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FREE CONVECTION ABOUT A RECTANGULAR PRISMATIC CRYSTAL GROWING FROM A SOLUTION

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Abstract – Sodium chlorate crystals growing from unstirred aqueous solutions were observed by schlieren techniques. Growth rate data were best correlated by $Sh = 0.48\ Ra^{0.25}$ with finite interface kinetics and a characteristic length L equal to the crystal height plus the inverse reciprocal sum of the horizontal dimensions. The growth rate was slightly higher when the plume was laminar than when it was irregular. The horizontal growth rate was greater than the vertical growth rate, with the ratio tending to decrease with increasing plume instability. The threshold Grashof number for plume instability was greatly decreased with decreasing height for crystals with the same horizontal dimensions. The frequency of eddy emission was proportional to $L^{-2}Ra^{0.8}$. The velocity of the eddies approached a constant value with increasing distance above the crystal surface, the terminal value being roughly proportional to $Ra^{1/2}/L$.

NOMENCLATURE

A,	aspect ratio, $h(a^{-1} + b^{-1})$;
a, b,	horizontal dimensions of crystal [m];
C,	solute concentration [mole/m^3];
ΔC,	supersaturation $(C_b - C_e)$ [mole/m^3];
C_p,	heat capacity of solution [J/kgK];
D,	diffusion coefficient of solute in solution [m^2/s];
d,	fraction of supersaturation required for mass transfer, $(C_b - C_i)/(C_b - C_e) = (C_b - C_i)/\Delta C$;
e, f,	constants;
F.	volume fraction of solute at crystal surface;
f,	frequency of eddy emission [s^{-1}];
g,	acceleration due to gravity [m/s^2];
Gr_c,	Grashof number for concentration differences, $\alpha(C_b - C_i)gL^3\rho^2/\mu^2$;
Gr_t,	Grashof number for temperature differences, $\beta(T_b - T_i)gL^3\rho^2/\mu^2$;
H,	average change in horizontal dimensions, $0.5(\Delta a + \Delta b)$[m];
h,	height of crystal [m];
h,	heat-transfer coefficient [W/m^2 K];
ΔH_f,	latent heat of crystallization [J/mole];
K,	mass-transfer coefficient [m/s];
	interface kinetics coefficient for linear kinetics [m/s];
L,	characteristic dimension of crystal as defined by equation (7) [m];
m,	slope as defined in equation (19);
N,	flux into crystal surface [mole/m$^2 \cdot$ s];
n,	number;
P,	perimeter of top surface, $2a + 2b$ [m];
Pr,	Prandtl number, $C_p\mu/\kappa$;
Q,	heat evolution per length of wire [W/m];
R,	residual as defined in equation (5);
r_{AB},	correlation coefficient between A and B variables, where one variable is correlated to some power (is actually for $\ln A$ vs $\ln B$);
Ra,	Rayleigh number, $g\Delta\rho L^3/\mu D = GrSc$;
Re,	Reynolds number of disturbances in plume, $v_{ed}L\rho/\mu$;
S,	Strouhal number, fL/v;
s,	standard deviation based on $n - 1$;
s',	standard deviation based on n;
Sc,	Schmidt number, $\mu/\rho D$;
Sh,	Sherwood number, $KL/D = N(1 - F) \times L/d\Delta CD$;
T,	temperature [K];
ΔT,	supercooling, 30°C − T [K];
t,	defined by equation (17);
V,	change in vertical dimension, Δh [m];
v,	velocity [m/s];
W,	minimum plume width (neck) [m];
y,	height above crystal surface [m].

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Greek symbols

α,	concentration densification coefficient, $\partial\rho/\rho\partial C$;
β,	thermal densification coefficient, $\partial\rho/\rho\partial T$;
κ,	thermal conductivity of solution [W/mK];
μ,	average viscosity of solution in boundary layer [kg/m · s];
ρ,	average solution density in boundary layer [kg/m^3];
$\Delta\rho$,	difference in solution density between bulk solution and interface [kg/m^3];
τ,	average period of eddy emission [s].

Subscripts

0,	at $y = 0$ (top crystal surface);
∞,	asymptotic value above crystal;
b,	value in bulk solution;
corr,	value for correlation;
e,	equilibrium value;
ed,	eddies;
exp,	value from experiment;
horiz,	horizontal value;
i,	interfacial value;
S,	schlieren;
vert,	vertical value.

Superscripts

i,	ith datum.

INTRODUCTION

POLYHEDRAL crystals are frequently grown by suspending the crystals in supersaturated solutions. When growth occurs, solute is removed from the solution; this usually lowers the solution density and causes a rising convection plume to issue from the vicinity of the crystal. A latent heat is also liberated at the crystal surface, causing a slight temperature difference between crystal and solution. As shown in Appendix A, however, this thermal effect is normally negligible in comparison to the compositional effect on natural convection.

Even so, growth of a polyhedral crystal from solution is significantly different from typical mass-transfer problems in several respects: (a) Transport about polyhedral objects has not been extensively studied. Even convection-free diffusion about a growing cube can be analyzed only by an approximate numerical solution [1]. (b) Another difficulty is the definition of an appropriate "characteristic length" L. Textbooks recommend the use of

$$L = (1/L_{\text{horiz}} + 1/L_{\text{vert}})^{-1} \tag{1}$$

in correlations for estimation of heat transfer about polyhedrons, although L_{horiz} is not specified. This can be traced back to a 1932 paper [2] citing data for spheres and cylinders and to an unpublished work by Montsinger on free convection about a single heated copper block in transformer oil or water. However, equation (1) is theoretically unacceptable since it leads to unreasonable values when either L_{horiz} or L_{vert} approaches zero. Therefore, a new formulation of L is required; in fact this is one of the aims of the present paper. (c) Additionally, the analogy between heat and mass transfer is not valid for a growing polyhedral crystal. The boundary condition at the surface of such a crystal is a constant flux, which neither corresponds to constant surface concentration nor to constant gradient of solute concentration [1].

