The Effect of Natural Convection on Ice Crystal Growth Rates in Salt Solutions

Many measurements of ice growth rates in NaCl solutions have been made to determine the role of natural convection in the control of such growth rates. The results of two analyses are presented. In one, only thermal natural convection is treated for growth in pure water. The second treats the simultaneous effects of both mass and thermal natural convection in salt solutions. This second analysis correctly predicts higher ice growth rates in dilute salt solutions than in pure water.

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SCOPE

The improved experimental apparatus of Kallungal has been used to obtain many new accurate data on the growth rate of ice crystals in salt solutions. Crystals have been grown vertically, both up and down, as well as horizontally to see if any differences exist, as would be predicted if natural convection affects the growth rates. The results of mathematical analyses of natural convection effects on crystallization are presented and compared to our observations.

A novel explanation of the effects of thermal natural convection is offered, based on an analysis of the maximum density temperature of aqueous NaCl solutions.

CONCLUSIONS AND SIGNIFICANCE

It is shown that NaCl solutions containing more than 2.37 mass % salt will behave very differently from those containing less than this salt concentration, because at 2.37 mass % NaCl the temperature of maximum density coincides with the freezing point, while for lesser concentrations the solution becomes more dense with increasing temperature. The opposite is true for solutions more concentrated than 2.37 mass %. This explains many but not all of our observations.

Our mathematical analyses of crystal growth with natural convection show that natural convection causes "anomalous" higher growth rates of ice in dilute salt solutions than in pure water for any given subcooling. An analysis using both thermal and mass natural convection correctly predicts the maximum growth rate to occur at about 0.9 mass % NaCl. However, the actual growth rates predicted are one order of magnitude lower than those observed. So we do not yet have an accurate theory of ice growth rates, but it is now reasonably certain that natural convection, both mass and thermal, strongly affects growth rates and explains the "anomalous" growth rates in dilute salt solutions.

INTRODUCTION

The freezing of ice from salt solutions is probably the second most important phase change to take place on this planet. Presumably the evaporation and condensation of water is the most important, but the formation of ice from the oceans determines the level of the sea and thus the extent of habitable land. It also caused the ice ages, glaciers, and the subsequent scouring of the land, with its formation of soil, valleys, icebergs, etc. We should thus understand something of the growth rate of ice from pure water or salt solutions, but there is not yet a viable theory of why ice grows at the rate it does. We have studied this interesting problem in the laboratory for twenty years and still do not have a complete answer to all our questions, but we get closer to those answers continuously. At first we were interested because of our work with desalting sea water by freezing methods, but we found that the problem of describing rates of crystal growth was a more general unsolved problem not confined solely to ice formation. In fact the design of industrial crystallizers is really an art rather than a science; i.e., it takes more experience than fundamental knowledge. Someday we hope to be able to predict with confidence the rate of growth at least for ice crystals under a wide variety of conditions. Perhaps when this is accomplished the models will be applicable to growth of all kinds of crystals.

Previous work by Fernandez (1967), Poisot (1968), Vlahakis (1974), and Kallungal (1975, 1977) on ice crystal growth rates in pure water and NaCl solutions has produced interesting effects, such as: (a) a faster growth rate of ice from dilute salt solution than from pure water, and (b) a lack of steady-state growth rates at certain combinations of salt concentration and growth direction. Neither phenomenon could be explained conclusively, although several theories were examined. We thus decided to get additional data on growth in solutions using the new, superior apparatus of Kallungal and Barduhn (1977) to prove or disprove various conjectures put forth, and especially to determine the role of natural convection on the results.

Extensive literature searches on our knowledge of ice crystal growth rates have been made and summarized by Kallungal (1975)

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TABLE 1. RANGE OF VALUES OF INDEPENDENT VARIABLES

Variable	Symbol	Units	Range
Subcooling	ΔT	K	0.1 to 1.0
Salt Concentration	W	Mass % NaCl	0.25 to 6.11
Forced Velocity	V	cm/s	0 and 6.8 $\times 10^{-5}$ to 78.0
Orientation of Growth, Gravity	—	—	Vertically up or down, or horizontal

and Huang (1975).

Kallungal (1977) showed that crystals growing downward in quiescent pure water do not grow at a steady state; only for upward or horizontal growth does one observe a steady state growth in quiescent water. He explained this convincingly by presuming thermal natural convection to control the heat transfer when no forced convection was applied.

