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CONVECTIVE FLOWS IN ENCLOSURES WITH VERTICAL TEMPERATURE OR CONCENTRATION GRADIENTS

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Abstract

The transport process in the fluid phase during the growth of a crystal has a profound influence on the structure and quality of the solid phase. In vertical growth techniques the fluid phase is often subjected to vertical temperature and concentration gradients. The main objective of the present work is to obtain more experimental data on convective flows in enclosures with vertical temperature or concentration gradients.

Among actual crystal systems the parameters vary widely. The parametric ranges studied herein for mass transfer experiments are mainly dictated by the electrochemical system employed herein to impose concentration gradients. Temperature or concentration differences are maintained between two horizontal end walls. The other walls are kept insulated.

Experimental measurements and observations were made of the heat transfer or mass transfer, flow patterns, and the mean and fluctuating temperature distribution. For purely thermal cases, the experiment covers the ranges of thermal Rayleigh number $Ra = 9 \times 10^6$ to 2×10^9 , aspect ratio $Ar = 0.65$ to 2.6 , Prandtl number $Pr = 4.0$ to 7.0 . The method used to visualize the flow pattern in the thermal cases is an electrochemical ph-indicator method. For purely solutal cases the experiment covers the ranges of solutal Rayleigh number $Ra_m = 1.0 \times 10^9$ to 2.4×10^{11} , $Ar = 0.68$ to 1.34 , $Sc = 2000$ to 2100 . Laser shadowgraphs is employed to visualize flow patterns in the solutal cases.

The results of the present experiments for the purely thermal cases are adequately correlated by the equation $Nu = 0.0862 Ra^{0.322}$ in the range $9 \times 10^6 < Ra < 2 \times 10^9$. For the purely solutal cases, the mass transfer data can be correlated by the equation $Sh = 0.1326 (Sc Gr_m)^{0.328}$ in the range $1 \times 10^9 < Ra_m < 2.4 \times 10^{11}$.

Nomenclature

Ar aspect ratio ($=d/L$)
 C_b bulk concentration of $CuSO_4$
 C_1 constant of heat transport correlation
 C_2 constant of mass transport correlation
 d height of enclosure

D solutal diffusivity
 F Faraday's constant
 Gr thermal Grashof number
 Gr_m solutal Grashof number
 g gravitational acceleration
 h average mass transfer coefficient
 I current
 i_L limiting current density
 L length of enclosure
 N buoyancy ratio
 Nu Nusselt number
 n exponential constant in transport correlation
 Pr Prandtl number
 Ra thermal Rayleigh number ($=Gr Pr$)
 Ra_m solutal Rayleigh number ($=Gr_m Sc$)
 Sc Schmidt number
 Sh Sherwood number
 T temperature
 T_h hot wall temperature
 T_c cold wall temperature
 t time
 V voltage
 W depth of enclosure
 x, y, z coordinates
 $\bar{x}, \bar{y}, \bar{z}$ dimensionless coordinates
 z^* charge number of copper
 $\bar{\beta}$ volumetric coefficient of solutal expansion
 β volumetric coefficient of thermal expansion
 δ_s thickness of solutal boundary layer

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ν kinematic viscosity

α thermal diffusivity

Introduction

The growth of crystals is a coupled process of heat and mass transfer, fluid flow, phase transformations, and chemical reactions. Although the technology of crystal growth is well grounded in physical chemistry, the treatment of the transport phenomena has been relatively rudimentary and empirical. Recently, it was recognized that convection, as a result of temperature or concentration gradients,¹ occurs in many aspects of materials processing including solidification, oxidation of surface material, and crystal growth.

The rate of crystal growth is often limited by the mass transfer because it is the slowest process. In almost all the commonly used techniques the parent phase is a fluid. As in most nonequilibrium processes involving a fluid, convection plays a dominant role in that it affects the fluid-phase composition and temperature at the phase interface. The fluid motion in many cases is determined by the locally generated motion. The flows are due either to the effects of temperature and/or concentration gradients in a body-force field.² In some crystal-growth techniques, the completely confined fluid phase is subject to vertical temperature and/or concentration gradients. To gain insights on such flows, an experimental program was initiated to study flows in rectangular enclosures with vertical temperature or concentration gradients between horizontal end walls.

The present work for purely thermal cases is intended to give a comprehensive understanding of natural convection heat transfer in enclosures with thermally insulated vertical walls by systematically varying the parameters Ra and Ar . The fluid is tap water in these thermal experiments.

An electrochemical system based on a diffusion-controlled electrode reaction was employed to create the vertical concentration gradients. The apparatus was a rectangular cavity with the horizontal end walls being the electrodes. All other walls were insulated. The fluid was a copper sulphate ($CuSO_4$)-acid (H_2SO_4) solution for purely solutal experiments.

Experimental Design

Dimensionless Parameters

Based on the basic differential equation for natural convection in an enclosure with thermal or solutal buoyancy forces (Fig. 1), it can be shown that the following dimensionless parameters are important in the present work.

Thermal Grashof number, $Gr = g\beta \Delta T d^3/\nu^2$

Prandtl number, $Pr = \nu/\alpha$

Schmidt number, $Sc = \nu/D$

Solutal Grashof number, $Gr_m = g\beta \Delta C d^3/\nu^2$

Aspect ratio, $A = d/L$

With the present system the ranges of the dimensionless parameters covered for purely thermal cases are $Pr = 4.0$ to 7.0 , $Gr = 1.3 \times 10^6$ to 5.0×10^8 , $Ar = 0.65$ to 2.6 , and for purely solutal cases are $Sc = 2000$ to 2100 , $Gr_m = 5.0 \times 10^5$ to

1.2×10^8 , $Ar = 0.68$ to 1.34 . All fluid properties are evaluated at the average temperature of the hot and cold walls in the thermal cases. In the solutal cases fluid properties are evaluated at the average concentration of the bulk solution.