EXPERIMENTAL DATA

As described in detail elsewhere [3,4], horizontal schlieren observations were made of NaClO$_3$ crystals growing from aqueous solutions. The crystals were all rectangular prisms with the top face approximately horizontal, and with a wide range of $a:b:h$ ratios. All solutions were saturated at 30°C. Both growth rates and plume behavior were determined. It was found that, as either supersaturation or horizontal crystal dimensions were increased, plume behavior changed from steady-state laminar flow to irregular or oscillatory flow with periodic shedding of eddies moving upward with the plume. Transitional plume behavior was also observed in which the periodic emission of eddies occurred only occasionally. The present paper contains improved correlations not given previously for growth kinetics and for plume behavior.

GROWTH KINETICS

crystals before and after growth had taken place for a known length of time at a constant temperature T. There were two major problems in correlating the growth data – unknown interface kinetics and considerable uncertainty as to the correct characteristic length L.

In order to extract mass-transfer coefficients from the data, it was assumed that the interface kinetics were linear and that an average surface concentration C_i of solute can be used. Using the derivations discussed in [1] we obtain

$$N = \frac{K(C_b - C_i)}{1 - F} = k(C_i - C_e) \tag{2}$$

where the symbols are defined in the table of nomenclature. The fraction of the supersaturation $(C_b - C_e)$ used in driving the mass transfer is found from equation (2) to be

$$d \equiv \frac{C_b - C_i}{C_b - C_e} = 1 - \frac{N}{k(C_b - C_e)}. \tag{3}$$

Thus, the Sherwood number is defined as

$$Sh \equiv \frac{KL}{D} = \frac{N(1 - F)L}{d(C_b - C_e)D}. \tag{4}$$

Similarly, the density difference in the Rayleigh number Ra is a function of d. The density difference, F, μ, D, and ρ all depend on d and on the supercooling ΔT; this is shown in Appendix B. For each definition of L described below, Sh was correlated vs Ra for a series of values of the interface kinetics constant k. It was assumed that, over the supercooling range studied (8°C), k was constant. Correlations were compared by calculating the correlation coefficient r and the residual

$$R = \frac{\left[\sum_{i=1}^{n} (N^i_{\text{exp}} - N^i_{\text{corr}})^2/n\right]^{1/2}}{s'_N}. \qquad (5)$$

The residual is the best measure since it gives the average error due to the correlation as a fraction of the average variation in N.

In order to obtain the most appropriate formulation of L, one must first note the recent results for free convection from horizontal surfaces [6, 7]. The appropriate L, over a wide range of shapes and Sh, was shown to be the ratio of the area to the perimeter, which for a rectangle is

$$L = 0.5(1/a + 1/b)^{-1}. \qquad (6)$$

This formulation can be applied to the present experimental data by assuming that only the top face of the crystal contributes to the plume behavior.

Alternatively, the formulation may be extended to three dimensional surfaces by noting [6, 7] that equation (6) corresponds to the average distance travelled by a fluid element from the perimeter of the surface to the point of convergence at the base of the plume. This concept, which seems intuitively reasonable, may be extended to a rectangular prism by defining L as the average path length from the bottom face to the top face, i.e.

$$L = h + (1/a + 1/b)^{-1}. \qquad (7)$$

Finally, we could combine equations (6) and (1) to obtain

$$L = (1/h + 1/a + 1/b)^{-1}. \qquad (8)$$

These three formulations of L were evaluated by comparison of correlations of the growth rate data.

Growth rates were determined in 53 runs with ΔT varied from 1.2 to 8°C, L [defined by equation (7)] from 4.3 to 12.5, and the aspect ratio A from 0.22 to 5.1. The correlation between A and h was only $r^2 = 0.07$, i.e. the crystal dimensions were varied independently of one another. The best least-squares correlations vs k for the three definitions of L are shown in Table 1. Inclusion of aspect ratio A in the correlations yielded only a slight improvement. Fits to the form $Sh = e + fRa^{1/4}$ yielded negative values of e and so were discarded.

The values for the residual in Table 1 are all virtually the same, and disappointingly high. Apparently this represents errors due to (a) errors in growth rate measurement due to inclusion of occluded solution in the weight measurements of the crystals, (b) surface irregularities on the crystals, and (c) deviations of the top surfaces from horizontal. We currently favor the correlation

$$Sh = 0.48\,Ra^{0.25}, \quad k = 0.003\ \text{cm/s} \qquad (9)$$

because it is very near the correlations for suspended cylinders and spheres [8–11], and because L, as defined in equation (7), is the only formulation that reduces to the proper values in the limits of $h \to 0$, $a \to 0$, or $b \to 0$. Hereinafter, we make exclusive use of L as defined by equation (7).

A plot of the equation (9) correlation is shown in Fig. 1. Note that the observations for an irregular plume were generally below the correlating line, while those for a laminar plume were generally above the line. By comparing deviations from the correlating line it was concluded that there is a 99.7% probability that this effect is real. In other words, we are forced to the surprising conclusion that the growth rate is lower for an unstable plume than for a stable one. Separate correlations for laminar and irregular plumes were found to be:

Laminar: $Sh = 0.78\,Ra^{0.22}$ $(r^2 = 0.68)$ (10)

Irregular: $Sh = 0.14\,Ra^{0.32}$ $(r^2 = 0.92)$. (11)

RELATIVE GROWTH KINETICS

Additional growth rate determinations were made by measuring the changes in vertical (V) and average horizontal crystal dimensions (H) caused by growth. These were measured by a micrometer in a set of 48 experiments. Supercooling ΔT was varied from 1.2 to 8°C, L from 4.3 to 18, and A from 0.22 to 3.9. H/V varied from 1 to 1.9 and was only poorly correlated by

$$\frac{H}{V} = 5.5\,Ra^{-0.089}A^{0.15} \quad (R = 0.81). \qquad (12)$$

This is plotted in Fig. 2. Note the almost complete separation according to plume stability. These data are replotted in Fig. 3 using the correlating equation for plume behavior given below. There is a clear trend, especially if one ignores the two points for which $H/V = 1$.

