

## 지상 및 미소중력 환경에서 물리적 승화법 공정에 미치는 불순물의 영향 분석: 염화제일수은에 대한 응용성

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## Numerical Analysis for Impurity Effects on Diffusive-convection Flow Fields by Physical Vapor Transport under Terrestrial and Microgravity Conditions: Applications to Mercurous Chloride

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### 초 록

본 연구에서는 지상 및 미소중력환경하에서 물리적 승화법 공정에서의 확산-대류유동에 미치는 불순물의 영향을 이론적으로  $\text{Hg}_2\text{Cl}_2\text{-I}_2$  시스템에 적용하여 규명하는 것이다. 이론적 해석은 증기상에서 확산-대류 흐름, 열 및 물질전달을 속도 벡터 흐름, 유선, 온도, 농도 분포를 통하여 제시된다. 결정 영역에서의 전체 몰플럭스는 중력가속도와 성분  $\text{I}_2$  불순물에 상당히 민감하게 반응한다. 성분  $\text{I}_2$ 를 증가시켰을 때, 농도 대류효과는 확산-대류 유동흐름을 안정화시키는 경향이 있다. 지상중력가속도의 0.001환경에서는 유동흐름은 1차원포물선의 흐름 구조를 나타내며, 확산지배형태를 보여주고 있다.  $10^{-3}$ 지상중력가속도 이하에서는 대류 영향은 무시할 수 있다.

### Abstract

In this study, impurity effects on diffusive-convection flow fields by physical vapor transport under terrestrial and microgravity conditions were numerically analyzed for the mixture of  $\text{Hg}_2\text{Cl}_2\text{-I}_2$  system. The numerical analysis provides the essence of diffusive-convection flow as well as heat and mass transfer in the vapor phase during the physical vapor transport through velocity vector flow fields, streamlines, temperature, and concentration profiles. The total molar fluxes at the crystal regions were found to be much more sensitive to both the gravitational acceleration and the partial pressure of component  $\text{I}_2$  as an impurity. Our results showed that the solutal effect tended to stabilize the diffusive-convection flow with increasing the partial pressure of component  $\text{I}_2$ . Under microgravity conditions below  $10^{-3}g_0$ , the flow fields showed a one-dimensional parabolic flow structure indicating a diffusion-dominant mode. In other words, at the gravitational levels less than  $10^{-3}g_0$ , the effects of convection would be negligible.

**Keywords:** microgravity, crystal growth

## 1. Introduction

Over the past 45 years since the results of the Apollo space flights, many researches on the role of gravity as a major parameter in materi-

als science and engineering, crystal growth have been extensively carried out in the United States, Russia, Germany, and Japan etc. In particular, through the International Space Station (ISS), various experiments have been carried out based on the American Skylab or Spacelab missions, the German D1 and D2 missions, the Russian Mir station of Foton flights, the European Eureka mission, or the German and the Japanese sounding rocket programs[1]. Fontana *et al.*[2] had investigated crystal growth of sodium chloride in both terrestrial and microgravity environments on the ISS. Single crystal growth diffraction