Test Apparatus

A sketch of the experimental system is presented in Fig. 1. The test cell is a rectangular enclosure formed with four insulating plexiglas plates and two copper plates (both thermally and electrically conductive). The width, L , is 7.7 cm or 8.2 cm and the height is variable so that a range of aspect ratios can be covered. The depth of the enclosure is relatively large (23.9 cm or 21.2 cm). The two copper horizontal end walls are 0.7 cm thick.

For the purely thermal cases, an electrical heating mat is bonded to the back of the bottom copper plate and a circulating water system cools the top copper plate (Fig. 2(a)), so that a uniform temperature can be imposed on the top surface. The plexiglas surfaces are wrapped with 5 cm thick fiberglass insulation to ensure an adiabatic boundary condition. There are six thermocouples embedded in each copper plate to measure the uniformity of the temperature over the entire surface. A thermocouple probe with a two-dimensional traversing mechanism is inserted into the test section to measure the temperature distribution inside the enclosure. A Leeds and Northrup millivolt potentiometer is used to measure the thermoelectric emf from the copper-constantan thermocouples.

The test field is a copper-sulphate-acid solution ($CuSO_4 + H_2SO_4 + H_2O$) for the purely solutal cases. When a voltage is applied to the electrodes, copper dissolves into the solution at the anode and is deposited at the cathode (Fig. 2(b)). As a result, the density of the fluid near the cathode (anode) becomes lower (higher) than that of the bulk of the solution. The migration of cupric ions in the electric field is eliminated by adding sulphuric acid to the solution, which acts as a supporting electrolyte, and thus the transport of the cupric ions is controlled only by diffusion and convection. The equipment for measuring the current and potential are shown schematically in Fig. 2. A dc power supply was used in the tests as the power source to the test cell and two digital multimeters are necessary to obtain values of current corresponding to each potential setting. A potential recorder was used to record the voltage as a function of time.

Test Procedure

The method used to visualize the flow patterns in the present work for purely thermal cases is an electrochemical method called the ph-indicator method.⁴ For this test thymol blue is added to the water in the container to produce 0.01 percent by weight solution, and then the ph of the solution is adjusted to place it on the yellow side of the end-point. Two electrodes are placed in the solution. When a dc voltage (about 6 V in the present experiment) is applied between the electrodes, a small volume of colored solution forms around one of the electrodes and moves from the electrode with the fluid motion forming a neutrally buoyant marker. The color change occurs

because the color of the basic form of thymol blue (blue) is different from that of the acid form (yellow).

Although the temperature of the copper walls can be easily measured by thermocouples, the concentration levels at the walls can not be so easily determined. One relatively simple way to set the concentration level at the cathode in the present system is to adjust the cell potential in such a way that the saturation (limiting) current is obtained. Under the limiting current condition the ion concentration at the cathode surface is zero. In other words, the change in concentration across the solutal boundary layer along the cathode ($\Delta C_{\text{cathode}}$) is C_b . The limiting-current condition is valid at the cathode but not along the anode surface. However, since the net mass fluxes at the cathode and anode are considered to be equal and since the concentration outside the solutal boundary layers of both walls is C_b , it is reasonable to expect that the average change in concentration across the solutal boundary layer along the anode (ΔC_{anode}) is nearly equal to $\Delta C_{\text{cathode}}$. Then, the overall concentration difference across the cell is $\Delta C = \Delta C_{\text{cathode}} + \Delta C_{\text{anode}} = 2C_b$ and thus Gr_m is defined based on $\Delta C = 2C_b$ in the present work. The above concept assumes that C_b remains constant, but as convection develops in the cell the concentration level remains constant only on the average. Moreover, due to electrolysis the copper wall surfaces become rough slowly with time, which causes the current densities at the walls to decrease and the cathode deviates gradually from the limiting-current condition. For this reason, the duration of each run for the pure solutal cases is limited to 2 hr, far shorter than the time required to attain steady solutal convection. It should thus be evident that the concentration boundary conditions are not as well-defined as the thermal conditions. After each run the electrodes are carefully cleaned and polished.

The cell voltage is increased stepwise by a manual control and the corresponding total current is read after each steady state is attained. This procedure is repeated until a potential-current plateau is obtained. The potential stepping rate is chosen as 20 mV/40 sec. The flow pattern and the concentration distributions are visually observed. Since the density of the solution changes with cupric ion concentration a qualitative picture of the flow configuration is obtained from the shadowgraph techniques.

Experimental Results

Purely Thermal Cases

The flow structures were studied by the aforementioned ph-indicator technique. Based on these observations the flow structures are shown in Figs. 3 to 5. "Thermals" rising from the bottom of the test cell and falling from the top of the test cell are evident. The dominant motion in the test cell is that of the thermals.^{5,6}

Figure 6(a) and (b) shows typical mean temperature profiles. The mean temperature distributions are presented in the dimensionless form. The magnitude of the temperature, $T_z - T_c$, is normalized by the temperature difference, ΔT , across the layer. The subscript z denotes a condition at a

distance z from the bottom plate. It was found that the temperature changed sharply near the top and bottom plates and the temperature distribution of the bulk flow was uniform. To record temperature disturbances induced by the thermals, the thermocouple probe was placed as near as possible to the bottom and top surfaces of the test cell. Time series records of the thermocouple output are shown in Figs. 7 and 8. According to Figs. 7 and 8 the amplitudes of the temperature oscillations increase near the top and bottom plates.

The Nusselt number data for water ($Pr = 5.5$ to 7.0) for various values of Ar are presented in Fig. 9. The predictions of Nu by others^{5,6} are also shown in this figure for comparison. Nu was found to be proportional to $Ra^{0.322}$ as seen in Fig. 9. The value of the exponent ($n = 0.322$) points to a heat-transfer mechanism by turbulent eddies. However, there appears to be a small but noticeable difference between the present data ($Hr \equiv L/W = 3.22$) and others' data ($Hr = 1$). Nu seems to increase slightly with decreasing Hr (L/W). This dependence of Nu upon Hr may be explained if one considers the probably weak horizontal flow motion which can be detected in Figs. 3 and 4.

