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Experimental Study of Freezing of Binary Solutions

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An experimental investigation of the freezing of hypoeutectic aqueous sodium chloride (20%) solutions in the presence of buoyancy driven thermosolutal convection is reported. The freezing was induced on one of the vertical walls of a rectangular cavity. The variation of temperatures and concentrations have been recorded. During freezing some of the rejected solute settled at the bottom of the melt forming a solutally and thermally stable, stagnant region ("stratified region") and the convective currents were confined to the upper region ("convection region"). In the stratified region, the temperatures show a decreasing trend followed by an increasing (i.e., reheating) trend while the concentrations increase with time. In the convection region for a lower initial superheat of 10°C, the temperatures show a continuous cooling trend (with no reheating), while for an initial superheat of 20°C there is slight reheating. However, the concentrations in the convection region are fairly uniform and constant.

Nomenclature

C = concentration in percent by weight
T = temperature, K
t = time, s
x = horizontal distance measured from cold wall, m
y = vertical distance measured from bottom of test cell, m

Subscripts
c = cold wall
f = fusion
h = hot wall
i = initial

Introduction

THE phase change (solidification and melting) of multicomponent solutions involves the flow of heat, fluid, and species transport processes. The occurrence of these processes in many natural and artificial systems spanning diverse fields has provided the impetus to study them. The cooling of magma chambers,[1] solidification of earth's inner core,[2] manufacture of castings and ingots,[3] and welding and soldering[4] are a few examples. The solidification of multicomponent solutions involves simultaneous interaction of buoyancy driven currents generated and sustained by thermal and concentration gradients, thereby making the thermophysical phenomena more complex than in systems in which there is only one driving force.

In general, the solidification of multicomponent solutions is characterized by the presence of three regions: 1) solid—where heat transfer is by conduction; 2) mush—which consists of frozen solid particles intermixed with liquid, thus both conduction and convection heat transfer may be present; and 3) liquid—where bulk natural convection currents are present.[5] It has been recognized that natural convection currents influence the rate of solidification and the physical structure of the solidified material.[5] But the effects of thermosolutal convection have not been clearly and thoroughly identified.

In order to understand and to be able to describe the complicated processes occurring in many systems involving the solidification of multicomponent solutions, it is imperative to have a clear and comprehensive appreciation of the problem. In particular, from a designer's or manufacturer's viewpoint, it is necessary to know how the solute and heat are transported to effect the formation of substances with different solute concentration at varied temperatures. The properties (structural, thermophysical, electromagnetic, and acoustic) of the solidified product, and therefore, its performance are influenced by the redistribution of the heat and mass.

At the lower end of the spectrum of multicomponent solutions is the binary solution. The solidification of binary solutions has been studied mainly by metallurgists, with application to the manufacture of alloy castings. Both alloys like Al-Cu and Pb-Sn and nonmetallic solutions like NH_4Cl, Na_2CO_3 and NaCl have been investigated. In particular, Terwilliger and Dizio[6] have investigated the salt rejection phenomena during the freezing of saline solutions in a cylindrical tube. The solute concentration was measured using microconductance probes. They found that the liquid interface concentration, controlled by constitutional supercooling, and the thermal driving potential for freezing are two important parameters influencing the redistribution of solute. The freezing of aqueous sodium chloride solutions from the top surface of a rectangular cavity has been experimentally studied by Grange et al.[7] using an interferometer. They observed thermosolutal convection in the form of salt fingers. At the bottom of the liquid region, the liquid settled with a stable density distribution due to solute redistribution but had an adverse temperature gradient. Hayashi and Komori[8] analyzed the freezing of aqueous salt solutions in small cubical cells. Their model considered the freezing process as freezing of a semi-infinite body and Neumann's solution with an approximate heat capacity was used. Good agreement was obtained between the predicted and measured experimental temperatures. Chellaiah and Viskanta[9] investigated the freezing of a water-NaCl solution on a vertical wall of a rectangular cavity. They presented detailed information about flow patterns in the bulk melt and morphology of the liquidus at different times. Braga and Viskanta[10] experimentally and theoretically investigated the solidification of an aqueous salt solution on a horizontal wall in the absence of natural convection. Good agreement was obtained between

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the predicted and measured temperatures. They found that concentration was as important as the initial and wall temperatures as far as the growth of the mushy zone was concerned.