In salt solutions, however, it is probable that both thermal and mass natural convection are operative and affect the growth rate, since the rejection of both salt and heat at the growing interface will create both concentration and thermal gradients which influence local velocities and thus heat and mass transfer and growth rates.

We thus present in plotted form more than 300 new data points in Figures 1–8 of ice growth rates in various salt solutions. The tabulated data are available (Barduhn et al., 1976; Huang, 1976). The independent variables are ΔT, the subcooling; W, the mass % NaCl in solution; V, the forced convection velocity; and the growth direction with respect to gravity. The applied forced convection, if any, is always directly opposite the growth direction. The crystallographic growth direction is always in that of the a-axis. The range of each independent variable used is shown in Table 1. The measured growth rates (v) ranged from 2×10^{-4} to 0.077 cm/s.

Figure 1 presents growth rate data in quiescent solutions as a function of % salt in the solution with subcooling as the parameter. Note that the ratio of the growth rates at the maximum (at about 1% NaCl) to that in pure water ranges from 2.8 to 4.6 and any theory of growth rates must account for this. These are increases of 180 to 360%. The data in pure water are from Kallungal (1975) and are calculated from his equation for this condition ($W = V = 0$) with upward growth: v (cm/s) $= 0.0118\ \Delta T^{2.17}$. Note also that the higher ratios are at the smaller subcoolings.

Many investigators have reported higher growth rates in dilute solutions beyond that in pure solvent, and the reasons put forth have been varied and altogether unsubstantiated. One reason given is that fluid motion is caused by shrinkage or expansion when the solid is formed from the liquid (because of density differences between the crystal and solution). Another reason given for higher growth rates is surface tension anomalies. Few investigators have even suspected natural convection. It has been reported (anon., 1976) that no fluid motion is observed during freezing under zero gravity conditions, so additional fluid velocity must certainly come from natural convection caused by gravity.

The solid lines in Figure 1 for $0 < W < 2\%$ NaCl are the steady growth rates observed directly. For the rates in 4 and 6.11% NaCl solutions no steady values were observed, but steady growth rates were measured with small forced velocities going down over the upward growing crystal and then extrapolated to 0 forced velocity.

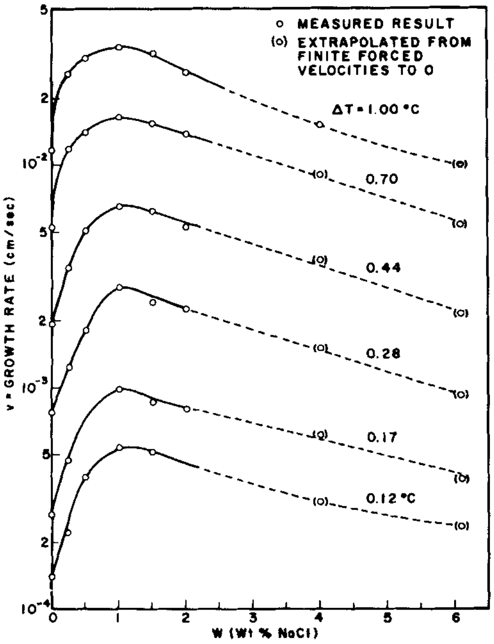

Figure 1. Measured and extrapolated growth rates of single ice crystals growing upward in quiescent NaCl solutions.

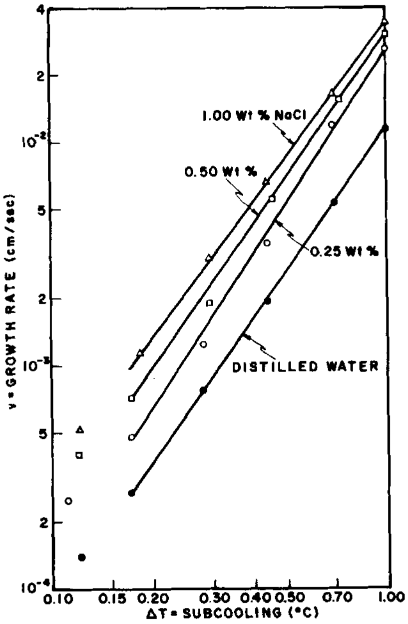

Figure 2. Measured growth rates of single ice crystals growing upward in quiescent, low-concentration NaCl solutions.