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shows no change in crystalline structure and cell parameters of NaCl in crystals grown under the microgravity condition of  $10^{-6}g_0$ , where  $g_0$  denotes the Earth's gravitational acceleration of  $981 \text{ cm s}^{-2}$ . In 2014, Japanese researchers of Nobeoka *et al.*[3] have reported the numerical simulations of InGaSb crystal growth crystal growth by temperature gradient method on board at the ISS to get a complete understanding for the transport phenomena occurring in the melt system, i.e., in the microgravity fields. Kinoshita *et al.*[4] have grown a silicon germanium alloy crystal  $\text{Si}_{0.5}\text{Ge}_{0.5}$  with 10 mm in diameter by traveling liquidus-zone method in microgravity. Abe *et al.*[5] have studied the numerical simulations of SiGe crystal growth by traveling liquidus-zone method under the microgravity environments. In recent years, many studies on numerical simulations have been carried out under microgravity conditions in various countries[6-18]. In particular, Konishi and Mudawar[19] reported reviews of flow boiling and critical heat flux in microgravity. Carotenuto[20] addressed reviews and relevant results on crystal growth from the vapour phase under microgravity environments, which would be contributed to future microgravity research on board the International Space Station. Yeckel and Derby[21] had investigated the g-jitter (transient acceleration) with steady magnetic fields in vertical Bridgman crystal growth system whose results were relevant to dynamics of three-dimensional convection in microgravity crystal growth. For transverse jitter at intermediate frequencies, flow oscillations were found to be increased, considering in most cases, application of a magnetic field suppresses flow oscillations. Zeng *et al.*[22] investigated three-dimensional oscillatory Marangoni convection in silicone oil liquid bridge under microgravity environments and provided temperature contour lines and projected velocity vector for the range of Marangoni number, from  $2.0 \times 10^4$  up to  $5.0 \times 10^4$ . Maekawa *et al.*[23] studied the possibility of growing a uniform binary compound crystal in space for InAs-GaAs binary semiconductor by the Bridgman and zone methods. The solution-crystal interface was found to be deformed by buoyancy driven convection in the case of the Bridgman method even in the gravitational acceleration of  $10^{-6}g_0$  which is supposed to be the gravity level in the International Space Station. Liu *et al.*[24] carried out simulation of directional solidification or refined Al-7 wt% Si Alloys with comparison with benchmark microgravity experiments.

In general, the crystal growth method by physical vapor transport (PVT) is referred to as a mechanism of sublimation-condensation. The source component-bearing species are vaporized from the source material and recrystallized on a seed crystal, in closed silica glass ampoules in temperature gradient imposed between the source material and the growing crystal. With regard to studies on the PVT in the vapor phase, recently Tebbe *et al.*[25] extended for transition to chaos flow fields in specialty materials of mercurous chloride in applications of microgravity experiments. They have addressed the underlying phenomena in the PVT processes on the relative importance and influencing parameters of diffusion-advection, thermal and/or solutal convection on mass transport. It is reported that  $\text{Hg}_2\text{Cl}_2$  single crystals have applications for acousto-optic and opto-electronic devices such as Bragg cells, X-ray detectors operating at ambient temperatures[26]. In study we employ a material of  $\text{Hg}_2\text{Cl}_2$  as a model sample and investigate numerically the influence of

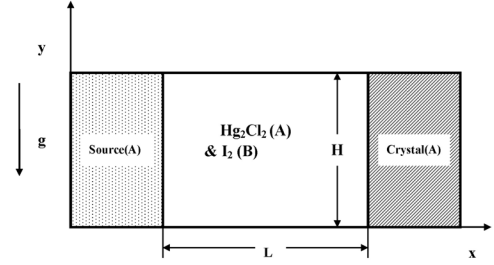


Figure 1. Schematic description of a two-dimensional PVT model for a  $\text{Hg}_2\text{Cl}_2$ - $\text{I}_2$  system.

gravitational accelerations on diffusive convection during the PVT of a mixture of  $\text{Hg}_2\text{Cl}_2$  vapor and impurity of  $\text{I}_2$ . The distinction of this study in a comparison with our previous study[27-28] is centered into the effects of gravitational accelerations and the partial pressure of component  $\text{I}_2$  with high molecular weight through numerical simulations of vector, streamline, temperature and concentration profiles.