Despite the work that has been performed to date, few studies have dealt with freezing of aqueous salt solutions on a vertical wall of a rectangular cavity. The transport of heat and mass due to thermosolutal convection has not been thoroughly investigated in these previous studies. Experimental studies of salt segregation in the freezing of NaCl solutions are only sparsely reported in the literature. This article focuses on the redistribution of solute (NaCl) and heat rejected during freezing due to the thermosolutal convective currents. The freezing of aqueous NaCl on one of the vertical walls of a rectangular cavity with its top and bottom surfaces insulated is considered.

Experimental Apparatus

The experiments were conducted in a rectangular Plexiglas® test cell, with inner dimensions of 15-cm in height, 19-cm in length and 20-cm in depth (see Fig. 1). The use of Plexiglas allowed for visual and photographic observations during the freezing process. Two copper heat exchangers constituted the vertical walls of the rectangular cavity. For the experiments in this study the test cell was filled with solution to a height of 9.5 cm. Therefore, an aspect ratio (height/length) of 0.5 was obtained. The heat exchangers were bonded to the Plexiglas with silicone glue. The silicone prevented the leakage of solution and was flexible enough to maintain a seal as the cavity experienced thermal contraction during the experiment. Coolant from two temperature-controlled baths was circulated through the serpentine counter-current channels in the heat exchangers. In this way, the temperature of the heat exchanger was held constant. Three Copper-Constantan thermocouples were embedded in each heat exchanger, close to the surface exposed to the solution, to monitor its temperature. The entire test cell was insulated with 5-cm thick Styrofoam® to minimize heat gain from the ambience. A removable Styrofoam plug was located in the front of the test cell for photographic and visual observations of the freezing process. A flourescent light (30-cm in length) was placed on the top, front surface of the test cell for illuminating the solution during photography. The entire assembly was placed on a steel plate fitted with four leveling screws to ensure that the test cell was level. Therefore, the effects of inclination on the freezing process were eliminated. Nineteen Chromel-Alumel immersion thermocouple probes were inserted into the test cell through a rigid top plate (Fig. 1). All thermocouples (including those in the heat exchangers) were connected to a Fluke Helios I data acquisition system controlled by a personal computer. Furthermore, all thermocouples were calibrated to an accuracy of ±0.1°C.

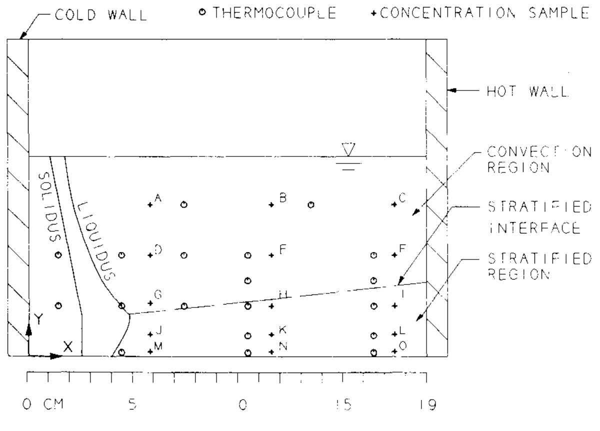

Fig. 1 Schema of experimental apparatus (note: the points A, D, G, J, and M were approximately 1 cm ahead of the point of maximum freezing on the liquidus. Thus, these locations moved as freezing progressed while others remain fixed).

Experimental Procedure

Degasified, deionized, distilled water and research grade sodium chloride was used to prepare the solution for each experiment. Using a laboratory balance, the appropriate mass of sodium chloride for a specific mass of water was weighed and carefully mixed with water. After mixing, the concentration of the solution was checked using a refractometer. Additional salt or water was added to obtain the desired concentration. The solution was then carefully siphoned into the test cell. During the entire preparation and filling process, violent agitation of the solution was avoided to prevent the entrapment of air in the solution.