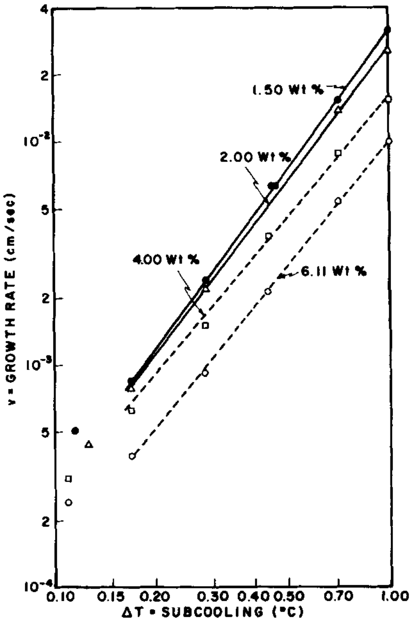

Figure 3. Measured and extrapolated rates of single ice crystals growing upward in quiescent, high-concentration NaCl solutions.

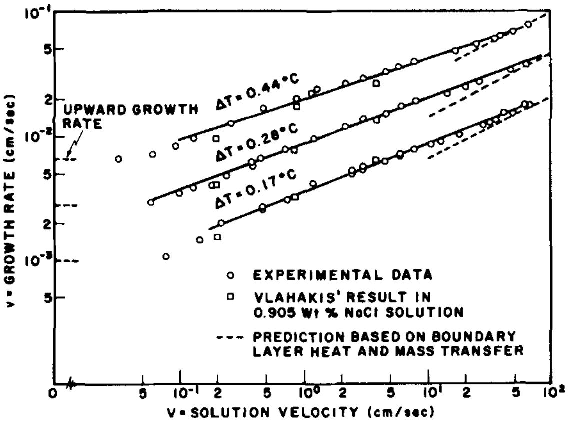

Figure 4. Horizontal growth rates of single ice crystals in 1.00 wt. % NaCl solution.

Figures 2 and 3 show the data on v vs ΔT for upward growth in a quiescent solution ($V = 0$) with W = from 0 to 6% NaCl as the parameter. The slope of the lines is 2.0 ± 0.2. These slopes do not agree with the natural convection analysis presented below nor to the forced convection solution of Fernandez (1967), both of which require a slope of 1.5.

In Figure 4 growth rate is plotted vs. solution velocity for 1.0% NaCl solutions with ΔT as parameter, while Figure 5 shows the same thing for 6% NaCl solutions. The dashed lines at the far righthand side of Figures 4 and 5 are based on the heat and mass transfer analysis for only forced convection of Fernandez (1967) and thus have slopes of 0.5.

Figures 6 and 7 give growth rates vs. salt concentration for fixed ΔT's of 0.44 and 0.28 K. The parameter is the forced velocity, V = 0 to 60 cm/s. Note that the enhanced growth rate gradually disappears as the forced velocity increases. This is as would be expected if natural convection causes this enhanced growth rate.

Figure 8 compares growth rates vs. the forced velocity in the three directions studied in 6% salt solution at two different ΔT's. From this plot we see that upward growth rates are slightly greater than horizontal ones at the lower forced velocities, but all three growth directions give substantially the same rate at $V > 0.2$ cm/s. Note that there is no steady state for downward growth with V less than 0.2 cm/s at this salt concentration.

The nature and shape (morphology) of the growing ice crystals is discussed extensively by Huang (1975).

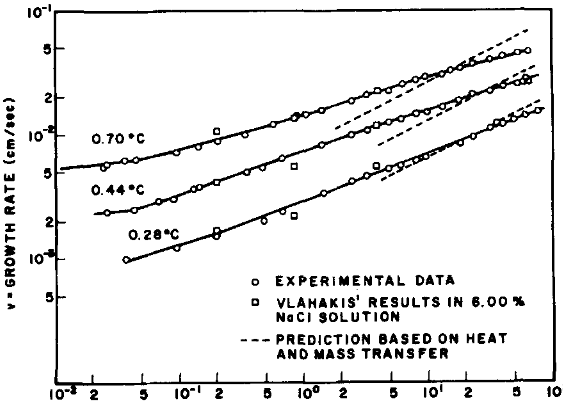

Figure 5. Horizontal growth rates of single ice crystals in 6.11 wt.% NaCl solution.