PLUME STABILITY

Plume stability was observed in 154 runs with Ra varied from 2×10^6 to 3×10^8, L from 3.8 to 20 mm,

Table 1. Summary of best growth rate correlations

L definition (equation)	k (cm/s)	Correlation	r^2	R	Average d
(6)	≥0.01	$Sh = 0.20\,Ra^{0.26}$	0.90	0.34	≥0.994
(7)	0.003	$Sh = 0.48\,Ra^{0.25}$	0.74	0.34	0.80
(8)	0.003	$Sh = 0.30\,Ra^{0.25}$	0.76	0.35	0.80

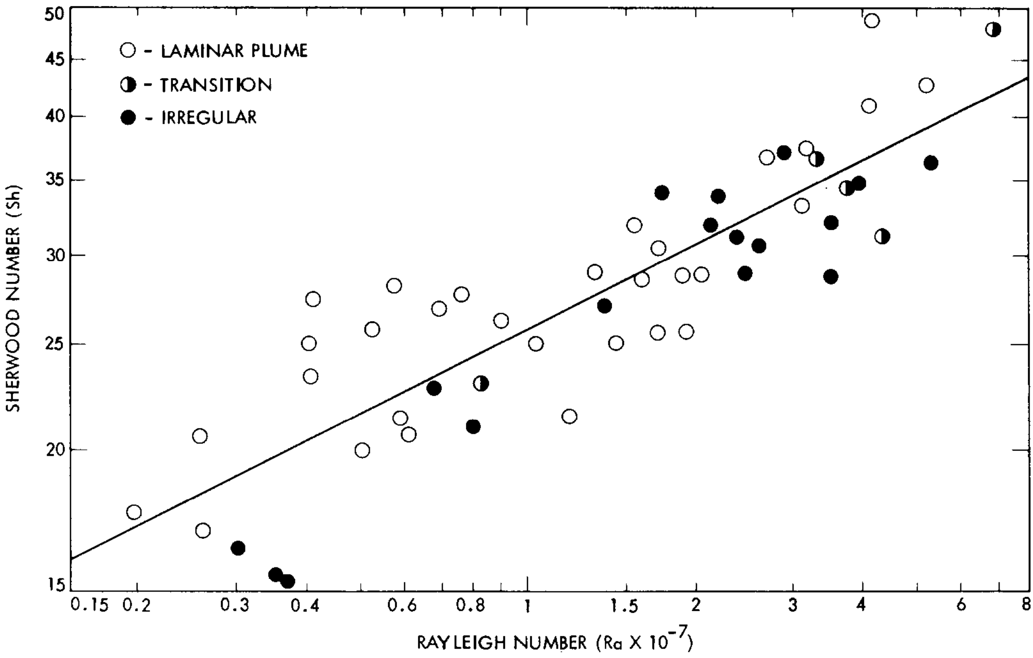

FIG. 1. Growth rate correlation with $k = 0.003$ cm/s. Sh calculated from experimental N, L, ΔT; Ra calculated from L, ΔT and equation (9) (to estimate d; see Appendix C). (Fit is slightly better than when Ra is also calculated from N, L, ΔT.) Line is equation (9). Plume behavior from correlation of Fig. 4.

ΔT from 1.2 to 9°C, and A from 0.22 to 5.2. Plume behavior for Ra vs A is plotted in Fig. 4. The equation of the straight portion of the line separating laminar from irregular plume behavior is

$$Ra = 1.3 \times 10^7 A^{2.2}. \qquad (13)$$

The few transition and irregular behavior points falling below the line are thought to be due to random vibrations in the system, which was shown [3, 4] to induce vortex shedding in plumes near to the critical condition.

It may readily be shown [3, 4] that the correlation of Fig. 4 corresponds to increasing plume instability as the height of the crystal is decreased – a very surprising result.

PLUME CHARACTERISTICS

As the plume emerged from the crystal it at first decreased in width, and then about h above the crystal began to increase. In other words, the plume had a minimum width W a finite distance above the crystal. Eleven measurements of W were made at $\Delta T = 8$°C, Ra ranging from 2×10^6 to 7.4×10^7, L from 6.6 to 11.1 mm, and A from 0.82 to 2.1. The results were correlated by

$$W/P = 2.4 \times 10^{-9}\, Ra - 0.0071 \quad (r^2 = 0.96) \qquad (14)$$

and

$$W/L = 8.9 \times 10^{-11}\, Ra^{1.3} \quad (r^2 = 0.96). \qquad (15)$$

Unfortunately, the influence of A could not be tested independently because the experimental parameters were varied in such a way that ln A and ln Ra had a correlation of $r^2 = 0.98$, i.e. A and Ra were varied in constant ratio inasmuch as h was kept at about 5 mm for all experiments.

In the same 11 experiments, the number of schlieren lines n_s in the plume were measured. These data were correlated very well by

$$\frac{W}{L(n_s + 1)} = 0.034 \quad (s = 0.0030). \qquad (16)$$

These dark lines on the schlieren image correspond to cycles of oscillation of the index of refraction gradient and therefore roughly indicate the number of refractive index minima in the plume. They may therefore indicate the number of cells in the plume. Equation (16) indicates that the width of each "cell" $[W/(n_s + 1)]$ at the plume neck is a fixed fraction of the crystal dimensions. As Ra is increased, more "cells" are added.

At Rayleigh numbers slightly above the critical value given by Fig. 4, only a few (n_{ed}) of the schlieren lines exhibited periodic irregularities or eddies. The others seemed to remain laminar. As Ra was increased, both the number of schlieren lines increased (as above) and the fraction of those containing eddies increased. The fraction containing eddies was correlated by

$$t \equiv \sqrt{2}\,\mathrm{erf}^{-1}(n_{ed}/n_s) = 0.024 + 2.8 \times 10^{-23}\, Ra^{2.9}$$
$$(r^2_{\ln t : \ln Ra} = 0.94) \qquad (17)$$

which is plotted in Fig. 5.