## 2. Numerical Analysis

A physical model is created with a two-dimensional rectangular enclosure with aspect ratio (transport length  $L$  to height  $H$ ) of 5, as shown in Figure 1. The source is maintained at a temperature  $T_s$ , while the growing crystal is at a temperature  $T_c$ , with  $T_s > T_c$ . PVT of the transported component A ( $\text{Hg}_2\text{Cl}_2$ ) occurs inevitably, due to presence of impurity, i.e., a component B ( $\text{I}_2$ ). 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), Eq. (1) with appropriate boundary conditions, Eqs. (2)-(4), which can be found in Refs.[27-29]. The density of the vapor mixture of component A ( $\text{Hg}_2\text{Cl}_2$ ) and B ( $\text{I}_2$ ) is assumed constant except the buoyancy body force term, which would be a function of both temperature and concentration.

$$\nabla \cdot (\rho u \phi) = \nabla \cdot (\Gamma \nabla \phi) + S \quad (1)$$

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

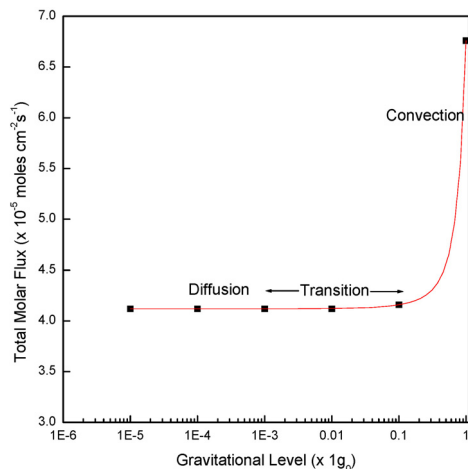
$$u^*(x^*, 0) = u^*(x^*, 1) = v^*(x^*, 0) = v^*(x^*, 1) = 0 \quad (2)$$

$$\frac{\partial \omega_A^*(x^*, 0)}{\partial y^*} = \frac{\partial \omega_A^*(x^*, 1)}{\partial y^*} = 0,$$

$$T^*(x^*, 0) = T^*(x^*, 1) = \frac{T - T_c}{T_s - T_c}$$

On the source ( $x^* = 0$ ,  $0 < y^* < 1$ ) :

$$\begin{aligned} u^*(0, y^*) &= -\frac{1}{Le} \frac{\Delta \omega}{(1 - \omega_{A,s})} \frac{\partial \omega_A^*(0, y^*)}{\partial x^*} \\ v^*(0, y^*) &= 0, \\ T^*(0, y^*) &= 1, \\ \omega_A^*(0, y^*) &= 1. \end{aligned} \quad (3)$$



**Figure 2.** Effects of gravitational levels on the total molar flux for various gravitational accelerations,  $10^{-5}g_0 \leq g \leq 1g_0$ , in the negative y-direction, where  $g_0$  denotes the Earth's gravitational acceleration of  $981 \text{ cm s}^{-2}$ . Based on aspect ratio = 5 ( $L = 10 \text{ cm}$ ), a linear wall temperature profile between  $T_s = 623.5 \text{ K}$  and  $T_c = 563.15 \text{ K}$ .

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

$$u^*(L/H, y^*) = -\frac{1}{Le(1-\omega_{A,c})} \frac{\partial \omega_A^*(L/H, y^*)}{\partial x^*} \quad (4)$$