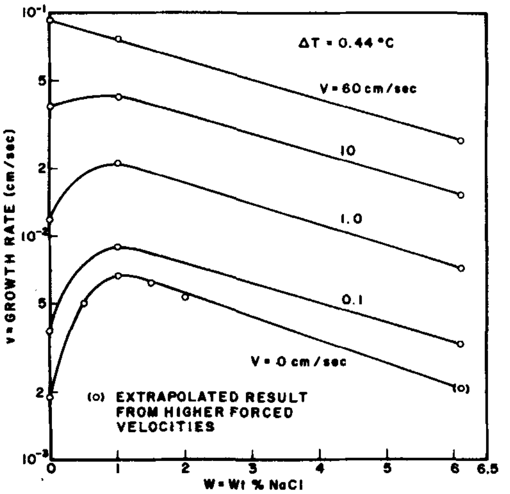

Figure 6. Ice crystal growth rate in NaCl solutions vs. concentration, with solution velocity as a parameter; subcooling = 0.44°C.

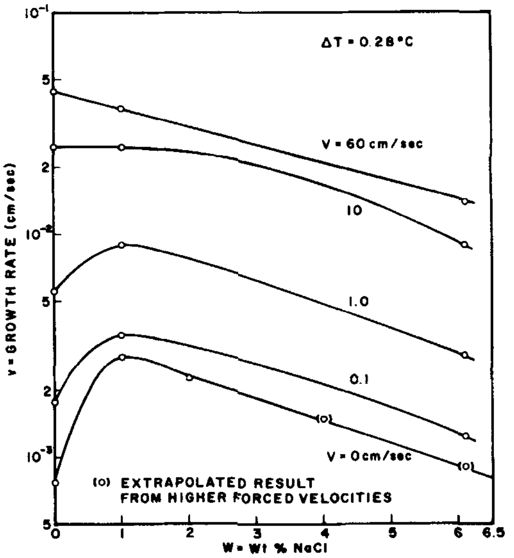

Figure 7. Ice crystal growth rate in NaCl solutions vs. concentration, with solution velocity as a parameter; subcooling = 0.28°C.

For any condition under which there is no steady state growth we can always get a steady state by applying an adequate forced velocity opposite to the growth direction. A thorough investigation of this was made. Figure 9 shows the forced velocity required to get steady growth for various growth directions as a function of the mass % NaCl in solution. The highest subcooling data available were used in each case. In the region above each curve steady growth is observed, while below each curve the rate drops off with time. Note that these values of forced velocity might change if the dimensions of the apparatus were changed, since natural convection exerts itself more easily in a large space than a small one.

Figure 9 shows that the forced velocity required for steady growth is lowest for upward growth, intermediate for horizontal growth, and highest for downward growth at any salt concentra-

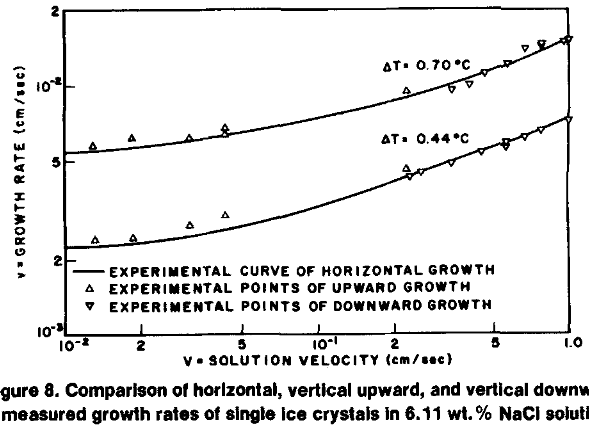

Figure 8. Comparison of horizontal, vertical upward, and vertical downward measured growth rates of single ice crystals in 6.11 wt.% NaCl solution.