The frequency of generating irregularities in a given schlieren line at a point on or near the crystal surface was relatively constant. This periodicity was measured in 22 experiments with Ra varied from 1.7×10^6 to 7.3×10^7, ΔT from 3 to 5°C, L from 3.8 to 12.6 mm, and A from 0.22 to 2.2. The average periodicity was correlated by

$$\frac{\tau\mu}{\rho L^2} = 9.6 \times 10^4 Ra^{-0.81} \quad (r^2 = 0.97) \qquad (18)$$

which is plotted in Fig. 6. This is perhaps comparable with the results of Aiba and Seki [12] in which the frequency of plume sway above a horizontal heated plate was found to be proportional to $(\mu/\rho L^2)Gr^{0.5}$.

Finally, the velocity of the eddies was found to increase with height above the top face of the crystal, approaching a constant value. Five experiments were made with measurements of eddy velocity v_{ed} vs height y, with ΔT varied from 3 to 8°C, L from 4.7 to 10.1 cm, A from 0.61 to 0.98, and Ra from 3.5×10^6 to 5.5×10^7. The data for each experiment were fitted to an equation of the form

$$\frac{Re_t - Re}{Re_t - Re_0} = e^{-my/L} \qquad (19)$$

by calculating the best value of m for a series of selected values of Re_t. The Re_t yielding the highest r^2 was taken as the best fit. Thereby, values of m were obtained of 1.5, 2.2, 2.3, 2.5 and 2.5. The square of the correlation coefficient r^2 ranged from 0.80 to 0.995, with the smallest being for $m = 1.5$. Thus, the data were all recorrelated using an average value of $m = 2.2$ for all. The resulting correlation for all five runs is shown in Fig. 7. The five values of Re_0 and Re_t resulting therefrom were, in turn, correlated by

$$Re_0 = 1.1 \times 10^{-7} Ra^{0.72} \quad (r^2 = 0.89) \qquad (20)$$

$$Re_t = 4.1 \times 10^{-6} Ra^{0.63} \quad (r^2 = 0.999!). \qquad (21)$$

These correlations are shown in Fig. 8. A series of 26 other runs were made in which only the terminal eddy velocity was observed; L was varied from 4.5 to 13.3 mm, A from 0.22 to 1.9, ΔT from 3 to 8°C, and Ra

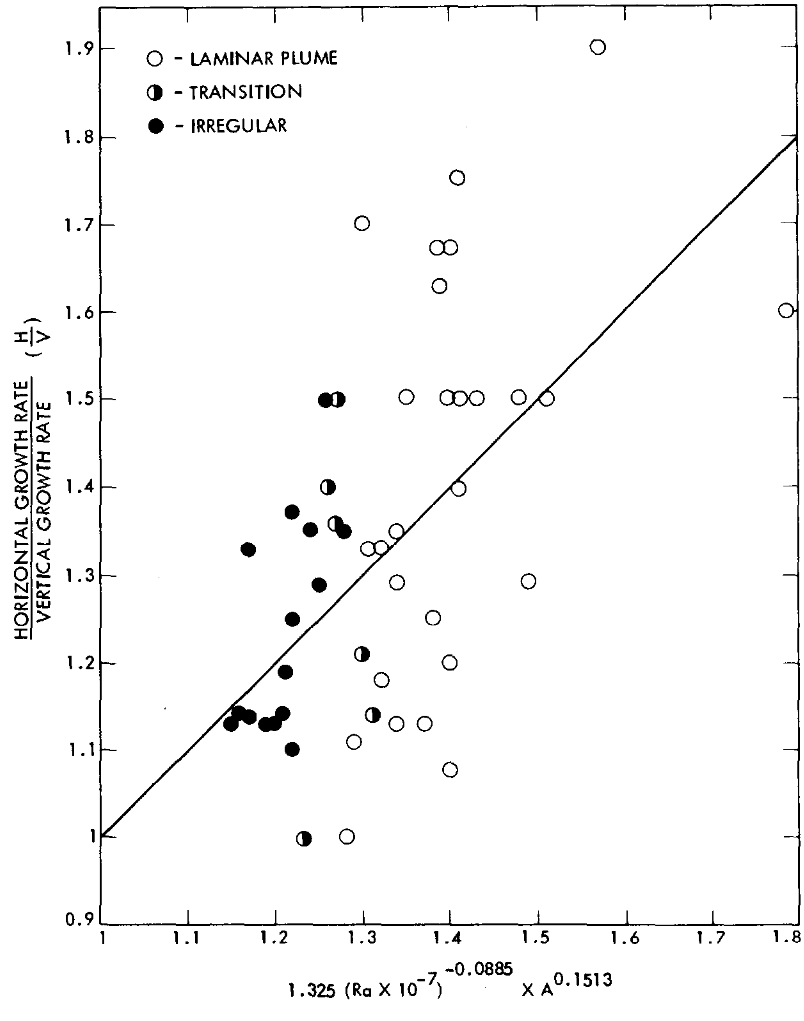

FIG. 2. Experimental ratio of horizontal growth rate to vertical growth rate (H/V) compared to correlation of equation (12).