Purely Solutal Cases

In the present electrochemical system operated under isothermal conditions the deposition of cupric ions on the cathode wall leaves behind less dense fluid. The lighter fluid rises toward the top of the test section. The downward flow in the boundary layer near the anode feeds heavier fluid toward the bottom wall.⁷ Simple shadowgraph studies (Figs. 10 and 11) suggested a very complex flow structure over the $Sc Gr_m$ range 1×10^9 to 2.4×10^{11} .

It may be useful to compare in detail the purely solutal convection flow structure with that for purely thermal convection because the overall flow structures look somewhat similar for both cases. An important fundamental contrast, however, in the flow structures produced by purely solutal convection and purely thermal convection is illustrated in Figs. 11 and 5. In the heat transfer situation there are far fewer convection columns (thermals) than for mass transfer.

The most convenient method for studying rates of mass-transfer by the free convection mechanism is by measuring the maximum rate of the electrode reaction (i.e., limiting current) which may be maintained in the quasi-steady state. With the limiting current per unit area of the cathodes, i_L , known the mass transfer according to Ref. 7 can be calculated as

$$h = \frac{i_L}{z^*} C_b$$

The dimensionless mass transfer rate, the Sherwood number, is defined as

$$Sh = h \frac{d}{D}$$

In the present electrochemical system the diffusion coefficient is very small so a long time is required to obtain truly solutal convection. Thus, the mass transfer rate measured herein is not a

steady-state value. However, the solutal boundary layers should be established on a time scale of δ_s/D , which is on the order of 10 sec and is small compared to the time at which the limiting current is measured. The measured mass-transfer rate is not expected to differ much from the steady-state value. This accuracy is probably acceptable particularly in view of the uncertainty in the concentration boundary conditions. The experimental error in Sh is estimated to be ± 10 percent. A correlation for Sh , as a function of solutal Rayleigh number ($Sc Gr_m$), is presented in Fig. 12.

Summary of Results

The results of the present experiments indicate that for the range of aspect ratios and Grashof (or Rayleigh) numbers covered:

1. The results for the purely thermal cases are adequately correlated by the equation

$$Nu = 0.862 Ra^{0.322}$$

in the range $9 \times 10^6 < Ra < 2 \times 10^9$.

2. Temperature oscillations and the exponent 0.322 found in the purely thermal cases reflect a high degree of flow instability in the present work.

3. For the purely solutal cases the mass transfer data can be correlated by the equation

$$Sh = 0.1326 (Sc Gr_m)^{0.328}$$

in the range $1 \times 10^9 < Ra_m < 2.4 \times 10^{11}$.

In the present work, comparison of the new correlations of mass-transfer data to the results of heat-transfer investigations could serve to give us some enlightenment about the mechanism of the transport process under discussion.

The figures 9 and 12 show clearly that all results are well correlated by the equations:

$$Nu = C_1 Ra^n,$$

$$Sh = C_2 Ra_m^n,$$

from a practical point of view $n = 1/3$ is acceptable as a regression parameter, at least within the experimental Ra (or Ra_m) range.

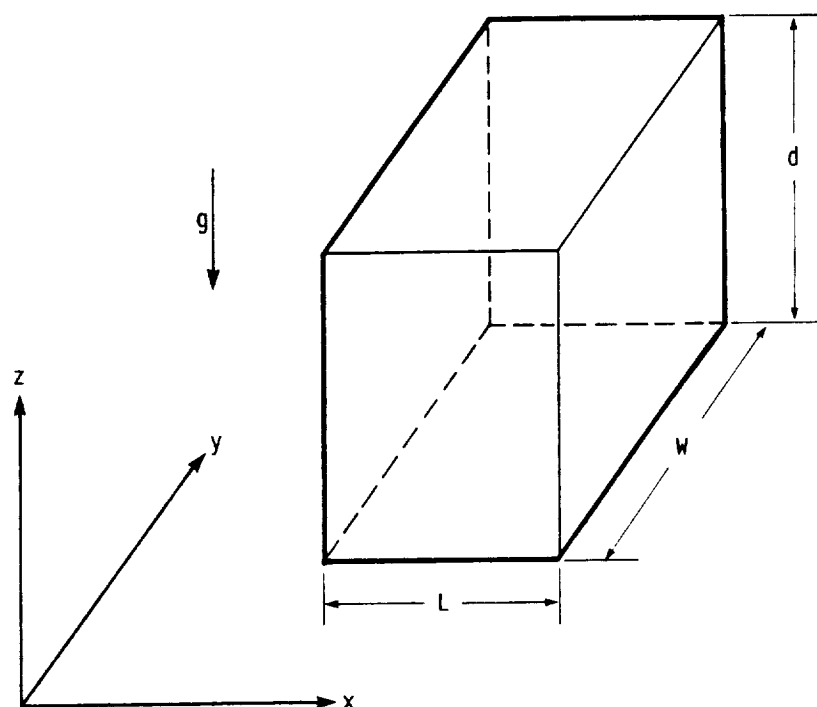
It enables a general understanding of convective flow for all important vertical crystal growth methods.

Acknowledgments

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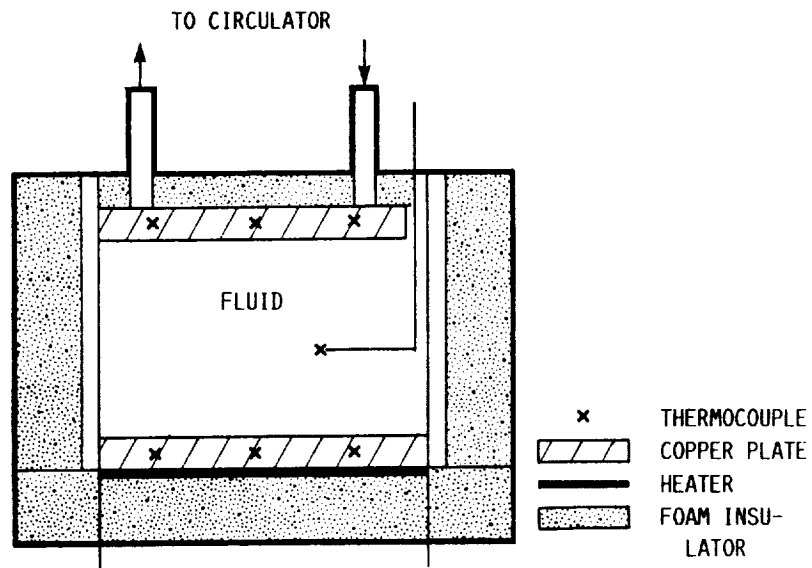


(a) SCHEMATIC.