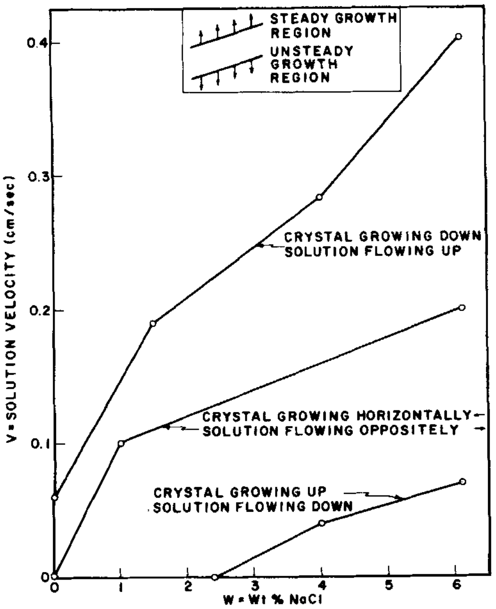

Figure 9. Topographical diagram of forced velocity required for steady crystal growth.

tion. Also for the same growth direction, higher velocities are required for the more concentrated solutions. It can thus be concluded that: (a) natural convection effects become stronger as the salt concentration increases; (b) natural convection is strongest in downward growth and weakest in upward growth; and (c) the net effect of natural convection in the region where mass and thermal natural convection oppose each other (>2.4% NaCl) is stronger than when they cooperate (<2.4% NaCl). This curious result is concluded because the former situation always requires higher forced velocities to overcome the natural convection in order to get steady growth. It may be that this just shows that mass natural convection is more important than thermal convection, especially in more concentrated solutions.

MATHEMATICAL ANALYSIS OF ICE GROWTH WITH NATURAL CONVECTION

Huang (1975) has made an analysis of the growth rate of ice crystals in pure water in which this rate is assumed to be controlled by thermal natural convection and the following additional assumptions are made:

1. The growth is upward with respect to gravity.
2. The viscosity and thermal conductivity are constant.
3. The crystal itself is all at one temperature.
4. Boundary layer approximations are made.
5. The growth rate is steady.
6. No forced velocity is applied.

The derivation followed closely that of Ostrach (1964).

The model of the growing ice crystal assumes that it grows as fast as the heat of fusion liberated at the tip can be dissipated into the surrounding solution. The crystal is assumed to be a two-dimensional parabolic cylinder (a drawing of the geometry is presented in Vlahakis, 1974). The heat balance at the growing tip yields $v\rho_i\Delta H_f$ = heat flux due to growth $= -k(\partial t/\partial x)$ where the temperature gradient is in the fluid at the ice-fluid interface. The rest of the analysis involves finding the temperature distribution in the solution for some assumed fluid mechanical regime, i.e., forced or natural convection. This allows calculation of the gradient which then can be put in the above heat balance and solved for v, the growth velocity, yielding $v = f(\Delta T, R)$ where R is the radius of the nose of the parabolic crystal. R can be eliminated, since it is not an independent variable (i.e., cannot be set experimentally) by maximizing v with respect to R to get:

$$v = 0.2689 \frac{k(Pr\beta g/\gamma T_M)^{1/4}}{(\rho_i \Delta H_f)^{3/4} \nu^{1/2}} \cdot \Delta T^{3/2} \tag{1a}$$

which for the ice-water system with $\gamma = 50$ mJ/m^2 (= 50 dyne/cm) is equivalent to

$$v(\mathrm{cm/s}) = 7.89 \times 10^{-4}\, \Delta T^{3/2} \tag{1b}$$

The maximizing of v with respect to R yields:

$$R = \frac{6\epsilon}{\Delta T} \tag{2}$$

which is exactly twice the R obtained by the analysis of Fernandez (1967) for forced convection heat transfer. Kallungal (1975) measured some radii of the tip and found them to be about four times larger than those predicted by Fernandez for forced convection over the growing crystal. The present analysis for natural convection makes Kallungal's measurements only about two times larger than predicted and thus a little more credible, or at least not so incredible. In any case, both the analysis and direct observation agree firmly that the radius of curvature of the parabolic cylinder at the tip of the growing crystal varies inversely with ΔT.

A more complete analysis for both thermal and mass natural convection simultaneously operating and controlling the growth rate, also was made by Huang (1976) with the additional assumption: The body force effects in the force balance are due to both temperature and concentration gradients creating density differences in the fluid with

$$\rho = \bar{\rho}[1 + \beta\Delta T + \beta'\Delta C] \tag{3}$$

Otherwise the fluid density is considered constant at $\bar{\rho}$. The other assumptions in this analysis are the same as those made in deriving Eq. 1a above.