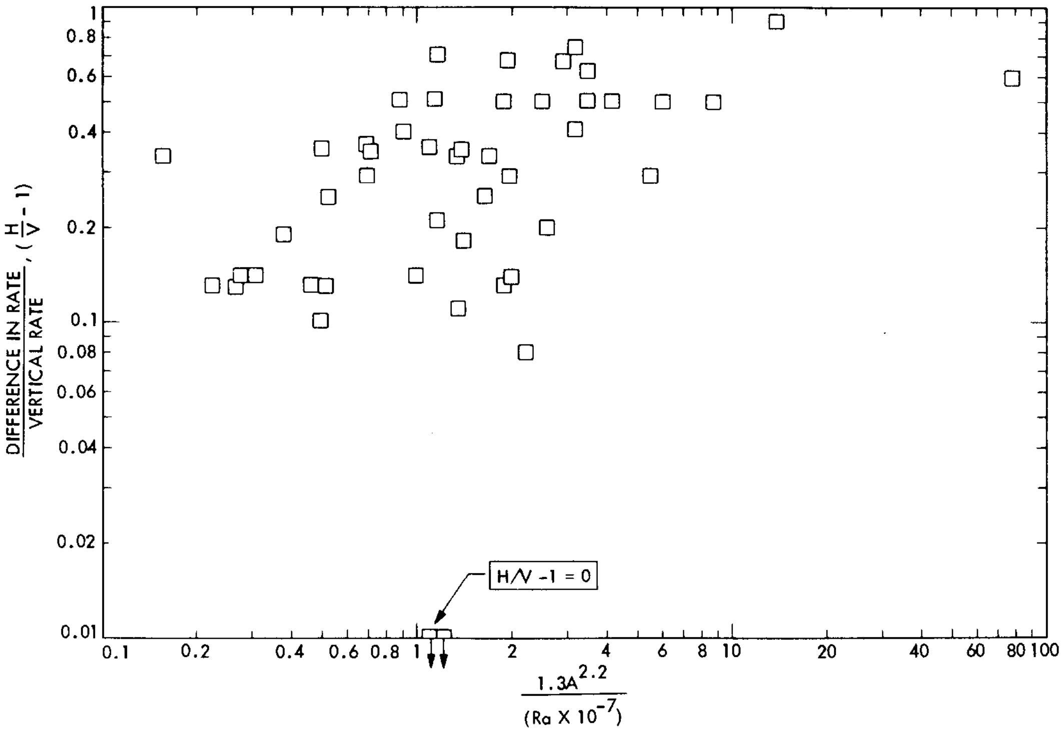

FIG. 3. *H*/*V* vs deviation from plume stability line in Fig. 4.

from 3 × 10^6 to 8.3 × 10^7. These data were correlated by

$$Re_t = 7.6 \times 10^{-5} Ra^{0.44} \quad (r^2 = 0.85), \qquad (22)$$

as also shown in Fig. 8. Note the values of Re_t determined by direct observation were generally lower than those determined by fitting v_e vs y data. We do not know if this represents an error in measurement (by not measuring at sufficiently large y) or a failure in equation (19) at large y.

These results do not agree with those for the plume above a linear heat source. For example, Nawoj and Hickman [13] found that the centerline velocity was proportional to $Q^{0.54}$ which with $Q \propto Ra^{1/4}$ would lead us to expect for our experiments $Re \propto Ra^{0.14}$. Gebhart *et al.* [14] predicted the velocity to be proportional to $y^{1/5}Q^{2/5}$, but their analysis was based on boundary-layer equations which are not applicable in our case.

It is also interesting to compare our plume behavior results to those on wakes in forced convection about spheres and cylinders. The Strouhal number $S = fL/v$ was found to increase slowly with increasing free stream velocity v [15–17]. Similar behavior in free convection would lead to the expectation that $1/\tau$ and Re would have almost the same Ra dependence. Equations (18) and (20) show that this is true within experimental error.

DISCUSSION

Our results indicate that free convection about suspended polyhedra is quite different from that expected by analogy to prior results on spheres, cylinders, and flat horizontal or vertical plates. We would hesitate to apply our results to heat transfer or to growth of anisotropic crystals. Even the validity of the present results to other crystals with cubic habit is uncertain. This is primarily because of the peculiar boundary condition in polyhedral crystal growth: (a) constant flux rather than constant concentration or constant gradient, and (b) finite and even site-dependent kinetics at the surface. In order to correlate the data, we were forced to assume that the concentration was constant over the entire surface of the crystal, albeit with finite interface kinetics. In other words, we assumed in essence a stagnant film model with no localized effects due to growth spirals or steps. We are not optimistic about the prospects for a realistic theoretical solution of this problem.

On the other hand, the analogous heat transfer problem may well admit of generalized experiments and exact theoretical solutions. We therefore recommend that heat-transfer experiments be performed on polyhedra, both with constant surface concentration and with constant heat flux. Professor Wirtz at Clarkson College of Technology is preparing to perform such experiments.

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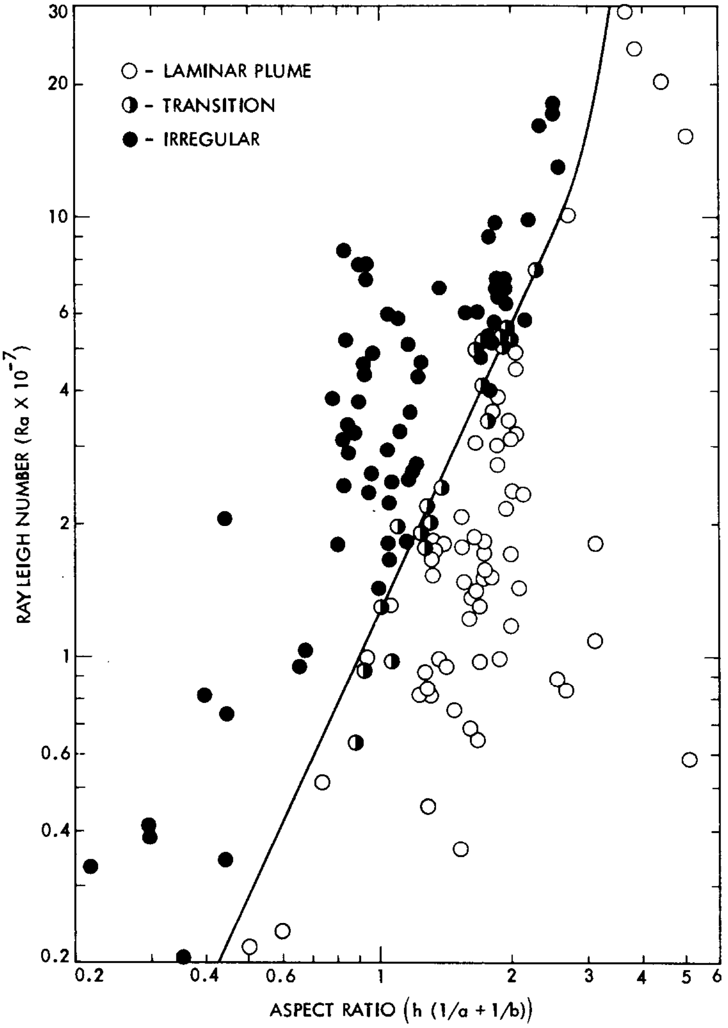

FIG. 4. Plume stability correlation. Laminar plumes below line, irregular plumes above.