The solution was then cooled to the initial temperature using two of the temperature-controlled circulating baths (Lauda Brinkmann RC6 and RK20). The cooling process typically took 12–14 h and was monitored by periodic temperature measurements to ensure that a uniform and steady temperature was achieved in the solution. Simultaneously, coolant (mixture of methanol and water) in a third bath (NESLAB ULT-95DD) was cooled to the desired cold wall temperature. After the solution in the test cell attained the desired initial temperature, freezing was initiated by circulating coolant from the third bath to one of the heat exchangers. The other heat exchanger (hot wall) was maintained at the initial temperature.

Hypodermic needles were used to extract small quantities of solution during the experiment from various locations in the test cell (Fig. 1). Using a refractometer, the solute concentration of the samples was determined. The accuracy of the refractometer was determined to be ±0.001 for refractive index which provides an accuracy of approximately ±0.5% for solute concentration. Photographs of the solid, mush, and liquid regions were taken periodically using a Nikon FE3 35-mm camera and Kodak 400 ASA Tmax film. The photographs were developed and printed to scale. Using a planimeter, the areas occupied by the solid and mush regions were determined. Then knowing the depth of the test cell and the scale of photography (in comparison with the size of test cell), the volume fractions were calculated. Some experiments were repeated to ensure consistency in data.

Results and Discussion

The solidification of binary solutions (of noneutectic composition) is different from that of pure substances, in that, in addition to the pure solid and pure liquid (bulk melt) region, there is a mushy region (where both solid and liquid phases coexist) between the solid and the liquid.[5] The size and shape of the three regions are influenced by the initial concentration, cold wall temperature, and initial superheat. Here, as a prelude to the discussion of experimental results, the freezing of aqueous sodium chloride is briefly reviewed. For a detailed discussion the reader is referred to Refs. 11 and 12.

Aqueous sodium chloride solution has an eutectic composition of 23.3% by weight and eutectic temperature of −21.12°C.[13] During the solidification of a hypoeutectic aqueous sodium chloride solution, the solute (sodium chloride) is rejected at the advancing freezing front (solidus). This solute dissolves in the adjacent solution, increasing its concentration. Thus, a concentration gradient is established which induces natural convective flow due to solutal buoyancy forces. Additionally, the thermal gradient between the hot wall and the freezing front causes thermally driven natural convective flow. The solution near the solidus is cold and solutally rich, whereas that near the hot wall is hot and solutally lean. Therefore, the thermal and solutal natural convective currents are in the same direction and augment each other.

The solution rises along the hot wall, turns toward the cold wall, and impinges on the top surface, thereby reducing the local rate of freezing. Then as it flows downward along the solidus/liquidus, its temperature decreases (due to cooling) while its concentration increases (due to absorption of the rejected solute). Some of this cold, high concentration liquid, that is too heavy to be convected settles at the bottom of the test cell. The increase in concentration lowers the freezing point and so the rate of freezing near the bottom of the test cell is also reduced (as the cold wall temperature remains constant).

As solidification progresses, more cold, solute-rich solution settles at the bottom of the test cell, eventually forming a stagnant region where no convective flow is present. For ease of discussion, this region is referred to as the stratified region while the solution above it where natural convective flow is present is called the convection region. A thin interface, about 3–4-mm thick, separates the two regions and extends from the hot wall to the liquidus. (It is recognized that actually it is a thin zone that separates the two regions, but because the sizes of convection and stratified regions are much bigger than this separating zone, the term "interface" is being used). The interface meets the liquidus at a distinct ridge, which will be called the "chin." This ridge was located no more than 5 mm above the location on the liquidus where the rate of freezing was maximum. The thickness of the stratified region increased from the liquidus to the hot wall. The morphology of the liquidus (not shown here for brevity) at later times in the experiment clearly indicates the development and advancement of the chin. With the growth of the stratified region, the chin advances upwards and outward from the cold wall. The morphology for similar experiments is discussed in Refs. 11 and 12.