The moving boundary is not taken into account. The final expression is:

$$v = 0.354 \frac{kPr^{1/3}(g/\gamma T_M)^{1/4}\,(A_1\beta + A_2\beta' m W_\infty)^{1/4}}{(\rho_i \Delta H)^{3/4}\, \nu^{1/2}\,(1 + \lambda m W_\infty)^{5/4}} \cdot \Delta T_s^{3/2} \tag{4}$$

$$\text{The constant} = \frac{5 \times 10^{1/4}}{12\left[\Gamma\left(\frac{1}{3}\right)\right]^{3/4}} = 0.35385$$

The tip radius R for this analysis agrees exactly with Eq. 2 above.

Differentiating v with respect to W_∞, and setting the derivative = 0, predicts the maximum growth rate to occur at

$$W_\infty = \frac{1}{4\lambda m} - \frac{5}{4}\frac{A_1\beta}{A_2\beta' m} \cong 0.86 \text{ mass \%} \tag{5}$$

Previous models not incorporating mass natural convection predict monotonically decreasing growth rates with increasing salt concentration. If we allow $W_\infty \rightarrow 0$ (pure water), Eq. 4 reduces to Eq. 1.

It is also interesting to note that both the cases of forced convection (Fernandez, 1967) and this case of natural convection predict the growth rate to vary as the 3/2 power of the subcooling, but our experimental data confirm the 3/2 power only for forced convection. For natural convection from our experiments the exponent is nearer to 2.0. Equation 4 predicts correctly the maximum growth rate to occur at about 0.9 mass % salt. It also predicts the ratio of $v_{\max}/v_0$ to be 1.72 independent of the subcooling. This 72% increase is substantial but not as great as we observed. Also, the growth rates predicted by Eq. 4 are about one order of magnitude below those observed (between 0.5 and 1.5 orders); see Figure 11.

So Equation 4 has some virtues: It predicts and explains the anomalous growth rates we have puzled over for so long. It predicts a maximum rate of growth to occur at about 0.9 mass % NaCl correctly. And it thus tends to reinforce our contention that natural convection of both mass and heat are operative in ice crystal growth.

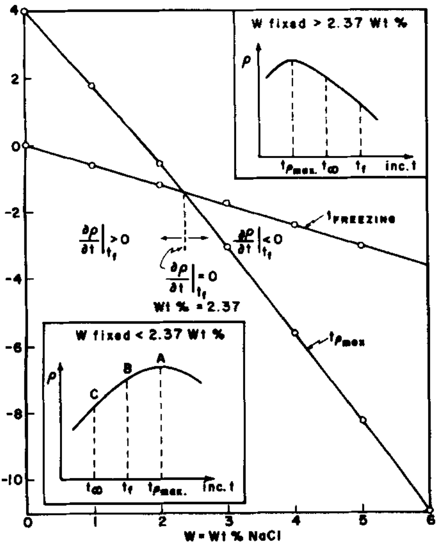

Figure 10. Freezing and maximum density temperatures of NaCl solutions.

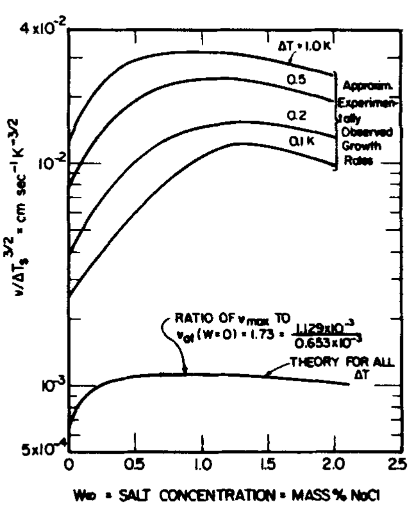

Figure 11. Comparison of theory (Eq. 4) with observed rates for ice crystals growing upward in quiescent NaCl solutions.

Additional analytical work that might be done would be to account for the moving boundary, but this is not expected to change the results in any substantial way, according to a private communication from W. N. Gill of the State University of New York at Buffalo. Also, one could treat the case where both forced and natural convection are operative.