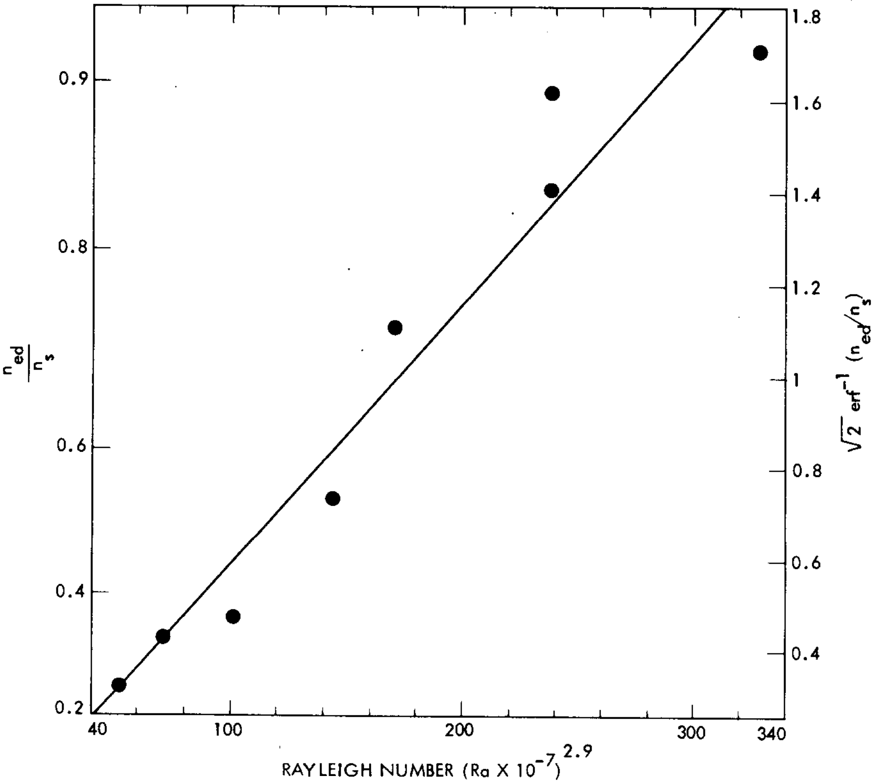

FIG. 5. Fraction n_{ed}/n_s of schlieren lines in plume that contain eddies, as function of *Ra* as per equation (17).

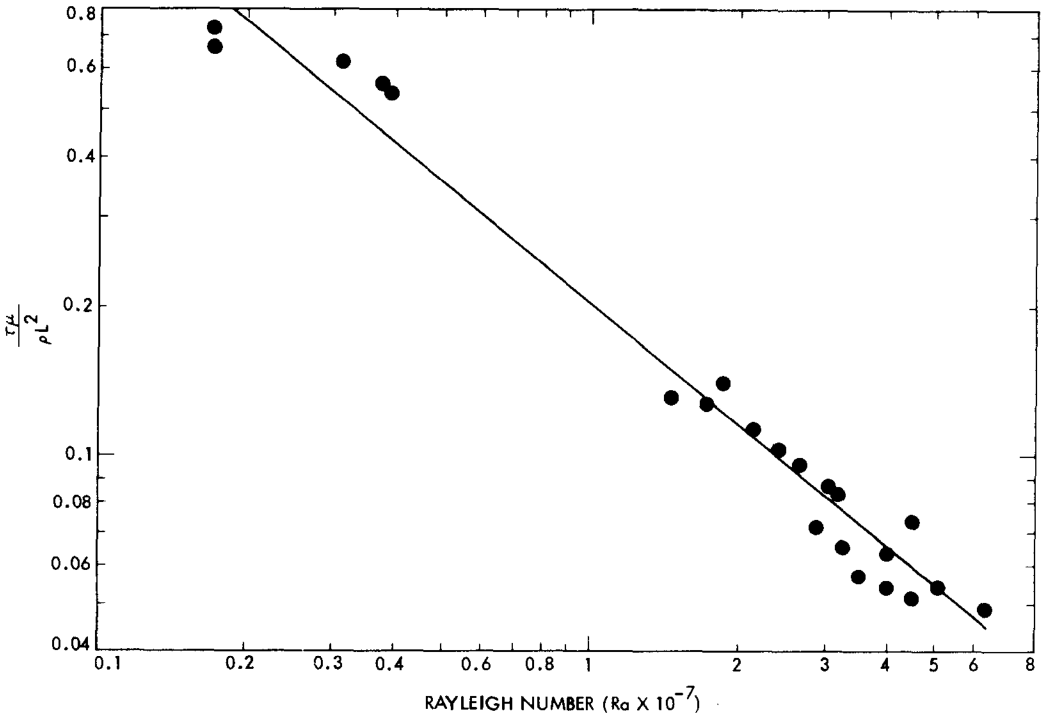

FIG. 6. Correlation for average period τ of eddy emissions into plume.

