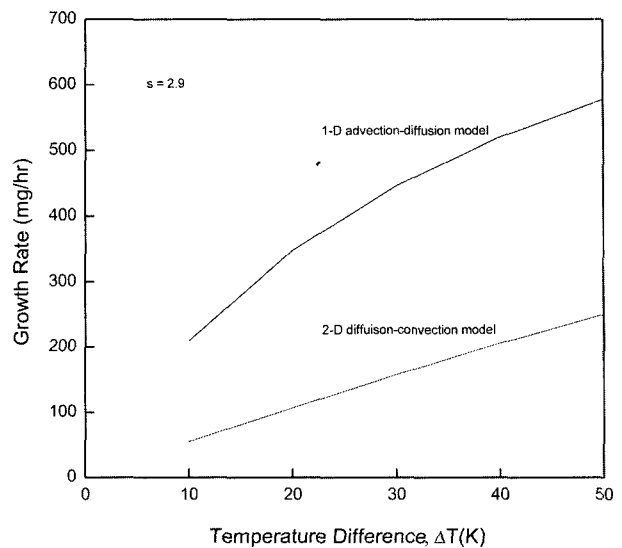
ity and the ground-based experimental conditions, the transport mechanisms are likely to be masked or superimposed by thermal convection, solutal convection, oscillatory double-diffusive convection, and Soret diffusion. From our predictions the growth rate based on 2-D is still very close to that of the Stefan flow case. Furthermore, the difference between the microgravity and ground-based experiments is not large. These results indicate that convection is likely to be very weak and steady.

Figure 5 shows the growth rate as function of temperature difference for $s = 2.9$, $0.366 \leq Pe \leq 1.00$, $4.65 \times 10^{-3} \leq Gr_t \leq 2.0 \times 10^{-2}$, $-8.5 \times 10^{-2} \leq Gr_s \leq -2.39 \times 10^{-2}$. $s = 2.9$ is chosen, which is close to experimental growth conditions (Zn-rich vapor phase) [26]. The trend similar to Fig. 4 is obtained, but the rate of 1-D case is slightly greater than the 2-D. The growth rate for the Zn-deficient case ($s = 0.2$) is greater than the Zn-rich case ($s = 2.9$) by approximately one order of magnitude, which is due to the effect of mass fluxes on the interfaces, i.e., sublimation and condensation. These effects appear through the dimensionless parameter, Peclet number. For the 10 K $\leq \Delta T \leq 50$ K, the former has $0.04 \leq Pe \leq 0.16$, whereas the latter has $0.366 \leq Pe \leq 1.00$.

Figure 6 shows the crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $0.2 \leq s \leq 1.999$, $0.04 \leq Pe \leq 8.04$, $0.04 \leq Gr_t \leq 0.056$, $0.01 \leq Gr_s \leq 0.33$, based on the 2-D numerical analysis based on
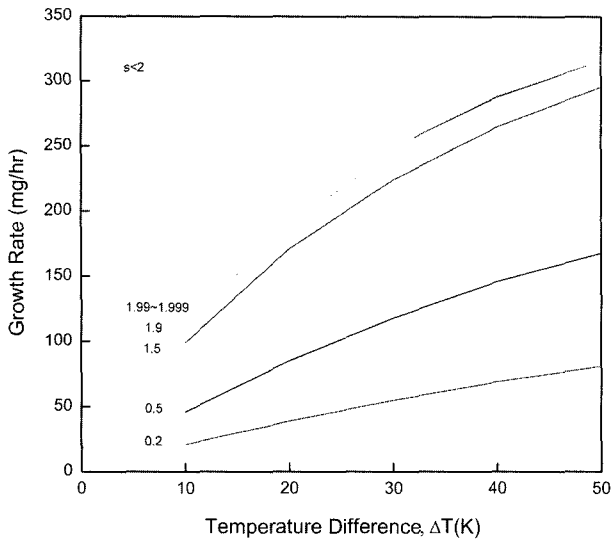
Fig. 6. The crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $0.2 \leq s \leq 1.999$, $0.04 \leq Pe \leq 8.04$, $0.04 \leq Gr_t \leq 0.056$, $0.01 \leq Gr_s \leq 0.33$, based on the 2-D numerical analysis based on the diffusion-limited model. For s $= 0.2$, $Pe = 0.04$, $Gr_t = 0.056$, $Gr_s = 0.01$; in the case of s $= 1.999$, $Pe = 8.04$, $Gr_t = 0.04$, $Gr_s = 0.33$.

the diffusion-limited model. The curves for $0.2 \leq s \leq 1.999$ have similar profiles for the temperature differences examined, $10 K \leq \Delta T \leq 50 K$, which indicates similar variation of the rate over the temperature difference. As the ratio of partial pressure approaches the stoichiometric value, s $= 2$ from s $= 1.5$, the rate increases sharply. From s $= 0.2$ to 0.5, the rate are slightly varied.