$$v^*(L/H, y^*) = 0,$$

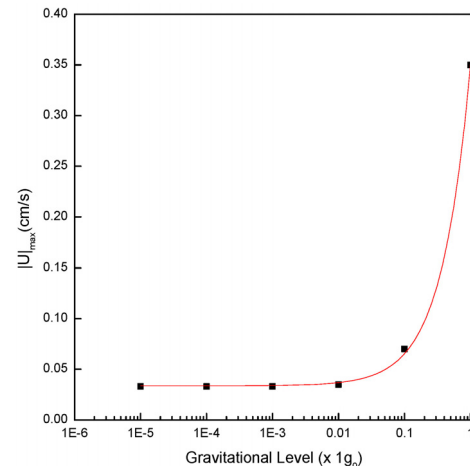
$$T^*(L/H, y^*) = 0,$$

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

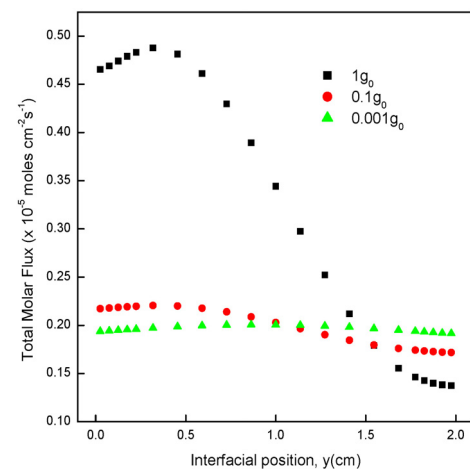
### 3. Results and Discussion

The purpose of this research is to perform the numerical analysis of the influence of impurity and gravitational levels on diffusive-convection flow fields by the PVT of  $\text{Hg}_2\text{Cl}_2$  with impurity of  $\text{I}_2$ , from a point of view of the total mass flux and interfacial distributions. Because the molecular weight of an impurity ( $\text{I}_2$ ) is not equal to that of the crystal component ( $\text{Hg}_2\text{Cl}_2$ ) during the physical vapor transport, both thermal and/or solutal convection always occur and also become much important in mass transport. In this study, a linear temperature profile at the walls is chosen. Detailed data such as typical process parameters and physical properties for the operating conditions could be found in Refs.[27-29].

Figure 2 shows our numerical results for the effects of gravitational levels on the total molar flux for various gravitational accelerations,  $10^{-5}g_0 \leq g \leq 1g_0$ , in the negative y-direction, where  $g_0$  denotes the Earth's gravitational acceleration of  $981 \text{ cm s}^{-2}$ . The convection is predominant over the diffusion for the range from  $1g_0$  down to  $0.1g_0$ . The total molar flux is sharply decreased with decreasing the gravitational level from  $1g_0$  down to  $0.1g_0$ . From a point of view of the total mass flux, the transition from the convection to the diffusion mode occurs in the range of  $10^{-2}g_0 \leq g \leq 0.1g_0$ , and reflects the diffusion-convection mode. There is little difference of the total mass flux between  $0.1g_0$  and  $10^{-2}g_0$ . The total molar flux in a convection region of  $1g_0$



**Figure 3.** Effects of gravitational levels on  $|U|_{\max}$  for various gravitational accelerations,  $10^{-5}g_0 \leq g \leq 1g_0$ , corresponding to Figure 2.



**Figure 4.** Interfacial distributions of the total molar flux for three different gravitational levels,  $1g_0$ ,  $0.1g_0$  and  $10^{-3}g_0$ .

is greater than in the diffusion region of  $10^{-2}g_0$  by a factor of 1.6. It should be noted that the flow fields under terrestrial conditions include both the convection and diffusion, but the convection is predominant over the diffusion.

Figure 3 shows the effects of gravitational levels on  $|U|_{\max}$  for various gravitational accelerations,  $10^{-5}g_0 \leq g \leq 1g_0$ , corresponding to Figure 2. The  $|U|_{\max}$  denotes the dimensional maximum magnitude of velocity vector in the vapor phase, and reflects the intensity of convection in the vapor phase during the physical vapor transport of  $\text{Hg}_2\text{Cl}_2$  with impurity of  $\text{I}_2$ . As the gravitational level is decreased from  $1g_0$  down to  $0.1g_0$ , the  $|U|_{\max}$  under the condition of  $0.1g_0$  is reduced to one-fifth of the  $|U|_{\max}$  under the condition of  $1g_0$ . With decreasing the gravitational level from  $0.1g_0$  down to  $10^{-2}g_0$ , the  $|U|_{\max}$  under the condition of  $10^{-2}g_0$  leads to one half of the  $|U|_{\max}$  under the condition of  $0.1g_0$ . Considering little difference of total mass flux between  $0.1g_0$  and  $10^{-2}g_0$  previously, from a point of view of dimensional maximum magnitude of velocity vector, the transition from the convection to the diffusion mode occurs in the range of  $10^{-3}g_0 \leq g \leq 0.1g_0$ . From Figures 3 and 4, it is concluded that the transition from the convection