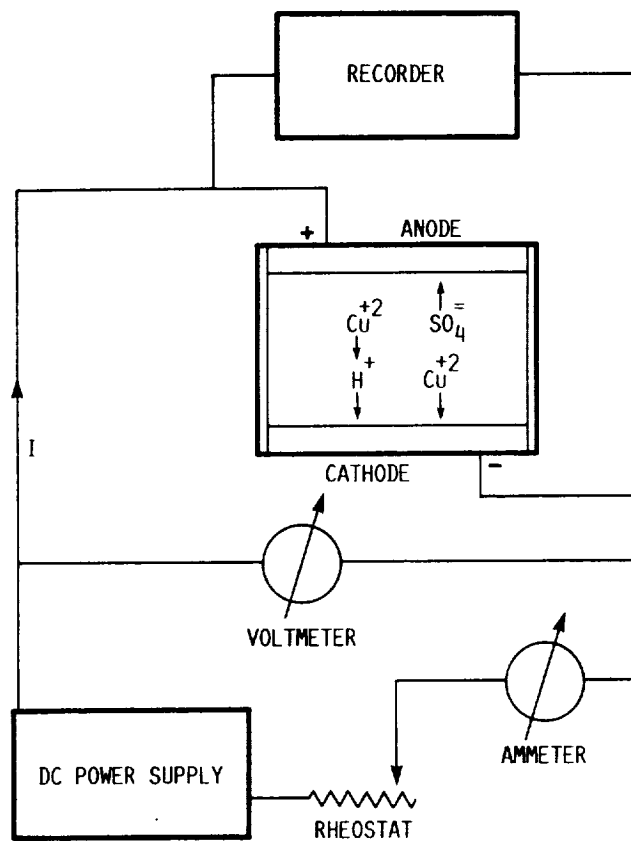
EXPERIMENTAL SYSTEM	L, MM	W, MM	d, MM	Ar
MASS TRANSFER	82	212	56	0.68
			84	1.02
			110	1.34
HEAT TRANSFER	77	239	50	0.65
			100	1.30
			200	2.60

(b) DIMENSIONS.

FIGURE 1. - TEST CELL.



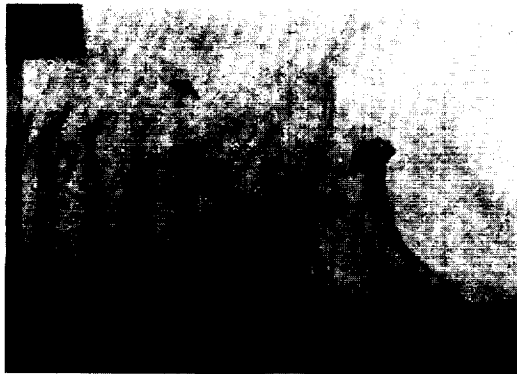
(a) HEAT TRANSFER.



(b) MASS TRANSFER.

FIGURE 2. - EXPERIMENTAL SYSTEMS.

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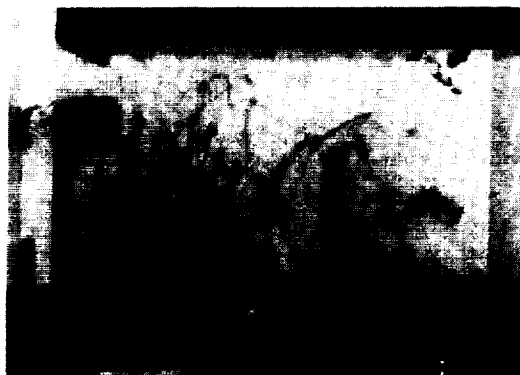
(a) t_1 .



(b) t_2 .



(c) t_3 .



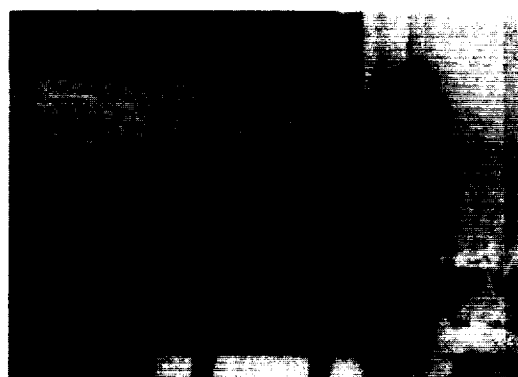
(d) t_4 .

FIGURE 3. - FLOW PATTERNS NEAR THE HEATED LOWER PLATE (PURELY THERMAL CASE) WITH
 $Ar = 0.65$; $Ra = 1.51 \times 10^7$; $t_1 < t_2 < t_3 < t_4$, $\Delta t = 5$ SEC; $\bar{y} = 0$ TO 0.5.

