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Transient Solidification of a Binary Mixture in an Inclined Rectangular Cavity

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This paper reports the main procedures and results of an experimental investigation of the problem of transient freezing of a binary mixture in an inclined rectangular cavity. The mixture was a solution of NH_4Cl in water. One wall of the cavity was cold and the remaining walls adiabatic. Three different regions existed in the cavity during the solidification phenomenon. A solid region, a liquid region, and a mixed-phase region. The mixed-phase region consisted of a solid matrix saturated with liquid and was located between the solid and the liquid regions. Characteristic temperature distributions in the cavity for a host of inclination angles were obtained. The history of the solidification process was studied by tracing the evolution of the interface between the solid and the mixed-phase regions as well as the interface between the mixed-phase and the liquid regions. The natural convection flow in the liquid, driven by both temperature and concentration gradients, seriously affected the solidification process. An engineering correlation for the dependence of the solidified volume fraction of the cavity on time, inclination angle, and initial concentration is also reported in the course of the study.

Nomenclature

C	=	concentration of solute, wt %
C_i	=	initial concentration, wt %
C_p	=	specific heat, J/(kg·K)
H	=	height of the cavity, mm
L	=	latent heat of fusion
T	=	temperature, °C
T_{eut}	=	eutectic temperature, °C
T_i	=	initial solution temperature
t	=	time, s
V^*	=	ratio of the volume of the solid region to the volume of the solid plus mushy regions
V^{**}	=	ratio of the volume of the solid plus mushy regions to the total volume of the cavity
V_T	=	total volume of the cavity
y	=	distance from cold wall
α	=	thermal diffusivity
θ	=	inclination angle of the cold wall with respect to the horizontal direction, Fig. 4
τ	=	dimensionless time $\{[\alpha_s C_{ps}(T_i - T_{eut})]/(H^2L)\}t$

I. Introduction

SOLIDIFICATION of mixtures and alloys is a phenomenon that occurs in many industrial applications. To this end, casting and coating processes in the manufacturing and the materials industry, freezing of food products in the food industry, and crystallization processes in the chemical industry all involve solidification of mixtures or alloys. In contrast to the voluminous literature that exists on heat transfer and fluid mechanics during phase change of pure substances, studies on these transport processes in the solidification of mixtures and alloys are limited. Representative investigations on freezing of alloys in enclosed spaces are discussed next.

Chellaiah and Viskanta[1] studied experimentally the phenomenon of freezing of a water and sodium chloride solution on a vertical wall of a rectangular cavity. Bennon and Incropera[2] derived a set of macroscopic equations based on the "continuum approach" for binary solid-liquid phase change problems. The same authors[3] used the above mentioned model to solve numerically the problem of solidification of a binary alloy in a rectangular cavity cooled from the side. Hayashi and Komori[4] conducted experiments on the freezing of salt solutions in cells cooled from above. Neilson et al.[5] utilized the theoretical developments of Ref. 2 to investigate numerically the phenomenon of double diffusion in a horizontal annulus charged with an aqueous salt solution. A numerical model of convection heat and mass transport in solidification systems has been published by Voller et al.[6] Fang et al.[7,8] reported theoretical and experimental results on the selective freezing and melting of a dilute salt solution on a cold ice surface. Huppert and Worster[9] presented a series of solidification experiments with various aqueous solutions and at different values or initial cooling temperatures and concentrations.

Using different concentrations of an *n*-octadecane–*n*-hexadecane paraffin mixture, Webb and Viskanta[10] reported an experimental and analytical study of solidification of a binary mixture cooled from below. Observations revealed the existence of a mushy zone comprised of small dendrites protruding vertically into the surrounding liquid. The origin of freckles during unidirectional solidification was studied by Copley et al.[11] In a transparent system (30 wt % NH_4Cl–H_2O), the freckles were caused by upward flowing liquid jets in the mushy zone. Szekely and Jassal[12] obtained temperature and velocity measurements as well as photographic observations of a solidifying NH_4Cl–H_2O mixture. Their quantitative observations of the jetlike eruption of the interdendritic fluid was in good agreement with findings of previous investigators.[11]

An assessment of the effects of varying the initial concentration and the thermal boundary conditions on the solidification of a NH_4Cl–H_2O solution in a rectangular cavity was made by Christenson and Incropera[13] and Christenson et al.[14] Solidification of a NH_4Cl–H_2O solution in a square cavity with a free surface was studied experimentally and numerically by Engel and Incropera.[15] The surface tension forces caused a cellular flow at the top of the cavity and enhanced its development with increasing time.

