염화제일수은 승화법 단결정 성장 공정에서의 대류 현상 연구

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Effects of Convective Flow Fields on the Physical Vapor Transport Processes of Hg_2Cl_2 Crystals

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1. Introduction

Mercurous chloride (Hg_2Cl_2) has many advantages in its applications to acousto-optic, and opto-electronic devices because it has the unique properties of a broad transmission range, well into the far infra-red, a low acoustic velocity, a large birefringence, and a high acousto-optic figure of merit [1]. Hg_2Cl_2 has a high vapor pressure, hence single crystals are usually grown by physical vapor transport (PVT) method in closed silica glass ampoules. We discuss the application of the laser Doppler velocimetry to measure the flow field inside a closed ampoule. The experimental results, are discussed its relationship to computational models and compared to their expectations.

2. Experiments

The crystal growth system employed in our experiment is schematically illustrated in Fig. 1, and includes a furnace, crystal transport ampoule with Hg_2Cl_2 material, and independently controlled temperature controllers. A two zone tubular resistance furnace with a 30 mm inner diameter quartz tube is modified slightly for our laser experiments with a 15 mm Kanthal heating wire spacing between each successive turn of the wire, as shown in Fig. 2. The ampoule is placed into the two-zone furnace with the bottom zone of the furnace at a higher temperature than the top zone. The bottom zone is kept either at 310 or 320 °C and the top zone at 290 °C. Single crystal growth is performed in the two zone furnace, with the growing crystal located at the top and the source material at the bottom. The ampoules were encapsulated under vacuum, 10^{-6} Torr, after purification [2].

3. Results and discussion

3.1 Experiments

The measurements of the radial velocity profiles for the experimental conditions in Table 1,

Fig. 3 and Fig. 4, show the presence of an asymmetric rolls for a fixed axial location. The velocity profiles corresponding to conditions 1 and 2 were made close to the source material, whereas condition 3 near the crystal interface. All 3 measurements indicate the presence of cellular convection. Further insight in the flow field characteristic was provided by taking a measurement in the perpendicular direction, corresponding to condition 1 in Fig. 3. This shows that these flow fields are complex and three dimensional. Due to the limited measurements taken axially, it is inconclusive whether or not the flow consisted of a single roll or a number of modes.

We measured the local radial temperature profile to establish the thermal boundary conditions which would lead insight in the velocity profile characteristics. The measured thermal field is shown in Fig. 5. This indicates that the furnace imposes a relatively large radial temperature gradient which also varies in the azimuthal direction. There is a correspondence between the radial temperature gradient and the velocity profile. The plane with the largest gradient corresponds to the larger velocity profile. This shows that the thermal boundary condition has an effect on the flow field.

According to the physics of PVT as postulated through the computational models, two sources of convection can be predominant, thermal or thermosolutal. Given that no inert gases were introduced inside the ampoule during preparation, residual gases would not be sufficient to drive solutal convection. We postulate that the source of convection is predominantly thermal. This simply means that solutally, mercurous chloride is advected passively in the flow field; thus the solutal Rayleigh number is zero. In light of the computational models that have predicted the flow field patterns in PVT for various boundary conditions, the flow field trends have given insight to our experimental findings. Even though, these models have been limited to two spatial dimensions, they capture the physics of the problem. In particular, the computational models [3-7] which addresses PVT in horizontal enclosures which is perpendicular to gravity (horizontal gradients) have shown that the predicted velocity profile is asymmetric. Whereas, the 2D model for the vertical cylinder [8], which uses the assumption of axisymmetry has predicted symmetric cells. Despite some of the limitations in boundary conditions (insulated), the models [9,10] which address vertical rectangular geometry parallel to gravity, in which the assumption of axisymmetry is not used, shows that the flow field is asymmetric. The computational models imply that either horizontal or vertical thermal gradients can yield asymmetric cells. In the case of horizontal gradients, there is no threshold for convection. Given the complexity of the thermal field which prescribes the boundary condition for the experiment, we cannot expect any one model to predict these velocity profiles. However, it is encouraging that asymmetric structure as measured through our experiments have been predicted for the vertical case parallel to gravity. One important outcome of the experiment is that it suggests that the assumption of axisymmetry for the vertical cylinders should be lifted for further computational modeling. Thereby the prediction of flow fields would not be limited to axisymmetric flows. Recent computational works [11,12] have also indicated the likelihood of multiple asymmetric modes.

