wall, its temperature rises and the heat transfer decreases. As the aspect ratio increases, the convective effect promotes greater heat-transfer rates for most of the height of the enclosure.

The average Nusselt number for the side wall is calculated according to the definition indicated in Huang and Hsieh.[8] It was suggested by Arpaci and Larsen[10] that the present $\overline{Nu_s}$ be represented in the form of

$$\overline{Nu_s} = 0.77(Ra)^{0.212}(Pr)^{0.291}(H/R)^{0.9}$$

for

$$0.5 \leq (H/R)2.0$$

$$10^4 \leq Ra \leq 10^6$$

$$100 \leq Pr \leq 200 \qquad (14)$$

to approximate the computed data within ±5% by least-squares curve fitting. It appears that this approximation agrees quite well with the results reported by Arpaci and Larsen[10] based on the two-length natural convection model and the scale analysis of the present configurations $\overline{Nu_s} \sim (H/R)^{-1}\ R^{0.25}\ Pr^{0.25})$. This indicates that the heat-transfer model used in the scale analysis accurately reflects the phenomenon inside the cylindrical enclosures.

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Evaluation of Transport Conditions During Physical Vapor Transport Growth of Opto-Electronic Crystals

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Nomenclature

a = radius of growth tube, cm
c = cold zone
D = diffusion coefficient, cm^2/S
H = hot zone
J = mass flux, $moles/cm^2.S$
L = transport length, cm
M = molecular weight, g/mole
N_e = dimensionless Peclet number
P = vapor pressure, Torr
$\hat{P}$ = average pressure, Torr
R = gas constant, atm/mole K
T = temperature, K
$\hat{T}$ = average temperature, K
v = growth velocity, cm/S
Ω = molar volume, cm^3

Introduction

MERCUROUS halides show great promise for acousto-optic devices applied to signal-processing and optical spectrum-analyzing systems, and have attractive properties for high-performance devices. These halides have 1) a large transmission range, 2) high acousto-optic figure of merit, 3) suitable photoelastic coefficients, and 4) very slow acoustic velocity. During the last few years, we have investigated[1-4] growth anisotropy, the effect of growth parameters on optical quality, and the effect of crystal quality on the fabrication and characteristics of mercurous chloride acousto-optic devices. The crystals have been grown in closed tubes by the physical vapor transport (PVT) method. In the ongoing investigation of PVT crystal growth, we are studying the effect of thermal and solutal convection during vapor transport. This Note reports the effect of source temperature on mass flow and the growth rate.

Experimental

Purification of Source Material

The as-supplied source material was listed at 99+% purity. It was sublimed several times in a hermetically sealed tube until water-white material was achieved. The purity was checked by spark-source spectrometry. Source material contains less than 15 ppm total metallic impurities.

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Growth-Rate Measurements

Growth rates were measured by a relaxation technique very similar to that described in Ref. 4. A two zone furnace with each zone controlled independently was utilized. All of the growth-rate measurements were made for the (110) orientation only in order to eliminate the effect of growth-rate anisotropy. Length of crystal was measured as a function of time to derive the growth rate for each thermal setting.

Results and Discussion

According to kinetic theory, flux J of molecules striking a unit area per unit time is given by the Hertz-Knudsen (H-K) equation:

$$J = \frac{\alpha P}{\sqrt{2\pi MRT}} \tag{1}$$

where α is the sticking coefficient of vapor molecules impinging on the crystal surface. This equation is applicable for the physical vapor-deposition growth in which a cerain sink action of the growing surface is assumed. For the physical vapor transport from the high-temperature to the low-temperature zone, the flux can be given[5] as

$$J = J_H - J_c = \frac{\alpha(P_H - P_c)}{\sqrt{2\pi MRT}} \tag{2}$$

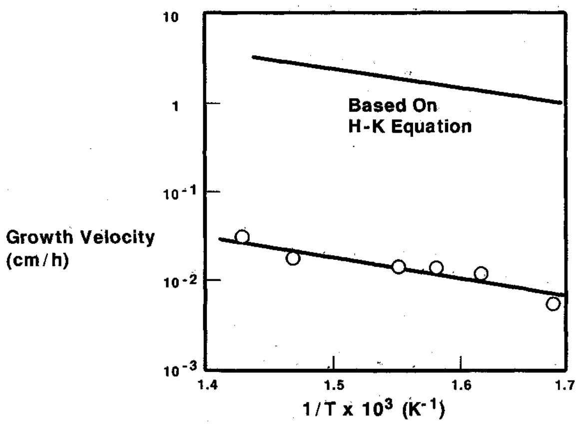

Fig. 1 Comparison of theoretical (H-K) and the experimental growth velocities.