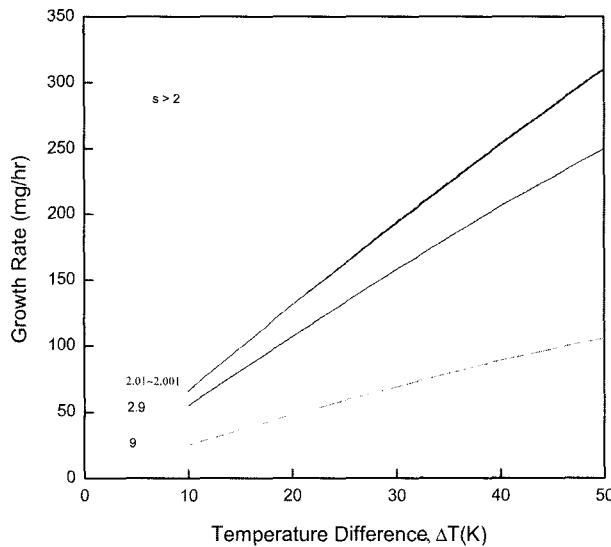

Fig. 7. The crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $2.001 \leq s \leq 9$, $0.2 \leq Pe \leq 7.35$, $Gr_t = 0.02$, $-0.039 \leq Gr_s \leq -0.12$, based on the 2-D numerical analysis based on the diffusion-limited model. For s $= 2.001$, $Pe = 7.35$, $Gr_t = 0.02$, $Gr_s = -0.12$; for s $= 9$, $Pe = 0.2$, $Gr_t = 0.02$, $Gr_s = -0.039$.

Figure 7 shows the crystal growth rate of ZnSe as a function of temperature difference, $\Delta T(K)$ for $2.001 \leq s \leq 9$. As the ratio of partial pressure approaches the stoichiometric value, s $= 2$ from s $= 2.9$, the rate increases sharply. From s $= 9$ to 2.9, the rate are slightly varied. As seen in Figs. 6 and 7, the similar profiles of the rate are obtained for s $= 0.2$ and 2.9, except for the difference of the rate by one order of magnitude between s $= 0.2$ and 2. Therefore, it is concluded that as the ratio of partial pressure approaches the stoichiometric value, s $= 2$ for the zinc-deficient case (s < 2) and zinc-rich case (s > 2), the rate increases sharply with the temperature difference, whereas as the ratio deviates from s $= 2$, the rate is increased slightly with the temperature difference.

For the effects of gravitational accelerations on the rate, the gravitational constant is varied from 1 g to 10^{-6} g for $\Delta T = 50 K$ and s $= 1.5$, the rates remain nearly constant, i.e., 211 mg/hr, which are not shown here and performed for our confirmations of other results [25, 26]. Therefore, the transport mechanism of diffusive-convection is independent of gravity. In other words, reducing the gravitational level such that Gr $\rightarrow$ 0, as provided by microgravity environments, would not affect the diffusive-convection flow fields for low vapor pressure system such as ZnSe vapor growth processes.

4. Conclusions

As the ratio of partial pressure approaches the stoichiometric value, s $= 2$ for the zinc-deficient case (s < 2) and zinc-rich case (s > 2), the rate increases sharply with the temperature difference, whereas as the ratio deviates from s $= 2$, the rate is increased slightly with the temperature difference. As the value of the ratio of partial pressure approaches the stoichiometric value, s $= 2$ from s $= 1.5$ through s $= 1.999$, the rate increases sharply. From s $= 0.2$ to 1.5, the rate are slightly varied. The similar profiles of the rate are obtained for s $= 0.2$ and 2.9, except for the rate by one order of magnitude. The mass flux based on one dimensional (1D model) flow for low vapor pressure system fall within the range of the predictions (2D model), which indicates the flow fields would be advective-diffusive. These results indicate the rate and the flow fields would be independent of gravity acceleration levels. Also, for the effects of gravitational accelerations on the rate, the gravitational constant is varied from 1 g to 10^{-6} g for $\Delta T = 50 K$ and s $= 1.5$, the rates remain nearly constant, i.e., 211 mg/hr, which Stefan flow is dominant over convection.

Acknowledgment

References

[1] H. Kukimoto, "Conductivity control of ZnSe grown by MOVPE and its application for blue electroluminescence," J. Crystal Growth 101 (1990) 953.

[2] H.J. Eichler, V. Glaw, A. Kummrow, V. Penschke and Wahi, "Optically bistable thin film devices using wide-gap II-VI compounds," J. Crystal Growth 101 (1990) 695.

[3] S. Guha, J.M. DePuydt, M.A. Hasse, J. Qiu and H. Cheng, Appl. Phys. Lett. 63 (1993) 3107.

[4] M.A. Hasse, J.M. DePuydt and H. Chen, Appl. Phys. Lett. 59 (1991) 1272.