3.2 Comparison with experiments

Fig. 7 shows a comparison of our numerical results with Laser Doppler Velocimetry (LDV) measurements of the radial velocity profile for the same parameters as the experiment at the axial location of x = 2.5 cm from the source interface, see also refs [11,13,14]. For comparison purpose, instead of a radial velocity (V_{old}) for LDV measurements, a transformed radial velocity (V_{tra}) is chosen, which is defined as:

$$V_{tra}\,(x, H\text{-}y) = V_{old}\,(x,y)$$

If one looks at the radial velocity profile by the LDV measurements in Fig.7 associated with the crystal growth ampoule, it is apparent that the radial velocity profiles of Fig. 4 and (c) shown in Fig.7 associated with the transformed velocity V_{tra} are essentially similar, differing only in their orientation in space. The radial velocity profile of Fig. 4 becomes the (c) profile by transforming the velocity profile. The LDV radial velocity profile for cylindrical enclosures corresponds to the numerically computed x-component velocity profiles at the corresponding axial location.

With respect to the velocity magnitude, the LDV has the same order of magnitude as the case 15-I (Ra = 1.04 x 104, Pe = 3.33, Pr = 1.25, Le = 0.58, Cv = 1.04, Ar = 0.124) with insulating walls and one order of magnitude larger than the case 15 (Ra = 1.04 x 104, Pe = 3.33, Pr = 1.25, Le = 0.58, Cv = 1.04, Ar = 0.124) with conducting walls, see ref. [11] for details. Note that the flow field characteristics in physical vapor transport is a function of the following parameters V = f (Ra, Ar, Pe, Pr, Le, Cv) [11,13]. The governing parameters for the LDV shown in Fig. 4 are slightly different from the cases 15 and 15-I [11] because of their adoptions of different total vapor pressure in the PVT process. Even though the conducting wall boundary condition is more realistic than insulating, better agreement is obtained with the insulating wall boundary condition. Note that the two-dimensional numerical model predicts that the convective flow is asymmetric which is in agreement with experiments. The numerical computations show that two-dimensional models can be useful in predicting trends in experiments in which three-dimensional effects are unlikely to exist.

4. Conclusions

The LDV system provides velocity profiles inside a PVT crystal growth system. The laser Doppler velocimetry measurements show that for a fixed axial location, the radial velocity profiles are asymmetric. The magnitude of the velocity profiles range from 0 to 4 cm/sec. The measurement of the velocity profile taken at a perpendicular plane, azimuthal variation, indicates that the flow field is three dimensional. In an actual experiment the measurement of the radial velocity profile at a fixed axial location does not imply its behavior over the entire field. The axial plane of the flow should be scanned to verify the number of asymmetric modes. In spite of the approximation of two-dimensional model, the insulating boundary condition predicts the correct trend of the experimentally measured velocity profile in both magnitude and planform.

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Table 1 Experimental conditions

Ampoule condition	1 (0°)	1 (90°)	2	3
Vertical Rayleigh number, Ra	5356	5356	9874	6120
Horizontal Rayleigh number, Ra_h	4552	2410	2962	2754
Aspect ratio, Ar	0.12	0.12	0.12	0.13
Peclet number, Pe	3.03	3.03	3.81	3.03
Prandtl number, Pr	1.2	1.2	1.2	1.2
Lewis number, Le	0.46	0.46	0.46	0.46
Concentration parameter, C_v	0.05	0.05	0.02	0.05
Diameter, D (mm)	22	22	22	23
Transport length, L (mm)	177	177	177	180
Materials	Medium purity Hg_2Cl_2			
Geometry	Parallel to the gravity vector			
Source temperature, T_s (°C)	310	310	320	310
Crystal temperature, T_c (°C)	290	290	290	290
Distance from source to laser positions, L_m (mm)	28	28	25	170
Distance from source to temperature gradient, L_0 (mm)	5	5	88	152

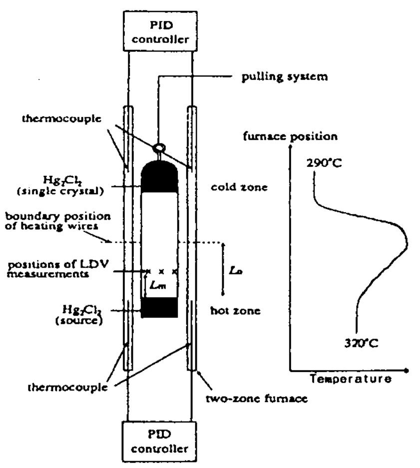

Fig. 1. A two-zone crystal growth system.