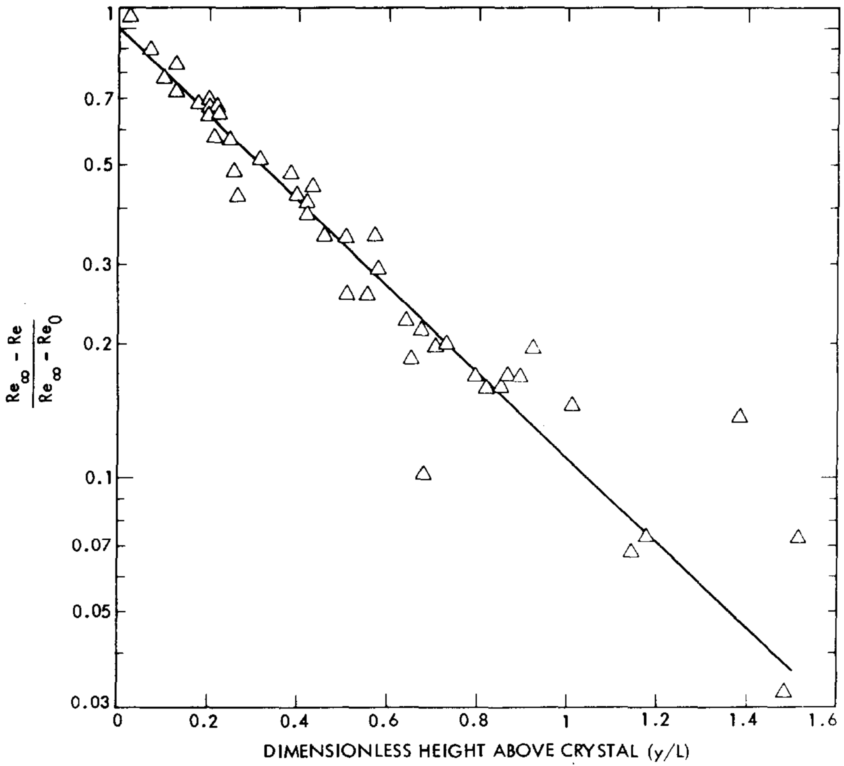

FIG. 7. Correlation for eddy velocity in plume vs height y above crystal surface. Re_∞ and Re_0 found for each of five experiments by regression analysis.

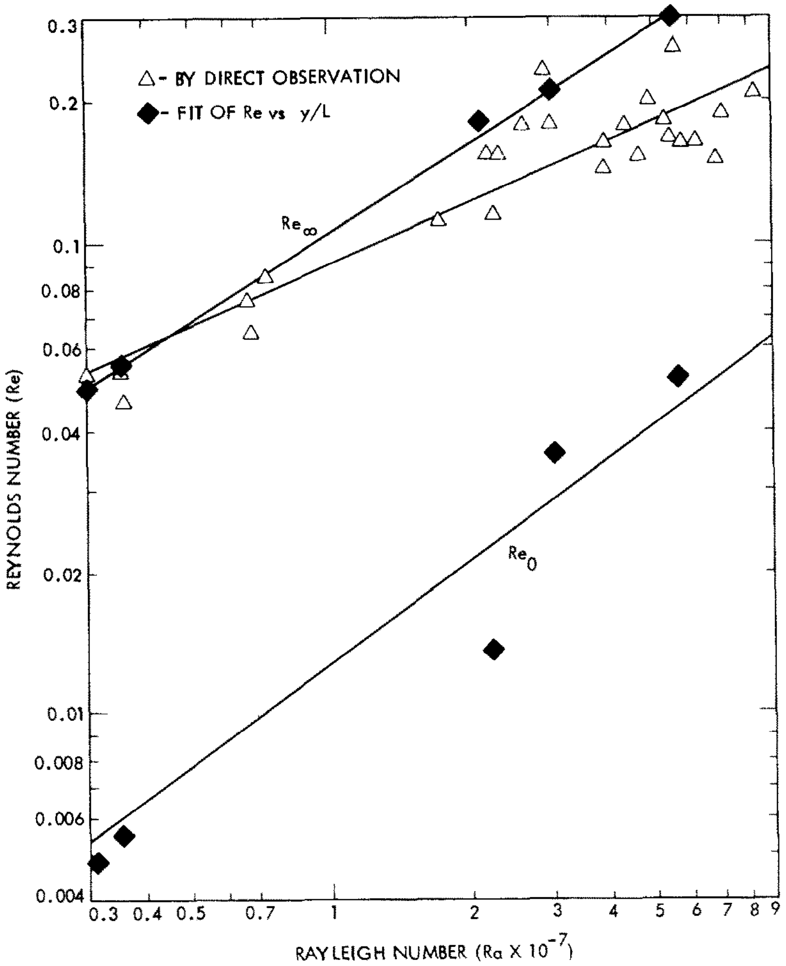

FIG. 8. Correlations for Re_0 and Re_∞.

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APPENDIX A

Importance of Heat Transfer in Crystal Growth from Solution.

Latent heat is liberated at the surface of the crystal during growth. Since the crystal is suspended in the solution, the latent heat must be dissipated in the surrounding solution. For the usual positive latent heat of crystallization, this causes the crystal to be warmer than the solution and contributes to the free convection about the crystal. The question is, how much of a contribution does it make to the free convection? We estimate that contribution here, using the film model and prior results for simultaneous heat and mass transfer.

At steady state, the rate of latent heat liberation equals the rate of heat transfer into the solution, i.e.

$$q = N\Delta H_f = h(T_i - T_b) = N\frac{K}{L}\Delta T. \tag{23}$$

There are two principal predictions for the relative driving forces for heat and mass transfer in free convection. One yields [18], with equations (4) and (23) and physical properties estimated for $NaClO_3$ growing from an aqueous solution:

$$\begin{aligned}\frac{\text{heat transfer driving force}}{\text{mass transfer driving force}} &= \frac{Gr_t}{Gr_c}\left(\frac{Sc}{Pr}\right)^{1/2}\\ &= \frac{\beta(T_i - T_b)}{\alpha(C_b - C_i)}\left(\frac{Sc}{Pr}\right)^{1/2}\\ &= \frac{\beta}{\alpha}\left(\frac{Pr}{Sc}\right)^{1/8}\frac{\Delta HF_f}{(1-F)\rho C_p}\\ &= 0.004. \end{aligned}\tag{24}$$

On the other hand, Gebhart and Pera [19] give with their $P = Q$,

$$\begin{aligned}\frac{\text{heat transfer driving force}}{\text{mass transfer driving force}} &= \frac{Gr_t}{Gr_c} = \frac{\beta(T_i - T_b)}{\alpha(C_b - C_i)}\\ &= \frac{\beta}{\alpha}\left(\frac{Pr}{Sc}\right)^{5/8}\frac{\Delta H_f}{(1-F)\rho C_p} = 0.05. \end{aligned}\tag{25}$$

Thus, heat-transfer contributes roughly between 0.5 and 5% of the free convection driving force in crystal growth from typical aqueous solutions. The enhanced convection increases the growth rate, while the increased temperature of the crystal decreases the growth. The net effect is certainly much less than our experimental errors.