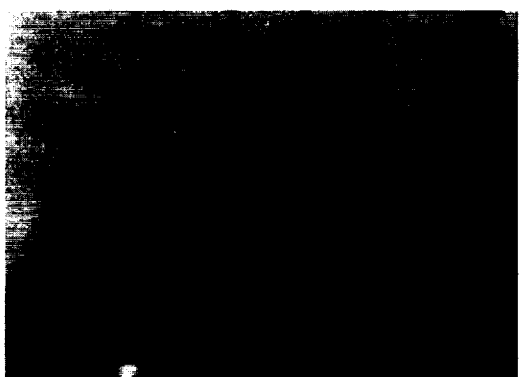
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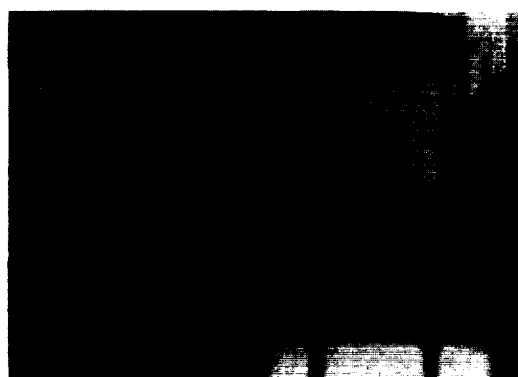
(a) t_1 .



(b) t_2 .



(c) t_3 .



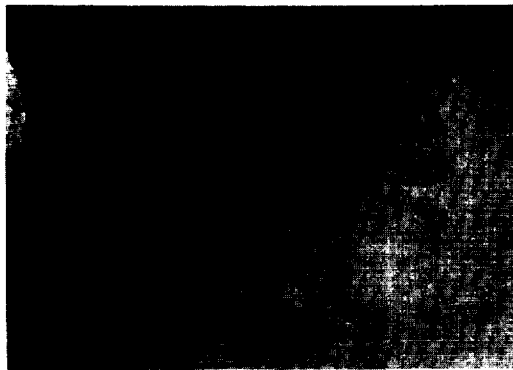
(d) t_4 .

FIGURE 4. - FLOW PATTERNS NEAR THE LOWER PLATE (PURELY THERMAL CASE) WITH $Ar = 0.65$;
 $Ra = 1.51 \times 10^7$; $t_1 < t_2 < t_3 < t_4$; $\Delta t = 5$ SEC; $\bar{y} = 0.6$ TO 1.0.

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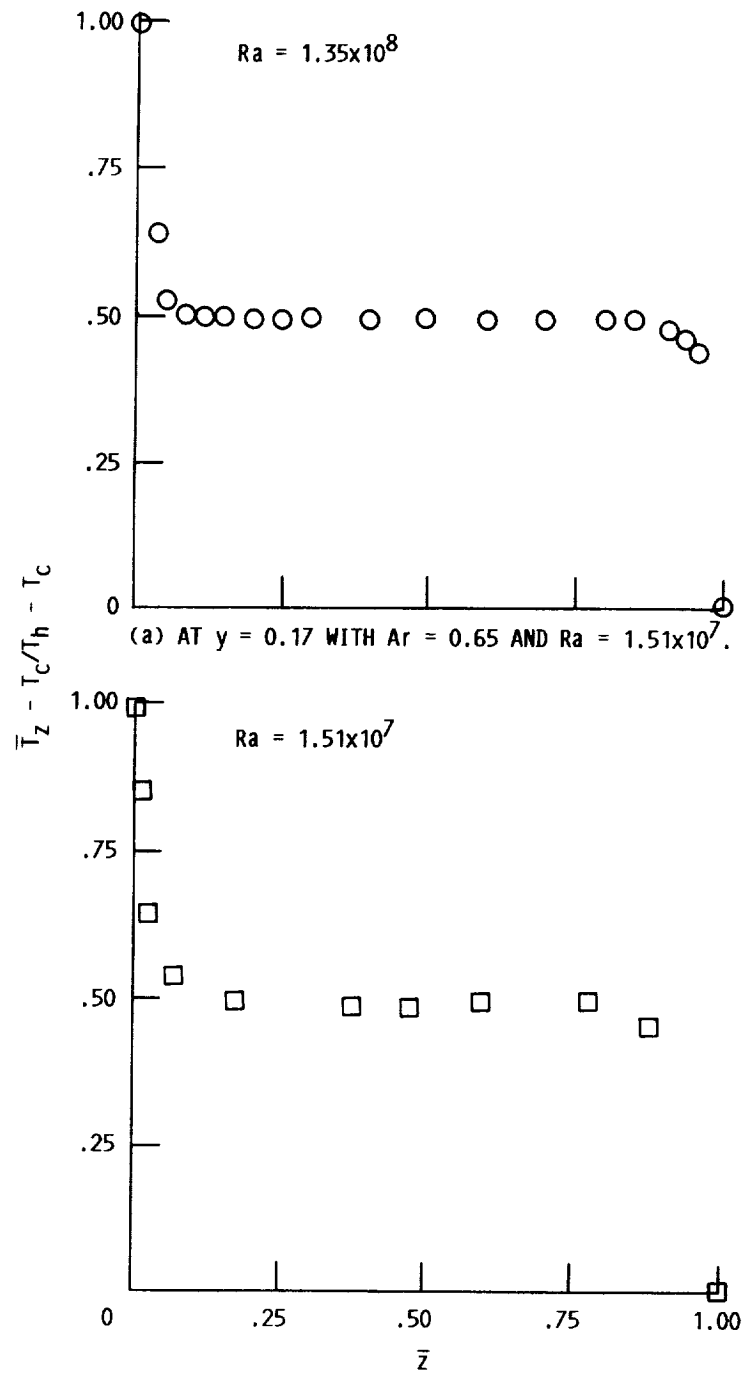


(a) t_1 .



(b) $t_2 = t_1 + 5 \text{ sec.}$

FIGURE 5. - FLOW PATTERNS NEAR THE COLD
TOP PLATE (PURELY THERMAL CASE) WITH
 $Ar = 1.30$; $Ra = 1.35 \times 10^8$; $t_1 < t_2$;
 $\Delta t = 5 \text{ sec.}$



(a) AT $y = 0.17$ WITH $Ar = 0.65$ AND $Ra = 1.51 \times 10^7$.
 (b) AT $y = 0.37$ WITH $Ar = 1.30$ AND $Ra = 1.35 \times 10^8$.
 FIGURE 6. - TEMPERATURE PROFILES AT $x = 0.52$
 FOR PURELY THERMAL CASE.