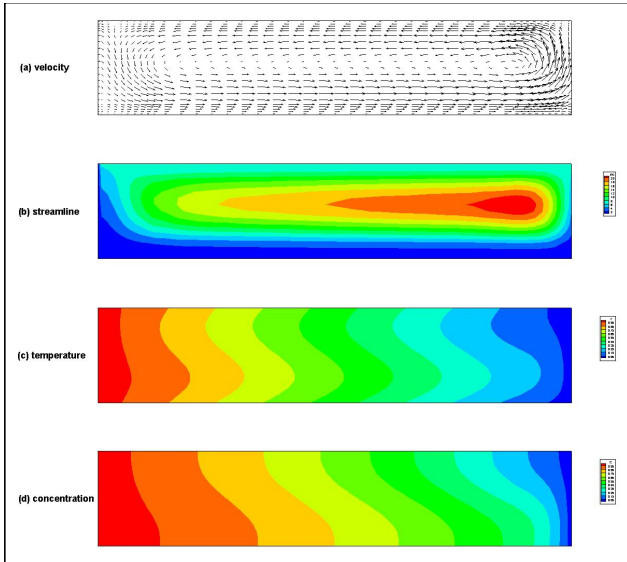


Figure 5. (a) velocity, (b) streamline ( $\Delta\psi = 2$ ,  $\psi_{\max} = 21.71$ ,  $\psi_{\min} = 2$ ), (c) temperature ( $\Delta T = 0.1$ ,  $T_{\max} = 1.0$ ,  $T_{\min} = 0$ ), (d) concentration ( $\Delta C = 0.1$ ,  $C_{\max} = 1.0$ ,  $C_{\min} = 0$ ), based on aspect ratio = 5,  $\Delta T = 60$  K (623.15 K  $\rightarrow$  563.15 K), and  $1g_0$ . A relative velocity vector with a magnitude of 100 has 1.4 cm. The maximum velocity vector is  $0.35 \text{ cm s}^{-1}$ .

to the diffusion mode occurs in the range of  $10^{-3}g_0 \leq g \leq 0.1g_0$ .

Figure 4 shows the interfacial distributions of the total molar flux for three different gravitational levels,  $1g_0$ ,  $0.1g_0$  and  $10^{-3}g_0$ . The case of  $1g_0$  shows the convective flow indicates asymmetrical at the position of  $y = 1$  cm and three dimensional flow structure. The case of  $0.1g_0$  is also likely to be slightly asymmetrical at the position of  $y = 1$  cm and three dimensional flow structure. It is clear that the profiles of the total molar flux against the interfacial position are symmetrical at the position of  $y = 1$  cm and nearly flat. The deviations of maximum and minimum total molar flux from the average total molar flux for  $0.1g_0$  and  $10^{-3}g_0$  are much smaller than for  $1g_0$ .

The mass transport phenomena occurring in the vapor phase when growing single crystals of  $\text{Hg}_2\text{Cl}_2$  by the method of vapor crystal growth under terrestrial and microgravity environments are numerically investigated. By examining plots of the distributions of vector and stream function, and temperature and concentration, an appreciation for the nature of the diffusive-convection and/or diffusive flow and the temperature, and the concentration fields can be obtained. Figures 5 and 6 show (a) velocity, (b) streamline, (c) temperature, (d) concentration field, based on  $\text{Ar} = 5$ ,  $\Delta T = 60$  K (623.15 K  $\rightarrow$  563.15 K), corresponding to  $1g_0$  and  $10^{-3}g_0$ , respectively. In Figure 5, a relative velocity vector with a magnitude of 100 has 1.4 cm and the maximum velocity vector is  $0.35 \text{ cm s}^{-1}$ , whereas in Figure 6, a relative velocity vector with a magnitude of 100 has 1 cm and the maximum velocity vector is  $0.033 \text{ cm s}^{-1}$ . As shown in Figure 5, all fields (5(a)-5(d)) are asymmetrical about the centerline axis of  $y = 1$  cm and exhibit a three dimensional flow structure with one single cell in the vapor phase. To study the effects of gravitational acceleration on flow fields of vector, stream function, temperature and concentration, a  $10^{-3}g_0$  of micro-