The results presented in this article are part of a larger study of solidification of binary solutions under different experimental conditions. Due to space constraints, only three experiments (all with an initial concentration of 20%) are selected to isolate and study the effects of a few parameters on the freezing process. The experimental conditions are presented in Table 1. The last two columns in the table are the initial superheat and the initial thermal-driving potential for freezing. The conditions were chosen so that the effect of varying the thermal driving potential for freezing (experiments 1 and 2) with nearly the same initial superheat and the effect of varying initial superheat (experiments 2 and 3) with nearly the same driving potential for freezing could be studied. It should be noted that superheat and the driving potential do not remain constant throughout the experiment. Although the hot and cold wall temperatures do not change, the freezing point varies with concentration, which is in contrast to the solidification of pure substances. The volume fraction, solute redistribution, and vertical temperature distributions are presented in the following subsections.

Volume Fraction

The volume fraction of solid (or mush) is the ratio of the volume of solid (or mushy) region to the total initial volume of solution in the test cell. The volume fraction of solid is the same as its frozen fraction, whereas for the mushy region, the amount of frozen material cannot be determined. Therefore, the frozen fraction in the mush cannot be calculated and the volume fraction will differ from the frozen fraction. Two volume fractions, one corresponding to the solid and other to the combined solid and mush are presented in Fig. 2 as a function of time for all the experiments. For a given experiment, at any time the difference between the two volume fractions is a measure of the volume of mush.

It is seen that initially, the rate of freezing is high and the amount of mush is small. With the progress of solidification, the rate of freezing slows while the amount of mush increases. The reason for this is as follows. As solidification is initiated impulsively by circulating cold coolant through one of the heat exchangers, the solution near it freezes very rapidly forming a solid region. However, it takes some time for the mush to form and natural convective currents to be established. Because the effect of convective flow is to retard the freezing process (as in the case of freezing of pure substances) the rate of freezing decreases at later times.[9,11,12]

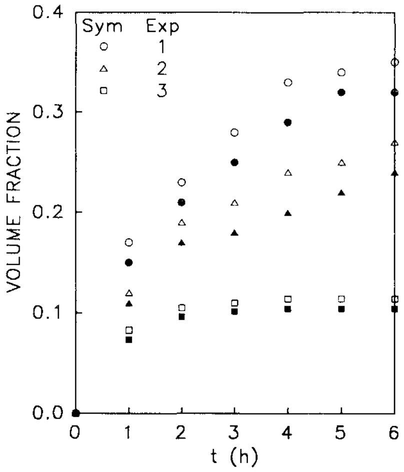

Fig. 2 **Variation of volume fraction with time (note: filled symbols correspond to solid region and open symbols correspond to solid and mush regions combined).**

After about 4 h, the ratio of the volume fractions for mush to the solid remains fairly constant. On average (average of values at 4, 5, and 6 h) for experiments 1, 2, and 3 the solid constitutes about 91.1, 86.7, and 83.3% of the combined solid and mush regions, respectively. In experiments 1, 2, and 3 the initial superheat was 10.3, 9.8, and 20.1°C, and driving potential for freezing was 49.2, 35.8, and 31.1°C, respectively. Experiment 2 had the least amount of initial superheat, while experiment 3 had the least amount of thermal driving potential for freezing. At any time in experiment 1, the volume fraction (corresponding to either solid or solid and mush combined) was the largest in experiment 1 and the smallest in experiment 3.

With an increase in superheat the longitudinal temperature gradient in the bulk melt increases and the convective flow is strengthened. This enhances heat transfer and subsequently, higher temperatures are found in the bulk melt; thus, the rate of freezing is reduced. As expected, with increased thermal driving potential, the freezing progresses faster. However, an increase in superheat of 10.3°C has significant effects on the volume fraction (refer to experiments 2 and 3 in Fig. 2). Whereas, an increase in thermal driving potential of 13.4°C increased the volume fraction by a small amount (refer to experiments 1 and 2 in Fig. 2).