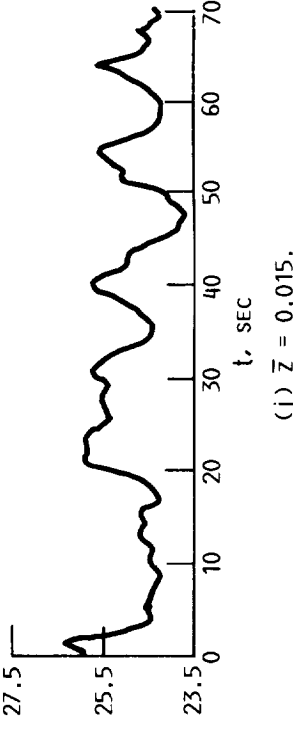
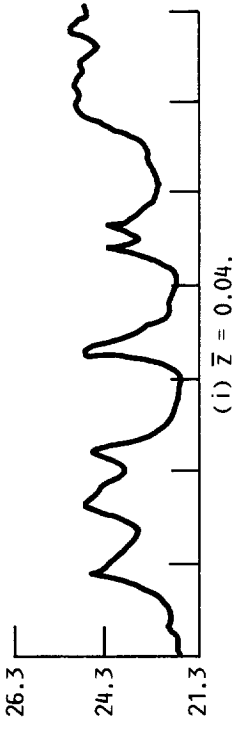
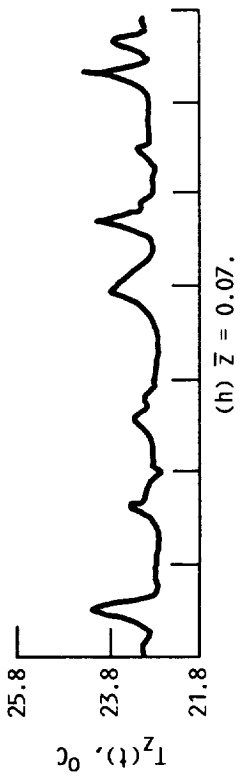
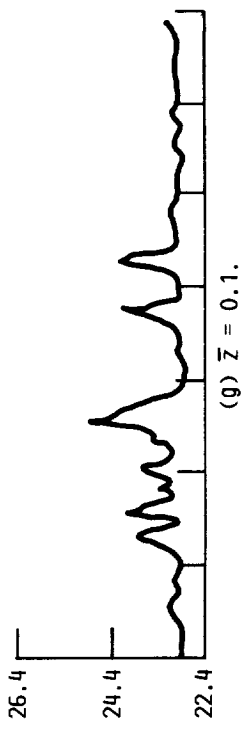
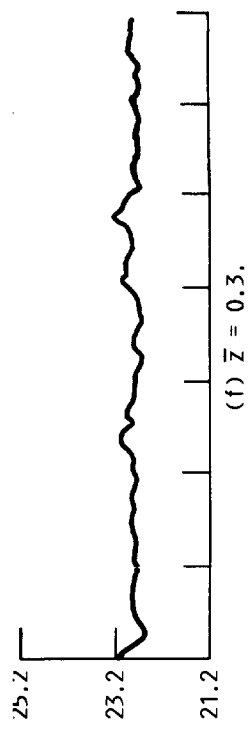
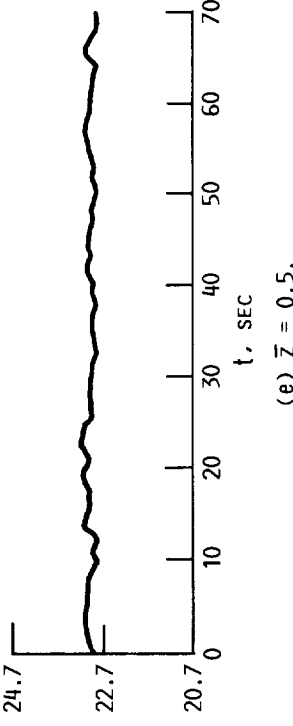
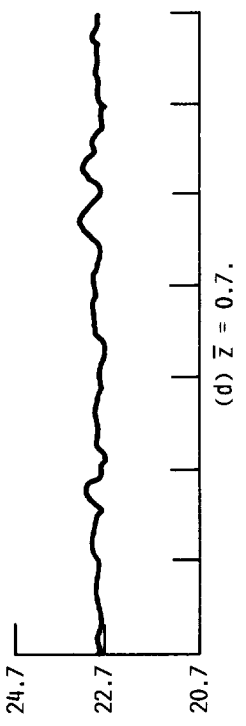
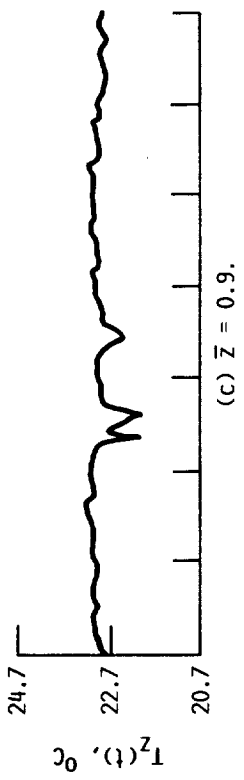
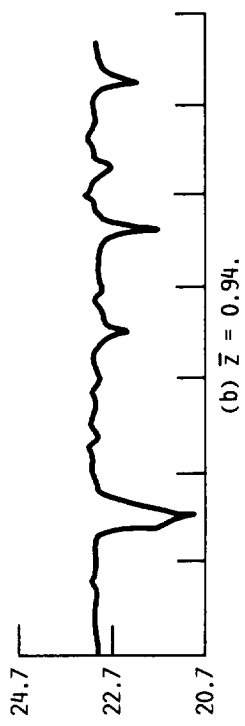
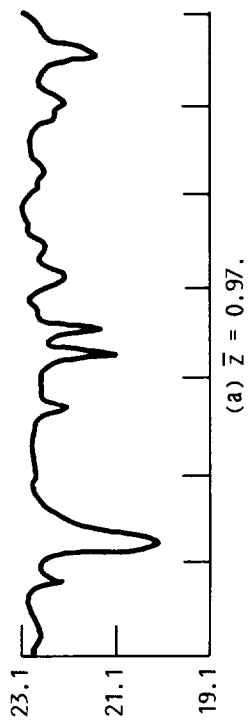


FIGURE 7. - INSTANTANEOUS TEMPERATURE PROFILES AT $x = 0.78$, $y = 0.59$ FOR PURELY THERMAL CASE WITH $Ar = 1.30$, $Ra = 1.35 \times 10^8$, AND $Th = 27.8^\circ C$.