[5] H. Jeon, J. Ding, A.V. Nurmikko, W. Xie, D.C. Grillo, M. Kobayashi, R.L. Gunshor, G.C. Hua and N. Otsuka, Appl. Phys. Lett. 60 (1992) 2045.

[6] W.C. Holton, R.K. Watts and R.D. Stinedurf, Eichler, J. Crystal Growth 6 (1969) 97.

[7] M.J. Kozielski, "Polytype single crystals of $Zn_{1-x}Cd_xS$ and $ZnS_{1-x}Se_x$ solid solutions grown from the melt under high argon pressure by bridgmans method," J. Crystal Growth 30 (1975) 86.

[8] I. Kikuma and M. Furukoshi," Melt growth of ZnSe crystals under argon pressures," J. Crystal Growth 41 (1977) 103.

[9] C.C. Chang, C.C. Wei, Y.K. Su and H.C. Tzeng, "Growth and characterization of ZnSe single crystals by closed tube methods," J. Crystal Growth 84 (1987) 11.

[10] K. Mochizuki, K. Masumoto and H. Iwanaga, "Morphology and photoluminescence (PL) of ZnSe single crystals grown frm Se and/or as solvents," J. Crystal Growth 84 (1987) 1.

[11] H. Hartmann, "Studies on the vapour growth of ZnS, ZnSe and ZnTe single crystals," J. Crystal Growth 42 (1977) 144.

[12] W. Palosz, "Growth of ZnS and $Zn_{1-x}Cd_xS(x < 0.07)$ single crystas by iodine transport," J. Crystal Growth 60 (1982) 57.

[13] K. Böttcher and H. Hartmann, "Zinc selenide single crystal growth by chemical transport reactions", J. Crystal Growth 146 (1995) 53.

[14] J.R. Cutter and J. Woods, "Growth of single crystals of zinc selenide from the vapor phase", J. Crystal Growth 47 (1979) 405.

[15] H.Y. Cheng and E.E. Anderson, "The growth of ZnSe single crystals by physical vapor transport", J. Crystal Growth 96 (1989) 756.

[16] Y.U. Korostelin, V.I. Kozlovsky, A.S. Nasibov and P.V. Shapkin, "Vapour and characterization of bulk ZnSe single crystals", J. Crystal Growth 161 (1996) 51.

[17] C.H. Su, S. Feth and S.L. Lehoczky, "In situ partial pressure measurements and visual observation during crystal growth of ZnSe by seeded physical vapor transport", J. Crystal Growth 209 (2000) 687.

[18] C.H. Su, M.A. George, W. Palosz, S. Feth and S.L. Lehoczky, "Contactless growth of ZnSe single crystals by physical vapor transport", J. Crystal Growth 213 (2000) 267.

[19] C.H. Su, S. Feth, L.J. Wang and S.L. Lehoczky, "Photoluminescene studies of ZnSe starting materials and vapor grown bulk crystals", J. Crystal Growth 224 (2001) 32.

[20] F. Rosenberger, "Fluid dynamics in crystal growth from vapors", Physico-Chemical Hydro-dynamics 1 (1980) 3.

[21] M.M. Faktor and I. Garrett, Growth of Crystals from the Vapour, Chapman & Hall, London (1974).

[22] Y.-G. Sha, C.-H. Su, W. Palosz, M.P. Volz, D.C. Gillies, F.R. Szofran, S.L. Lehoczky, H.-C. Liu and R.F. Brebrick, "Mass flux of ZnSe by physical vapor transport", J. Crystal Growth 146 (1995) 42.

[23] C.-H. Su, "Growth rate of CdS by vapor transport in a closed ampoule," J. Crystal Growth 80 (1987) 333.

[24] D.W.G. Ballentyne, S. Wetwatana and E.A.D. White, "Kinetics of the vapour-phase growth of II-VI compounds in a sealed tube," J. Crystal Growth 7 (1970) 79.

[25] H. Zhou, A. Zebib, S. Trivedi and W.M.B. Duval, "Physical vapor transport of zinc-telluride by dissociative sublimation", J. Crystal Growth 167 (1996) 534.

[26] N. Ramachandran, C.H. Su and S.L. Lehoczky, "Modeling studies of PVT growth of ZnSe: current status and future course", J. Crystal Growth 208 (2000) 269.

[27] G.T. Kim, "Convection in the growth of zinc telluride single crystal by physical vapor transport," J. Korean Crystal Growth and Crystal Tech. 4 (2003) 187.

[28] R.F. Brebrick and H.-C. Liu, High Temp. Mater. Sci. 35 (1996) 215.

[29] R.C. Reid, J.M. Prausnitz and B.E. Poling, Bird, The properties of gases & liquids (McGraw-Hill, New York, NY, 1988).

[30] D. Siche, K. Bottcher, U. Rinas and H. Hartmann, "Crystal growth of zinc selenide under μg-conditions," J. Crystal Growth 244 (2002) 249.