Solute Redistribution

Every hour samples of solution were extracted from different locations. Some locations showed an increase in concentration, while others remained constant or indicated a slight decrease. It was found that the concentration of solution in the bulk melt is primarily a function of height (y) with negligible variation longitudinally. Therefore, the concentration data obtained at sample locations of the same height were averaged, which also served to minimize experimental errors. Figures 3a–c show the solute distribution in the bulk melt as a function of time for experiments 1, 2, and 3, respectively. The concentrations are plotted in a dimensionless form $(C - C_i)/C_i$. Thus, negative values indicate depletion of solute and positive values denote gain of solute.

The general trend found in the concentration figures is the stable stratification of concentration in the bulk melt (i.e., the concentration decreases with increase in height). The maximum gains in concentration were 14.2, 14.7, and 11.1% for experiments 2, 1, and 3, respectively. Note that the data from experiments 1 and 2 do not differ significantly indicating that the 13.4°C increase in thermal driving potential had a negli-

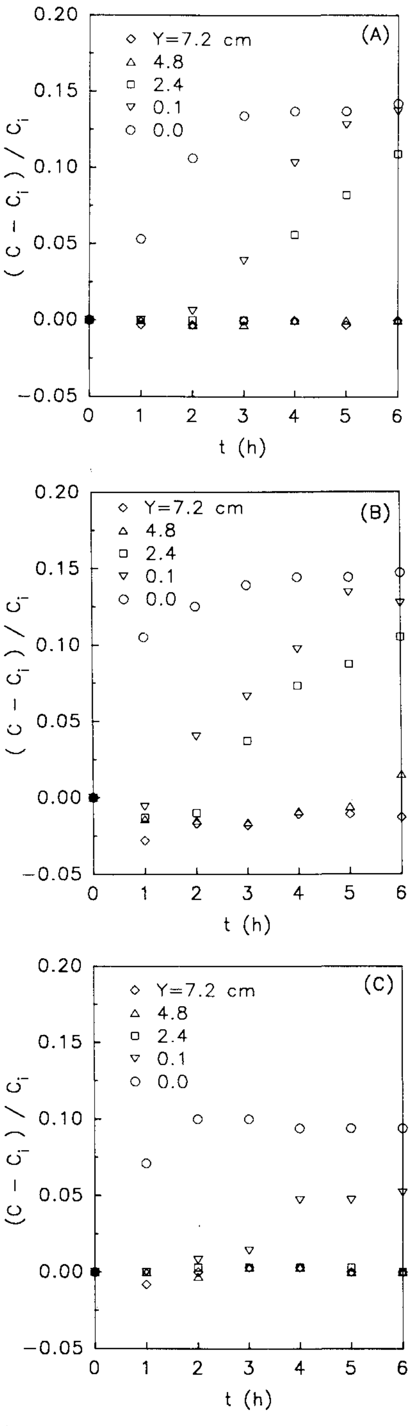

Fig. 3 Vertical concentration distribution at x = 10.5 cm for experiments: a) 1, b) 2, and c) 3.

gible effect on the amount of solute settling in the stratified region.

The redistribution of solute is directly related to the growth of the stratified region and the location of the thin interface separating the convection and stratified regions. Figures 4–6 show the location of the thin interface at the liquidus (which is also the location of the chin) at different times in the three experiments. The location also corresponds to the minimum height of the stratified region. From this figure, it is evident that the stratified region was the smallest in experiment 3, which had the largest amount of superheat (Table 1). With increase in superheat, the convective currents are stronger and the rate of freezing is reduced (Fig. 2). The reduction in the rate of freezing causes less solute to be rejected into the bulk melt. Hence, less cold and solute-rich solution exists in the liquid region. As the stratified region is formed by the settling of this cold and solute-rich solution, a decrease in its availability reduces the size of the stratified region. For this reason, the smallest increase in solute concentration from initial value was observed in experiment 3 compared to other two experiments which have less initial superheat.