APPENDIX B

Physical Properties

All solutions were saturated at 30°C. Solubility, density and viscosity were measured versus temperature and concentration, for concentrations corresponding to saturation at the measurement temperature up to saturation at 30°C [3, 4, 20]. Diffusion coefficient and viscosity data for undersaturated solutions were also taken from Landolt–Bornstein. It was assumed that all properties are linear in supercooling and temperature, and least-squares fits were made to the data where possible. Because of insufficient data for D, it was assumed that $D = 1.5 \times 10^{-5}\ \mathrm{cm^2/s}$ for a saturated solution at 30°C. Literature data were used to extrapolate to other conditions.

With these assumptions and data, the average property values in the boundary layer were estimated to be given by

$$\rho = 1.446[1 + (0.00047 - 0.000888d)\Delta T]\mathrm{g/cm^3} \tag{26}$$

$$\mu = 1.875[1 + (0.0268 - 0.0038d)\Delta T]\mathrm{cP} \tag{27}$$

$$D = 1.5 \times 10^{-5}[1 - (0.029 - 0.021d)\Delta T]\mathrm{cm^2/s}. \tag{28}$$

Similarly, the interfacial volume fraction of solute is estimated to be

$$F \doteq 0.282 - 0.0017d\Delta T \tag{29}$$

and the supersaturation ΔC and supercooling ΔT are related by

$$\Delta C = 4.268 \times 10^{-5}\Delta T\ \mathrm{mole/cm^3} \tag{30}$$

with ΔT in °C. The density difference between interface and bulk solution is

$$\Delta\rho = 0.00257d\Delta T\ \mathrm{g/cm^3}. \tag{31}$$

APPENDIX C

Calculation of Ra *from* L *and* ΔT

Because the interfacial conditions both depend on Ra and influence Ra, an iterative process is required if the growth rate is not known independently. While iteration between equations (3), (4) and (9) is feasible, convergence is slow. Much more rapid convergence is attained if d is estimated by the following simultaneous solution of equations (3), (4) and (9):

$$d = \left[1 + \frac{0.48}{1-F}\left(\frac{0.00257dg\Delta TD^3}{L\mu}\right)^{1/4}\right]^{-1}. \tag{32}$$

Thus d from a previous iteration is used on the RHS of the equation to estimate a new value. Three or four iterations suffice to give d to three significant figures, with typical values being near 0.8.

CONVECTION NATURELLE AUTOUR D'UN CRISTAL DE FORME PRISMATIQUE RECTANGLE ET CROISSANT A PARTIR D'UNE SOLUTION

Résumé—On observe, par la technique strioscopique, des cristaux de chlorure de sodium qui croissent à partir de solutions aqueuses immobiles. La vitesse de croissance est traduite par $Sh = 0{,}48Ra^{0.25}$ avec une cinétique finie à l'interface et une longueur caractéristique L égale à la hauteur du cristal plus l'inverse de la somme des inverses des dimensions horizontales. La vitesse de croissance est légèrement plus grande quand le panache est laminaire que lorsqu'il est irrégulier. La croissance horizontale est plus forte que la croissance verticale, avec un rapport qui tend à diminuer lorsque l'instabilité du panache augmente. Le nombre de Grashof du seuil d'apparition des instabilités décroit fortement lorsque la hauteur diminue pour des cristaux ayant même dimensions horizontales. La fréquence d'émission des tourbillons est proportionnelle à $L^{-2}\,Ra^{0.8}$. La vitesse des tourbillons approche une valeur constante quand la distance au dessus de la surface du cristal augmente, la valeur limite étant à peu près proportionnelle à $Ra^{1/2}/L$.

FREIE KONVEKTION UM EINEN AUS EINER LÖSUNG WACHSENDEN RECHTECKIGEN PRISMATISCHEN KRISTALL

Zusammenfassung—Natriumchloratkristalle, die aus nicht gerührten wäßrigen Lösungen wachsen, wurden durch Schlierentechnik beobachtet. Die Daten für das Wachstum wurden am besten durch $Sh = 0.48Ra^{0.25}$ korreliert, bei Kinetik mit endlicher Grenzfläche und einer charakteristischen Länge L gleich der Höhe des Kristalls plus der invers reziproken Summe der horizontalen Abmessungen. Die Wachstumsrate war bei laminarer Strömung etwas höher als bei turbulenter. Die horizontale Wachstumsrate war größer als die vertikale, wobei das Verhältnis mit zunehmender Turbulenz der Strömung zum Abnehmen tendierte. Der Schwellwert der Grashof-Zahl für turbulente Strömung nahm bei gleichen horizontalen Abmessungen mit abnehmender Kristallhöhe stark ab. Die Frequenz der Wirbelbildung war proportional zu $L^{-2}Ra^{0.8}$. Die Geschwindigkeit der Wirbel näherte sich mit zunehmendem Abstand von der oberen Kristallfläche einem konstanten Wert, wobei dieser Endwert ungefähr proportional $Ra^{1/2}/L$ war.

СВОБОДНАЯ КОНВЕКЦИЯ ВОКРУГ ПРЯМОУГОЛЬНОГО ПРИЗМАТИЧЕСКОГО КРИСТАЛЛА, ВЫРАЩИВАЕМОГО ИЗ РАСТВОРА

Аннотация — С помощью теневых методов наблюдался рост кристаллов хлорноватокислого натрия из невозмущенных водных растворов. Данные по скорости роста наилучшим образом обобщались выражением $Sh = 0{,}48Ra^{0.25}$ при использовании кинетики для конечной поверхности раздела и характерной длины L, равной сумме высоты кристалла и обратной сумме горизонтальных размеров. Скорость роста кристалла при ламинарном подъеме свободноконвективных струек была несколько выше, чем при хаотическом. Горизонтальная составляющая скорости роста превышала вертикальную, причем отношение этих величин уменьшалось с увеличением нестабильности струек. Пороговое значение числа Грасгофа неустойчивости струек значительно снижалось с уменьшением высоты кристаллов при тех же горизонтальных размерах. Вихри образовывались с частотой, пропорциональной $L^{-2}Ra^{0.8}$. Скорость вихрей стремилась к постоянному значению с увеличением расстояния от поверхности кристалла, причем ее конечное значение было примерно пропорционально величине $Ra^{1/2}/L$.