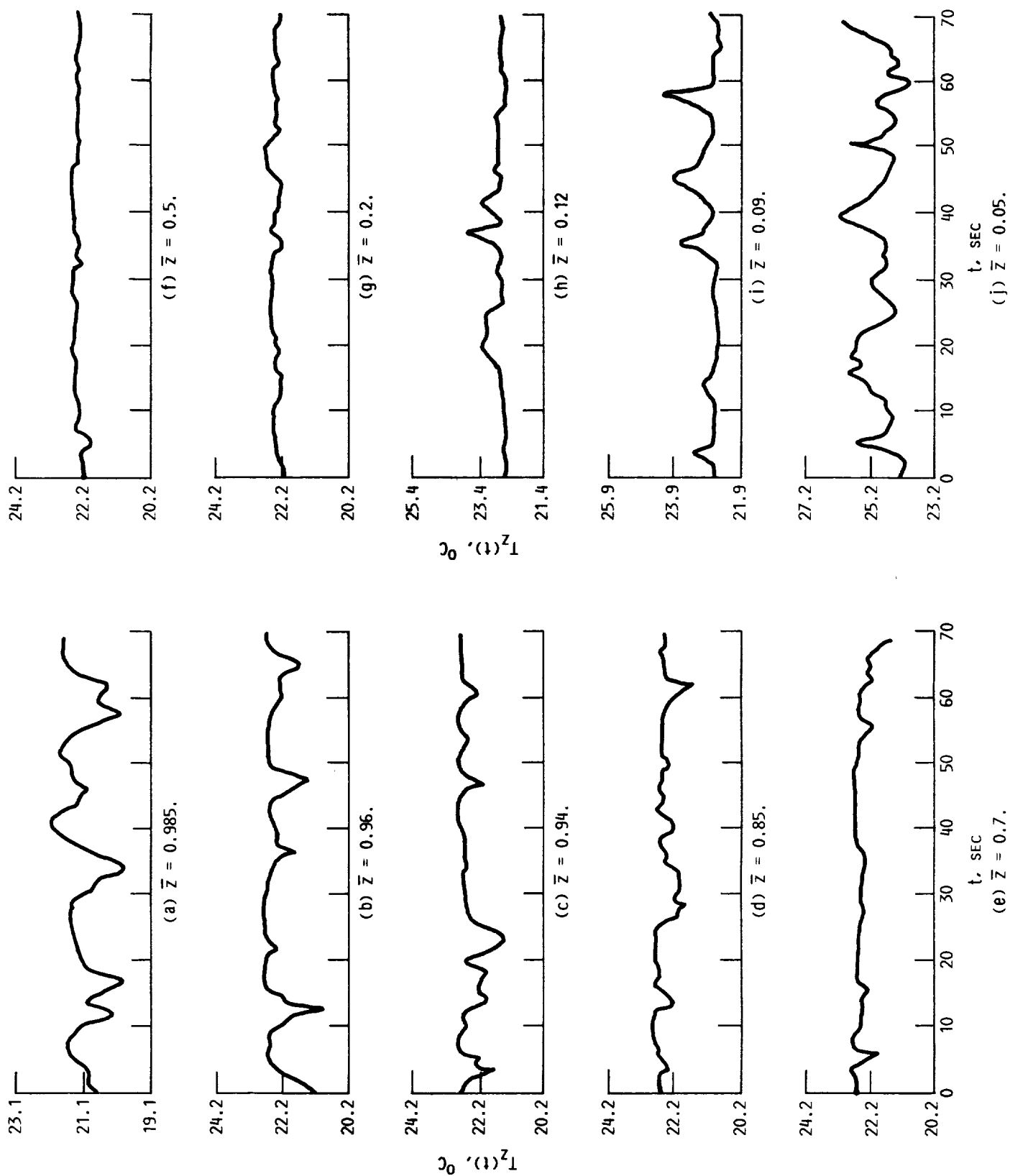


FIGURE 8. - INSTANTANEOUS TEMPERATURE PROFILES AT $x = 0.52$, $y = 0.17$ FOR PURELY THERMAL CASE WITH $Ar = 1.30$, $Ra = 1.35 \times 10^8$.