Comparing experiments 1 and 2, experiment 1 has a 13.4°C higher driving potential and approximately the same initial superheat as experiment 2. Thus, experiment 1 has a slightly higher rate of freezing than in experiment 2 (Fig. 2). The growth of the stratified region is approximately the same for both experiments (Figs. 4 and 5) and the solute distributions are also similar. This further supports the correlation between solute redistribution and the growth of the stratified region.

Vertical Temperature Distribution

As mentioned earlier, the pure solution consists of two separate regions: 1) stratified region, and 2) convection region. In the stratified region convective flow is not present and conduction is the only mode of heat transfer. In the convection region natural convective currents are present. A thin interface that is about 3–4-mm thick, separates the two regions. The stratified region is bounded by the bottom of the test cell, the liquidus, hot wall, and the thin interface at the top. The convection region is bounded by the liquidus, hot wall, the thin interface at the bottom, and the free surface at the top.

As freezing progresses, the liquidus advances reducing the width of the two regions. Simultaneously, the stratified region moves upwards and increases in height. Consequently, some thermocouples that were earlier in the path of convective flow get engulfed by the stratified region. These thermocouples then indicate a local temperature increase or "reheating" (i.e., the temperature distribution as recorded by these thermocouples show cooling followed by heating). It is surmised that in the absence of convective flow there is heat transfer by conduction from the hot wall into the stratified region. It is the presence of the two regions in bulk melt with their different flow regimes and solute concentration distributions which directly affects the temperature distribution in the solution. Thus, the bulk melt is a region of two moving boundaries, the advancing liquidus, and the rising thin interface.

Figures 4–6 show the vertical temperature distribution at hourly intervals at a typical location, x = 10.5 cm for experiments 1, 2, and 3, respectively. In order to provide visual clarity, for each experiment the data corresponding to the first 3 h are shown in one figure and those for the next 3 h are shown in a separate figure. As freezing progresses, global cooling takes place in the bulk melt. But after some time, reheating occurs in the stratified region and eventually in the lower portions of the convection region. This reheating causes the data points to move upward in the figures back on top of earlier data points. To avoid cluttering of the data two figures were used for each experiment. It should be noted that the choice of placing data from 1–3 h in one figure and 4–6 h in another is not meant to imply that this reheating occurs between 3–4 h in all the experiments.

In the first 3 h of experiment 1 (Fig. 4), the temperatures at all locations show continuous cooling. But as time progresses, the temperatures at locations y = 0.1, 1, 2.4, and 3.6 cm increase with time while the cooling trend continues at higher locations. This can be explained by considering the growth of the stratified region. The height of the stratified region increased from a minimum at the liquidus to a maximum at the hot wall, i.e., it was approximately 0.7–1 cm higher at the hot wall than at the liquidus. At t = 4 h, the minimum thickness was 3.80 cm. Hence, an approximate average value for the thickness of the stratified region would be 4.20 cm. From Fig. 4, it is evident that the reversal in temperature trends (continuous cooling against reheating) occurs between 3.6–4.8 cm. Thus, it can be inferred that reheating is experienced by the thermocouples that are engulfed

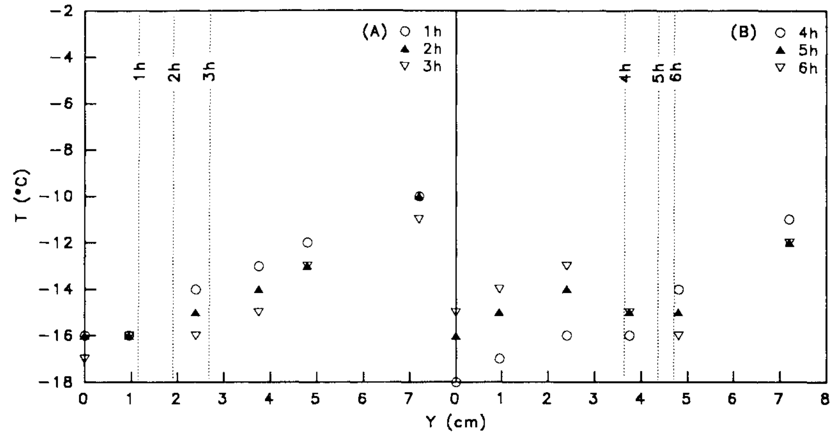

Fig. 4 Minimum height of the stratified region and vertical temperature distribution for experiment 1 at a) 1–3 h, and b) 4–6 h.