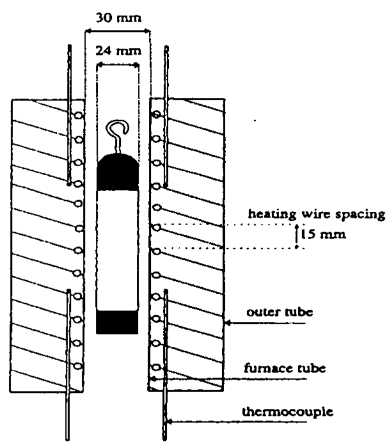

Fig. 2. Illustration of a modified two-zone furnace.

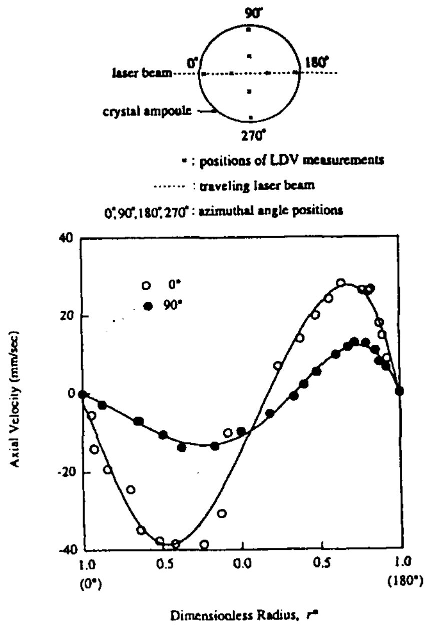

Fig. 3. Velocity profiles taken at 0° (0°–180°, $Ra_h = 4552$) and 90° (90°–270°, $Ra_h = 2410$) for ampoule condition 1 ($Ra = 5356$).

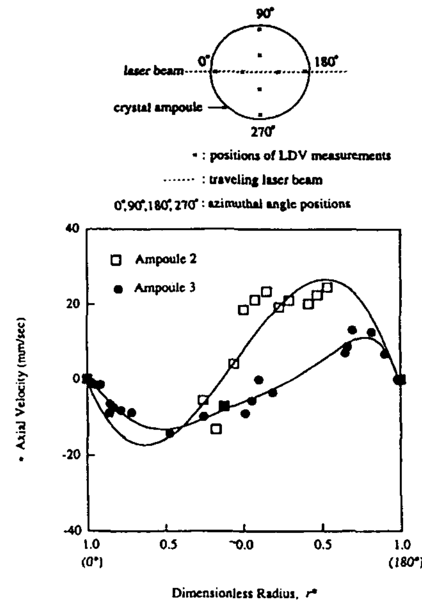

Fig. 4. Velocity profiles along the radial direction for ampoule condition 2 ($Ra = 9874$, $Ra_h = 2962$) and condition 3 ($Ra = 6120$, $Ra_h = 2754$).

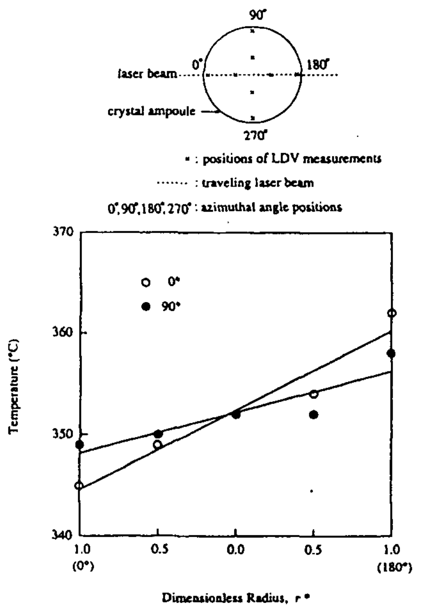

Fig. 5. Radial temperature profiles taken at 0° (0°–180°, $Ra_h = 4552$) and 90° (90°–270°, $Ra_h = 2410$) for ampoule condition 1 ($Ra = 5356$). 90° (90°–270°, $Ra_h = 2410$) for ampoule condition 1 ($Ra = 5356$).

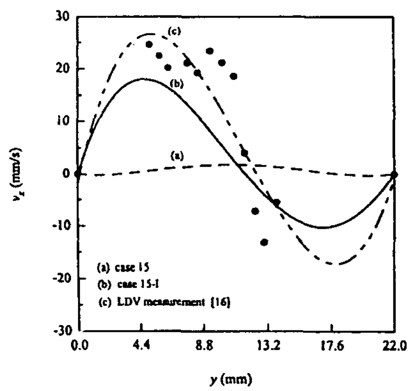

Fig 6. Comparison of numerically computed (cases 15 and 15-1) and experimentally (laser Doppler velocimetry) measured radial velocity profiles at fixed axial location $x = 2.5$ cm.