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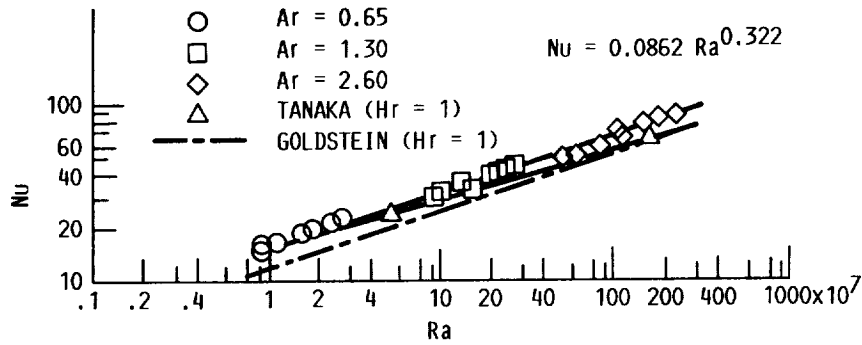
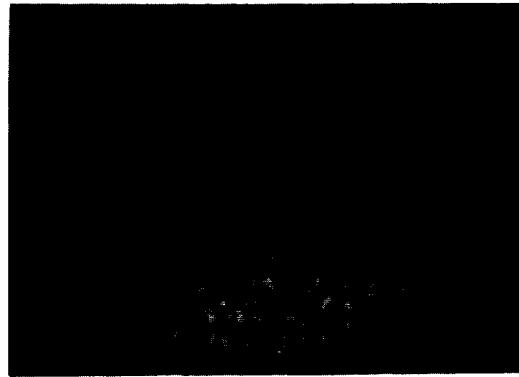
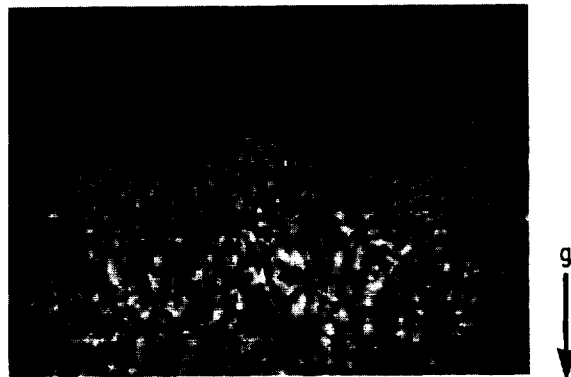


FIGURE 9. - CORRELATION OF Nu FOR PURELY THERMAL CASE,
 $Hr = 0.322$ (PRESENT CASE).



(a) t_1 .

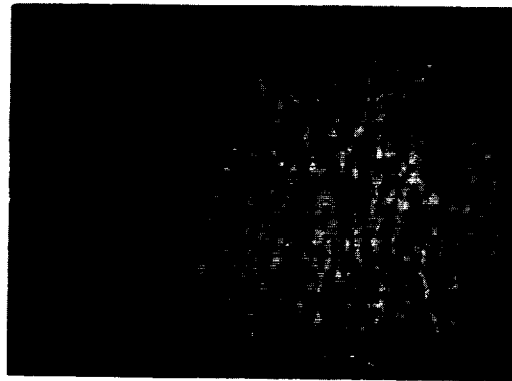


(b) $t_2 = t_1 + 5 \text{ SEC.}$

FIGURE 10. - FLOW PATTERNS NEAR THE CATH-
ODE LOWER PLATE (PURELY SOLUTAL CASE),
 $Ar = 1.02$, $Ra_m = 2.08 \times 10^{11}$, $t_1 < t_2$,
 $\Delta t = 5 \text{ SEC.}$



(a) t_1 .



(b) $t_2 = t_1 + 15$ SEC.

FIGURE 11. - FLOW PATTERNS NEAR THE ANODE TOP PLATE (PURELY SOLUTAL CASE),
 $Ar = 1.02$, $Ra_m = 2.08 \times 10^{11}$, $t_1 < t_2$,
 $\Delta t = 15$ SEC.

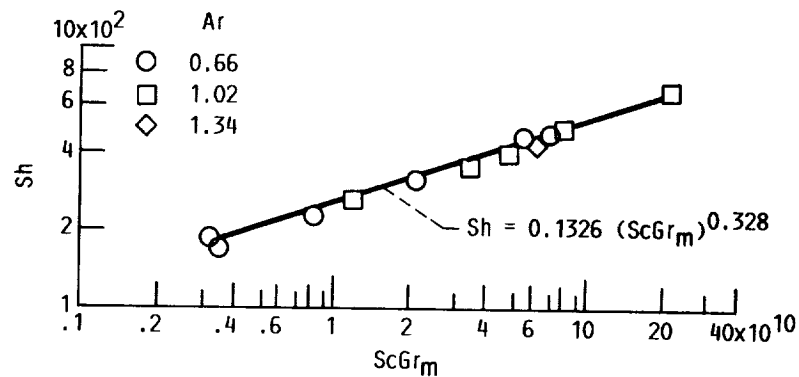


FIGURE 12. - CORRELATION OF Sh FOR PURELY SOLUTAL CASE.

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				6. Performing Organization Code	
7. Author(s) L.W. Wang, A.T. Chai, and D.J. Sun				8. Performing Organization Report No. E-4421	
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16. Abstract The transport process in the fluid phase during the growth of a crystal has a profound influence on the structure and quality of the solid phase. In vertical growth techniques the fluid phase is often subjected to vertical temperature and concentration gradients. The main objective of the present work is to obtain more experimental data on convective flows in enclosures with vertical temperature or concentration gradients. Among actual crystal systems the parameters vary widely. The parametric ranges studied herein for mass transfer experiments are mainly dictated by the electrochemical system employed herein to impose concentration gradients. Temperature or concentration differences are maintained between two horizontal end walls. The other walls are kept insulated. Experimental measurements and observations were made of the heat transfer or mass transfer, flow patterns, and the mean and fluctuating temperature distribution. For purely thermal cases, the experiment covers the ranges of thermal Rayleigh number $Ra = 9 \times 10^6$ to 2×10^9 , aspect ratio $Ar = 0.65$ to 2.6 , Prandtl number $Pr = 4.0$ to 7.0 . The method used to visualize the flow pattern in the thermal cases is an electrochemical ph-indicator method. For purely solutal cases the experiment covers the ranges of solutal Rayleigh number $Ra_m = 1.0 \times 10^9$ to 2.4×10^{11} , $Ar = 0.68$ to 1.34 , $Sc = 2000$ to 2100 . Laser shadowgraphs is employed to visualize flow patterns in the solutal cases. The results of the present experiments for the purely thermal cases are adequately correlated by the equation $Nu = 0.0862 Ra^{0.322}$ in the range $9 \times 10^6 < Ra < 2 \times 10^9$. For the purely solutal cases, the mass transfer data can be correlated by the equation $Sh = 0.1326 (Sc Gr_m)^{0.328}$ in the range $1 \times 10^9 < Ra_m < 2.4 \times 10^{11}$.					
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