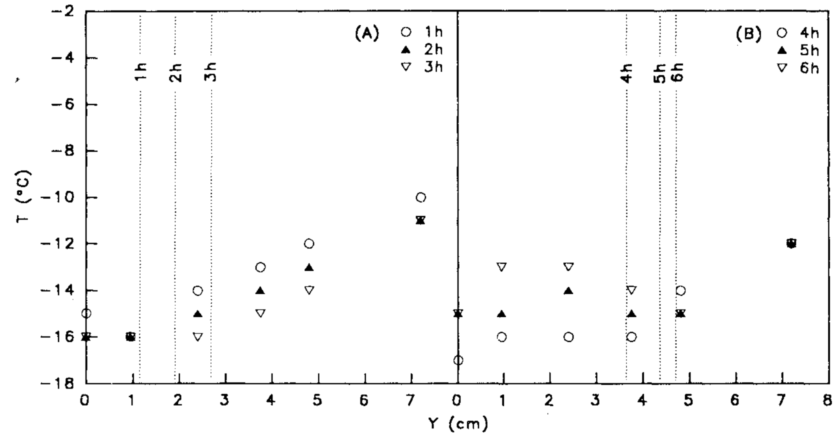

Fig. 5 Minimum height of the stratified region and vertical temperature distribution for experiment 2 at a) 1–3 h, and b) 4–6 h.

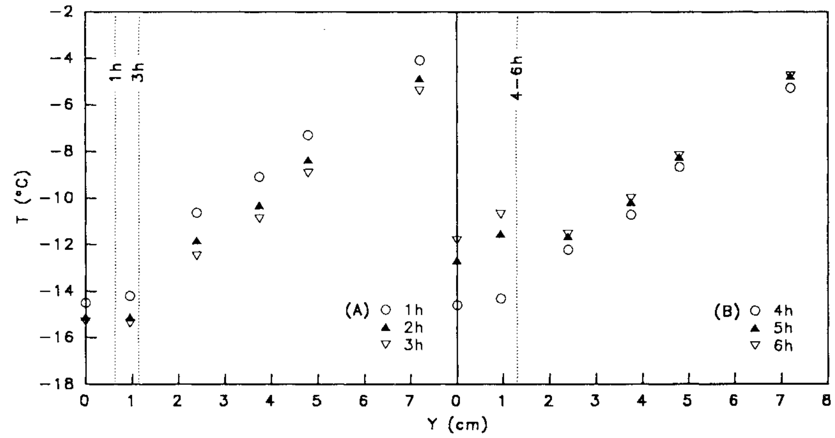

Fig. 6 Minimum height of the stratified region and vertical temperature distribution for experiment 3 at a) 1–3 h, and b) 4–6 h. (Note: the height at 2 h is not shown to avoid cluttering of lines).

by the growing stratified region. The thermocouples at higher locations remain in the convection region throughout the experiment and therefore experience continued cooling. The data in experiment 2, (Fig. 5), show trends similar to those in experiment 1. The concentrations in the stratified regions are only slightly different (Fig. 3).

In experiment 3 (Fig. 6), the continued cooling and reheating trend occurs for all the data points, i.e., reheating is observed both in the stratified and convection regions. The reason for this is unclear. One of the possible explanations is that the higher superheat in experiment 3 enhances heat transfer between the reheating stratified region and the convection region at the bottom, thus causing a bulk reheating of convection region. This phenomenon is under further investigation by the authors.