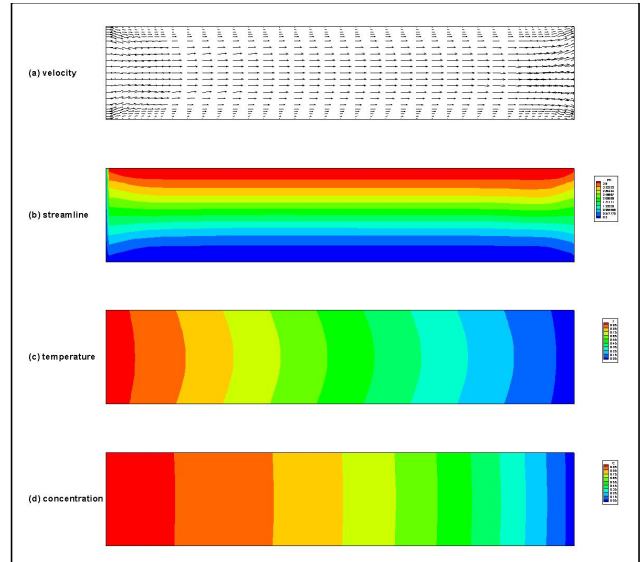


Figure 6. (a) velocity, (b) streamline ( $\Delta\psi = 0.37$ ,  $\psi_{\max} = 3.6$ ,  $\psi_{\min} = 0.2$ ), (c) temperature ( $\Delta T = 0.1$ ,  $T_{\max} = 1.0$ ,  $T_{\min} = 0$ ), (d) concentration ( $\Delta C = 0.1$ ,  $C_{\max} = 1.0$ ,  $C_{\min} = 0$ ), based on aspect ratio = 5,  $\Delta T = 60$  K (623.15 K  $\rightarrow$  563.15 K), and  $10^{-3}g_0$ . A relative velocity vector with a magnitude of 100 has 1 cm. The maximum velocity vector is  $0.033 \text{ cm s}^{-1}$ .

gravity acceleration is chosen, under whose condition it would increase the diffusive flow and yield one-dimensional parabolic flow pattern in the vapor phase. This indicates thermal and/or solutal buoyancy driven convection is very weak under  $10^{-3}g_0$  of microgravity, it is impossible for the effect of buoyancy force to contribute significantly to heat and/or mass transport. Thus, our results at the  $10^{-3}g_0$  of microgravity show that both temperature and concentration gradients become uniform near the crystal region, which is an ideal growth condition for the high quality of crystal. For the simulations shown in Figures 5 and 6, we choose a field strength of  $P_B = 100$  Torr for the cases considered here.

As shown in Figure 5(a), the magnitudes of velocity vector near the crystal region are shown to be greater than those near the source interface. The magnitudes of velocity vector in the lower half region are nearly same those in the upper half region along the centerline, dimensional  $y = 1$  cm, which indicates a strong convective structure under operating conditions of  $\Delta T = 60$  K (623.15 K  $\rightarrow$  563.15 K), and  $1g_0$ ,  $P_B = 100$  Torr, aspect ratio (transport length-to-width) = 5.0, Prandtl number = 1.1, Lewis number = 0.7, thermal Grashof number =  $1.0 \times 10^5$ , solutal Grashof number =  $4.7 \times 10^5$ , thermal diffusivity of  $0.07 \text{ cm}^2 \text{ s}^{-1}$ , kinematic viscosity of  $0.08 \text{ cm}^2 \text{ s}^{-1}$ , binary diffusivity of  $0.10 \text{ cm}^2 \text{ s}^{-1}$ . The temperature profile along the centerline of dimensional  $y = 1$  cm exhibits a cosine wave curve, indicating a uniform imposed temperature profile on ampoule walls. As shown in Figure 5(d), with regards to concentration profile, the concentrations in front of the source interface are relatively uniform, and significantly varied near the crystal interface. As shown in Figure 6, all fields (6(a)-6(d)) are symmetrical about the centerline axis of  $y = 1$  cm and exhibit a one-dimensional flow structure, diffusion-dominant in comparison with the