We observed single ice crystals to grow steadily in the vertically upward direction in quiescent solution ($V = 0$) containing less than 2% NaCl. However, no steady state growth rates are observed in quiescent solution either (a) for vertically upward growth in 4 or 6% NaCl solutions, or (b) for vertically downward or horizontal growth in all NaCl solutions. The explanation for this complex but interesting observation lies in the peculiar density of salt solutions and its effect on natural convection induced by the temperature and concentration gradients created by the growing ice crystal.

The phenomenon of accelerated crystal growth rates in quiescent or slow-flowing NaCl solutions over those in distilled water has been investigated thoroughly again. It will be observed that the growth enhancement is a function of forced velocity and that it vanishes at solution velocities greater than about 10 cm/s. This also shows the role of natural convection in the enhanced growth phenomena previously called "anomalous."

EFFECT OF THERMAL AND MASS NATURAL CONVECTION ON ICE CRYSTAL GROWTH IN QUIESCENT OR SLOW-FLOWING NaCl SOLUTIONS

When a single ice crystal grows in any orientation in subcooled NaCl solutions, rejection of salt and release of heat of crystallization proceed continuously, thus disturbing the bulk salt solution and generating both a concentration and a temperature gradient. Two categories of transport mechanism will result, namely, thermal and mass diffusion from the gradients, and thermal and mass natural convections due to the density differences caused by both gradients. The net heat and mass transfer will depend on the combination of these mechanisms. The transport of the disturbed solution due to each individual mechanism can be described as follows: The diffusion fluxes are of course always in the direction opposite to the gradients and tend to reduce the crystal growth rate. The mass natural convection alone will always cause downward fluid flow in free space, since NaCl solutions of higher concentration are always more dense and tend to sink. The flow direction induced by the thermal natural convection, however, will depend upon whether the maximum density of a salt solution occurs at a temperature above or below its freezing point, because, unlike the monotonically increasing density-concentration relation, the density-temperature curve for a given NaCl solution has a maximum, as does pure water. The data on freezing points, t_f, and temperature of maximum density, $t_{\rho\max}$, in NaCl solutions (I.C.T., 1928) are plotted on Figure 10. Note that the freezing points are above or below the maximum density temperatures when the solutions contain more or less than 2.37 wt. % NaCl, respectively. In the ice crystal growth study the temperatures of the bulk NaCl solutions in the growth tube are always lower than the freezing points, since there must be a finite subcooling required to obtain any growth. From the lower subsidiary diagram in Figure 10, it follows that subcooled solutions containing less than 2.37 wt. % NaCl will have the property that below t_f, $d\rho/dt > 0$; i.e., the warmer the solution, the more dense it is. Therefore, the solution current due to thermal natural convection will be downward at these concentrations. On the other hand, subcooled solutions containing more than 2.37 wt. % NaCl show that below t_f (but above $t_{\rho\max}$), $d\rho/dt < 0$; i.e., warmer fluid has lower density and tends to rise. A brief summary of the results of the foregoing arguments is shown in Table 2.

The critical NaCl concentration of 2.37% is not an exact figure, and one should not place too much reliance on its precise value because the precision of the data on maximum density is not known. Even if the data were precise, the growth of ice from solutions takes place within a small range of salt concentrations. There are gradients of concentration with ice growth; at the maximum density point $d\rho/dt$ is zero, and it is very small close to either side of this point. This creates only feeble thermal natural convection currents. We can say, however, that at 2% NaCl the direction of these currents is opposite to that at 3% NaCl.

Similar data on sea salts are also available from the same source (I.C.T., 1928) as on NaCl, and these show that the critical sea salt concentration for $d\rho/dt = 0$ at the freezing point of sea water occurs at 2.8% sea salt. Since normal sea water contains 3.5% salts, we would expect normal or more concentrated sea water to behave qualitatively like 4 and 6% NaCl and not like 1 or 2% NaCl solutions.