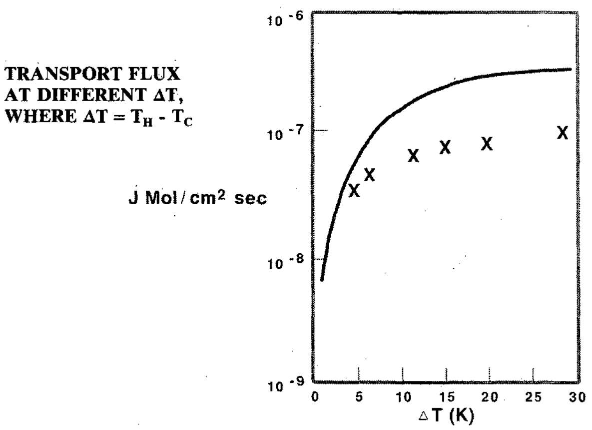

Fig. 2 Comparison of theoretical (one-dimensional model) and experimental mass-transport rate.

where J_H and J_c are the flux from the hot and cold temperature zone, and P_H and P_c denote the vapor pressures at the hot and cold zone. Figure 1 shows the Arrhenius plot of the experimental growth rates and the theoretical curve calculated from the H-K equation. In the calculation, we used the vapor pressure quoted from Ref. 6. The experimental line corresponds to the calculated values corrected by the sticking coefficient of 0.01. Since all of the growth-rate measurements were made in the (110) direction and at the identical aspect ratio (radius 0.55 cm, transport length 10 cm), the change in growth rate was attributed to thermal conditions only. Details of the comparison with theories based on other mechanisms are discussed elsewhere.[4] We have observed growth rates of orders of magnitude lower than the predicted values. This again suggests that disparity can be accounted for by the sticking coefficient.

Experimental data were used to examine the validity of the one-dimensional diffusive physical vapor transport model. This model is based on Stefan flow, which describes the movement of vapor in a closed ampoule due to a thermal gradient. The rate of mass transport J can be given by the equation[7]:

$$J = \frac{D\hat{P}}{R\hat{T}L} N_e \tag{3}$$

The value of N_e can be given as

$$N_e = \ln\left[\frac{\hat{P} - P_c}{\hat{P} - P_H}\right] \tag{4}$$

The value of $\hat{P}$ and $\hat{T}$ has been calculated by averaging the pressure and temperatures in a manner similar to that reported in Ref. 8. Figure 2 shows the flux of mercurous chloride vapor at different temperature differences between the source and crystallization zone. Experimental values were derived from the growth-rate measurements as follows:

$$J_{\text{condensing}} = \frac{\pi a^2 v}{\Omega} \tag{5}$$

A comparison between the values predicted and those derived from the experimentally observed growth rates showed that the one-dimensional diffusive model does not fully satisfy the growth rate of mercurous chloride crystals. This indicates the

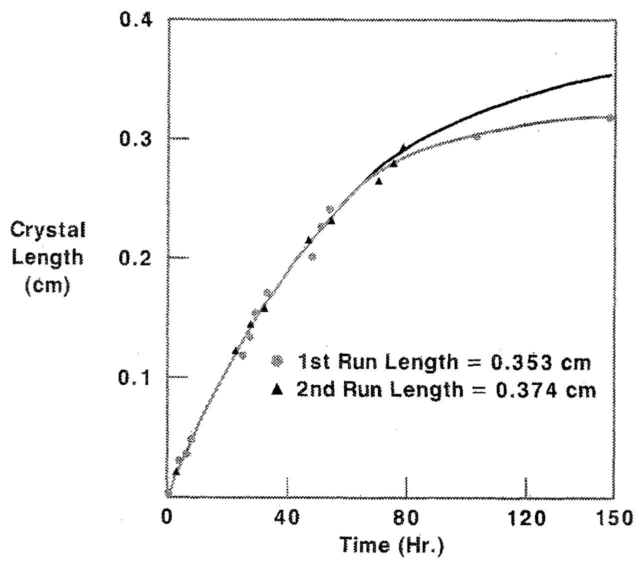

Fig. 3 Length of growing crystal at different time intervals: beginning of growth and end of the growth.

possibility of convecto-diffusive transport, and further studies are in progress.[2] We have extensively studied[9] the effect of aspect ratio and hence Rayleigh number on the growth rate to evaluate the importance of convection during PVT growth.

The role of impurities was studied to examine the contribution of impurities to lower the transport rate. Figure 3 shows a length of growing crystal at different time intervals. Growth rates were measured at the beginning and end of the growth run. If the impurity boundary layers were significant, the initial growth rates should be much higher because of the low concentration of impurity. On the other hand, the growth rate in the latter part of the run should be much lower due to the continuous buildup of impurity boundary layers at the interface. As it is clear from Fig. 3, the growth rates do not change, and we conclude that the effect of the impurity boundary layer is negligible.

Summary

The measured growth rates of mercurous chloride crystals do not fully satisfy the one-dimensional diffusive PVT model, indicating that growth is occurring in the convecto-diffusive range. The significance of the impurity boundary layer was negligible in the present system.

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