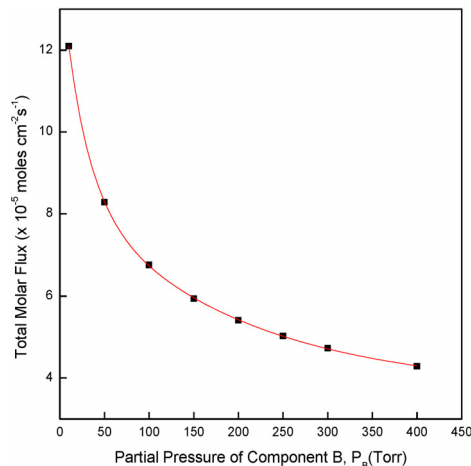


Figure 7. Effects of partial pressure of component B,  $P_B$ (Torr) on the total molar flux for various partial pressure of component B,  $P_B$ ,  $10 \leq P_B(\text{Torr}) \leq 400$ .

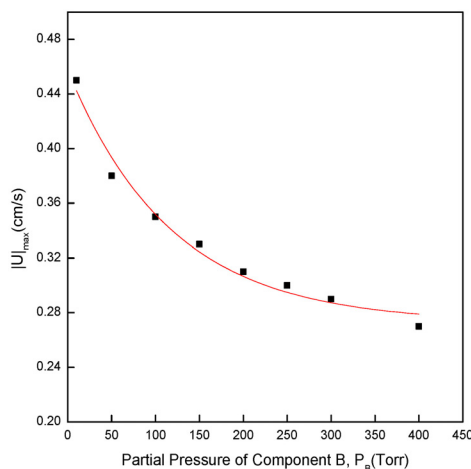


Figure 8. Effects of partial pressure of component B,  $P_B$ (Torr) on  $|U|_{\max}$  for various partial pressure of component B,  $P_B$ ,  $10 \leq P_B(\text{Torr}) \leq 400$ , corresponding to Figure 7.

effect of convection.

One of the practical methods for controlling the crystal growth rate is to increase the partial pressure of the component B as an impurity. Thus, we investigate the influences of the impurity on diffusive-convection flow fields by physical vapor transport. Figure 7 shows the effects of partial pressure of component B,  $P_B$  on the total molar flux for various partial pressure of component B,  $10 \leq P_B(\text{Torr}) \leq 400$ . The total molar flux decays second-order exponentially with increasing the partial pressure of component B, for  $10 \leq P_B(\text{Torr}) \leq 400$ . For  $10 \leq P_B(\text{Torr}) \leq 100$ , the total molar flux falls sharply, and for  $10 \leq P_B(\text{Torr}) \leq 400$ , the total molar flux is nearly linearly decreased with the partial pressure of component B. The total molar flux at  $P_B = 10$  Torr is nearly doubled than that at  $P_B = 150$  Torr, with decreasing the partial pressure of component B by a factor of one-seventh. Figure 8 shows the effects of partial pressure of component B,  $P_B$ (Torr) on  $|U|_{\max}$  for various partial pressure of component B,  $P_B$ ,  $10 \leq P_B(\text{Torr}) \leq 400$ , corresponding to Figure 7. The  $|U|_{\max}$  decays sec-