With the aid of Table 2 an explanation of the steady and unsteady quiescent vertical growth phenomena can be made based

TABLE 2. NATURAL CONVECTION EFFECTS IN VARIOUS NaCl SOLUTIONS

		Concentration, W (wt. % NaCl)	
		<2.37	>2.37
$d\rho/dt\vert_{t_f}$		Positive	Negative
Solution Flow Direction due to Natural Convention	Warmer Fluid	Downward	Upward
Effect of	More Concentrated Fluid	Downward	Downward

on the combined effect of natural convections. When single ice crystals grow in quiescent subcooled solutions containing about 2 wt. % NaCl or less, the warmer and more concentrated solution created due to the release of heat and the rejection of salt during crystal growth will sink, since in this concentration range both thermal and mass natural convections will carry the disturbed solution downward. Therefore, when single ice crystals grow vertically upward, *the growing crystal tip will be in continuous contact* with fresh solutions at t_∞ and W_∞ during its growth and thus show steady growth. Conversely, when single ice crystals grow vertically downward, the crystal tip will grow into a warmer and more concentrated region and the growth rate will decrease since both the concentration increase and rise in temperature will reduce the degree of subcooling of the solution. (When the concentration of a subcooled NaCl solution is increased, its freezing point will be depressed and the degree of subcooling reduced.)

For crystal growth in subcooled quiescent 4 or 6 wt. % NaCl solutions, the thermal and mass natural convections will tend to carry the disturbed solution in opposite directions, i.e., the thermal effect will be upward and mass effect will be downward. The final direction of the disturbed solution flow will depend upon the relative strength of the thermal vs. mass natural convection effects. If the natural convection of mass dominates that of thermal effect, the disturbed solution will sink; and vice versa. However, when the *ice crystals grow in subcooled quiescent 4 to 6 wt. % NaCl solutions*, experiment shows unsteady results for both upward and downward growth. This implies that when ice crystals grow upward, the thermal natural convection dominates, and when they grow downward the mass effect dominates. An estimation based on boundary-layer and stationary interface assumptions shows that the density difference caused by concentration gradients is much stronger than that caused by thermal gradients (Huang, 1975). This apparent contradiction cannot be explained by the primitive qualitative picture of natural convection presented. However, the relative strength of the thermal and mass natural convection will possibly be influenced by the geometry of the apparatus and the moving boundary effect at the crystal tip.

SUMMARY

We have determined and presented many new data on the growth rates of ice in salt solutions as a function of the subcooling, the forced velocity (including quiescent solutions), and the % salt in solution, when the direction of growth was upward, downward, and horizontal with respect to gravity. Under several conditions no steady growth was observed.

The lack of steady growth has been explained by a new analysis which shows that for solutions of NaCl of more than 2.37 mass % (or sea salts of more than 2.8%) the results should be quite different from ice growth in solution less concentrated than this.

An analytical solution for natural convection control of growth rates predicts correctly a maximum growth rate as a function of salt concentration at about 0.9 mass % NaCl. This reinforces the idea that natural convection is important in crystal growth.

ACKNOWLEDGMENT

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NOTATION

A_1 $= (1/2)Sc^{-1/3} - (1/6)Sc^{-2/3}Pr^{1/3}$
A_2 $= (1/3Sc)^{-1/3}$
D = diffusivity of NaCl in water, cm^2/s
g = gravity, m/s^2
ΔH_f or ΔH = heat of fusion of ice, J/kg
k = thermal conductivity of water or salt solution, J/s·mK
m $= (k/D)(1/\rho_i \Delta H)(Pr/Sc)^{1/3}$, dimension = K^{-1}
Pr = Prandtl number = $Cp\mu/k$
R = radius of nose of growing crystal, m
Sc = Schmidt number = ν/D
t_f = freezing temperature of ice on a flat surface, $R \rightarrow \infty$
$t_{\rho\max}$ = temperature of maximum density
T_M = freezing point of pure solvent
T_i = temperature at interface, K
ΔT or ΔT_s = subcooling, K
v = growth rate, cm/s
V = forced velocity, cm/s
W = mass % NaCl in solution

Greek Letters

β = temperature coefficient of density $(1/\rho)[(\partial\rho/\partial t)_{P,W}]_{T\infty,W\infty}$
β' = concentration coefficient of density $(1/\rho)[(\partial\rho/\partial W)_{P,T}]_{T\infty,W\infty}$
γ = interfacial tension between solution and ice, N/m or J/m^2
ϵ $= (\gamma T_M)/(\rho_i \Delta H)$, m·K
λW_i = depression of freezing point at interface
λW_∞ = decrease in freezing point due to salt, K
λ $= (\lambda W_\infty)/W_\infty$, dimension = K/mass %
μ = absolute viscosity, kg/ms
ν = kinematic viscosity = μ/ρ, m^2/s
ρ = solution density
ρ_i = ice density

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