The effect of increased superheat can be inferred by comparing Figs. 5 and 6. The increase in superheat increases the vertical temperature gradient in the liquid region and across the test cell. At any time, the vertical temperature gradient in experiment 3 is about 1.5–2 times that in experiment 2. The increased superheat causes a stronger convective flow which exhibits a vertically stratified temperature distribution. This temperature distribution is similar to that seen in natural convective flow in rectangular cavities in the absence of phase change.

The effect of increased driving potential can be inferred from Figs. 4 (experiment 1) and 5 (experiment 2). The temperature profiles are similar and the values are very close. Increased driving potential increases the rate of freezing. With a larger frozen mass the size of the bulk melt is reduced. The

Table 1 Summary of experimental conditions

Experiment	C_i, %	T_f, °C	T_i, °C	T_h, °C	T_c, °C	$\lvert T_h - T_f \rvert$, °C	$\lvert T_c - T_f \rvert$, °C
1	20	−16.5	−6.2	−6.2	−65.7	10.3	49.2
2	20	−16.5	−6.7	−6.7	−52.3	9.8	35.8
3	20	−16.5	3.6	3.6	−47.6	20.1	31.1

bulk melt is contained by the hot wall, the liquidus, and two adiabatic horizontal surfaces. Thus, the liquid in the melt is bounded by the hot wall temperature and the local temperatures along the fusion front. Therefore, the convective currents in melt are not directly affected by the cold wall temperature. Its influence is felt only through the rate of freezing which influences the size of the bulk melt region. Since for both experiments 1 and 2 the rate of freezing (Fig. 2) and the size of stratified region (Figs. 4 and 5) are not significantly different, the strength of the convective flow is nearly the same in both the experiments. Hence, the temperatures are very close and the trends are similar.

Conclusions

The freezing of hypoeutectic, aqueous sodium chloride solutions on a vertical wall of a rectangular cavity was studied experimentally. The natural convective currents generated by the thermal and solutal gradients in the bulk melt augmented each other. As expected, an increase in initial superheat reduced the rate of freezing and an increase in driving potential increased the rate of freezing. With the progress of freezing, the solute rejected due to freezing dissolved in the solution. Some of this cold, solute-rich solution settled at the bottom forming a stagnant region (called stratified region) where there was no flow. The convective currents were confined to the liquid region (called convection region) above the stably stratified region. A thin interface (about 3–4-mm thick) separated the two regions.

In the stratified region concentration increased with time, the maximum increase being near the bottom of the test cell. In the convection region the solute distributions remained approximately constant. The height of the stratified region increased progressively from a minimum at the liquidus to a maximum at the hot wall. For an initial superheat of approximately 10°C the thermocouples in the convection region showed continuous cooling. Some thermocouples, whose locations which were initially in the convection region and later submerged into the stratified region, indicated cooling followed by reheating after entering the stratified region. This is due to the absence of convective flow which dissipates thermal gradients and by heat transferred by conduction from the hot wall. For an initial superheat of 20°C all thermocouples showed reheating at later times although they remained in the convective region. This was probably due to heat transferred by diffusion from the stratified region below. An increase in initial superheat of approximately 10°C affected solute redistribution and the height of the stratified region decreased significantly. Whereas, an increase in the thermal driving potential of about 13°C had a negligible effect on solute redistribution and the height of the stratified region. However, the increase in thermal driving potential did have a small effect on the rate of freezing and caused an increase in the total volume fraction.

The method of determining concentration by withdrawing samples is cumbersome and time-consuming. There is a need for developing nonintrusive or less intrusive methods for measuring concentrations and temperatures simultaneously at any location. Miniature sensors using microelectronic components that can detect both temperature and concentration variation should be researched. The mathematical models of this freezing process must consider the settling of solute rich region at the bottom and the formation of a stratified region. The convective flow occurs in a region (convection region) that is bounded by two moving boundaries—not including the free surface. For accurate modeling this aspect should be considered.

Acknowledgments

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