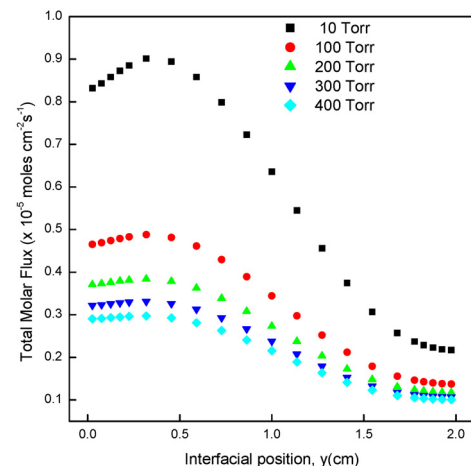


Figure 9. Interfacial distributions of the total molar flux for five partial pressures of component B,  $P_B$ (Torr).

ond-order exponentially with increasing the partial pressure of component B,  $P_B$ , for  $10 \leq P_B(\text{Torr}) \leq 400$ . The  $|U|_{\max}$  decreases with partial pressure of component B,  $P_B$  by a gradient of  $8.6 \times 10^{-4} \text{ cm s}^{-1} \text{ Torr}^{-1}$  for  $10 \leq P_B(\text{Torr}) \leq 150$ . The  $|U|_{\max}$  decreases directly and linearly with partial pressure of component B,  $P_B$  by a gradient of  $2.4 \times 10^{-4} \text{ cm s}^{-1} \text{ Torr}^{-1}$  for  $150 \leq P_B(\text{Torr}) \leq 400$ . The gradient in the range of  $10 \leq P_B(\text{Torr}) \leq 150$  is found to be 3.58 times than that in the range of  $150 \leq P_B(\text{Torr}) \leq 400$ .

Figure 9 shows the interfacial distributions of the total molar flux for five partial pressures of component B,  $P_B$  to study non-uniformities against the center of  $y = 1$  cm. The convection enhances the overall mass transport of component A ( $\text{Hg}_2\text{Cl}_2$ ), and, then results in an increase in its total molar flux, but pays for the expense of uniformity in the total molar flux, with the specific interfacial distribution revealing dominance of diffusive convection. This result is consistent with earlier results[30-31]. The total molar fluxes for all cases under consideration, as shown in Figure 9, imply the occurrence of one cell in front of the crystal region, and the results for  $P_B = 100$  Torr, and  $1g_0$  are found to be confirmed, as shown in Figure 3. All profiles of the total molar flux against dimensional interfacial position are asymmetric against the center position of  $y = 1$  cm. Moreover, the maximum value of the total molar flux for all cases occurs between  $y = 0$  cm and  $y = 0.5$  cm, the maximum value of the total molar flux for the case of  $P_B = 10$  Torr is about twice that for  $P_B = 100$  Torr. It should be noted that fluid is assumed to cling to solid surfaces, i.e., walls with which it is in contact. In other words, the velocities of the mixture of vapor A and B at walls, i.e.,  $y = 0$  cm and  $2$  cm, are assumed to be zero. As shown in Figures 7 through 9, our results have indicated that by increasing the partial pressure of component B, the solutal effect is found to have the stabilizing effect of diffusive-convection flow and, thus, the maximum total molar flux near at  $y = 0.5$  cm is decreased.

#### 4. Conclusions

For the study of the influences of impurity on diffusive-convection flow fields by physical vapor transport of  $\text{Hg}_2\text{Cl}_2$  under terrestrial and microgravity conditions, we carried out numerical simulations. It is concluded that the diffusive convection flow fields, total molar flux at the crystal regions are much sensitive to the gravitational accelerations, partial pressure of component of B. Under microgravity conditions below  $10^{-3}g_0$ , the flow fields are found to be a one-dimensional parabolic flow structure which implies a diffusion-dominant mode. For the convection region of  $10^{-1}g_0 \leq g \leq 1g_0$ , one cell in the vapor phase occurs for all cases under consideration of the total molar fluxes for  $10 \leq P_B(\text{Torr}) \leq 400$ . Also, at the gravitational level of  $10^{-3}g_0$ , the effect of convection is likely to be negligible.

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