Grange et al.[16] used interferometry to monitor the liquid phase concentration layer, which developed ahead of an advancing solid/liquid interface. Using an ethylene glycol solution as the phase change material, Fukusako and Yamada[17]

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examined photographically the effect of the initial concentration of the solution on the properties of the dendritic ice crystal. Incropera and Bennon[18] performed an experimental study on solidification of an aqueous $Na_2CO_3-H_2O$ solution in a horizontal cylindrical annulus. Cao and Poulikakos[19] investigated experimentally the phenomenon of solidification of a binary alloy (water and NH_4Cl) cooled through its top surface. Despite the fact that the mushy zone was permeable, the flow within it was weak. The heat transport in the mushy zone was conduction dominated.

The present study is novel because it investigates the effect of the *inclination angle* of the cold wall of a rectangular cavity with respect to the gravity vector on the solidification phenomenon. In addition, this study focuses on the *transient regime* and reports results on the evolution of the solid and the mushy regions. Note that the temperature of the cold wall is low enough to allow for the presence of a solid region in the system. Observations on the double diffusion flow structure in the liquid region are also reported.

II. Experimental Apparatus and Procedure

The experimental apparatus consisted of a test section and two supporting devices. The two supporting devices were a data acquisition system and a bath refrigerator circulator.

The test section was a rectangular cavity with two circular tracks attached to it. The tracks were used to rotate the cavity at any desirable angle (Fig. 1). To this end, all the three basic configurations (freezing along a "top" wall, a vertical "side" wall, and a "bottom" wall) are encountered. The internal dimensions of the water mixture space measured 5.1 cm long by 5.1 cm tall by 5.7 cm deep. The cold wall of the apparatus was constructed of stainless steel of thickness 0.685 cm. This wall was machined to allow for five thermocouples and a counterflow heat exchanger. The heat exchanger was constructed by milling four channels into the stainless steel surface. The direction of flow of the coolant in the heat exchanger was alternated between adjacent channels to establish isothermality of the wall. Indeed, the stainless steel wall of the apparatus was isothermal within 0.5°C for all the experiments.

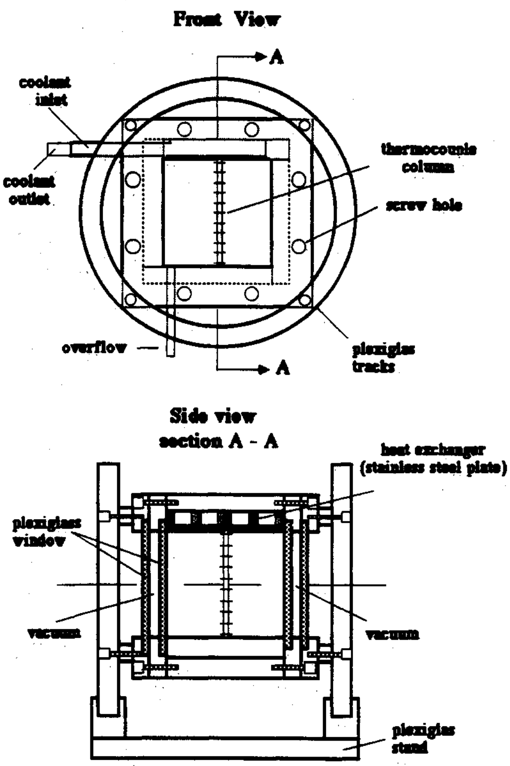

Fig. 1 Front view and side view of the test section.

The coolant was a 50% water/ethylene glycol solution precooled by a bath circulator refrigerator.

The remaining walls of the apparatus were constructed of Plexiglas® of thickness 0.685 cm. To allow for observation and photographing, the front and back walls were made of a "double" sheet of plexiglass with a vacuum gap between them (Fig. 1). This eliminated condensation on these walls and served as insulation between the water space and the environment. An additional 2-cm-thick styrofoam insulation surrounded the apparatus and helped to eliminate heat losses practically. A thermocouple column consisting of nine Chromel-Alumel thermocouples spaced every 6.2 mm was placed at the center of the cavity, perpendicular to the cold wall (Fig. 1). It was held in place by friction; it fit firmly between parallel walls of the cavity. This provided enough support for the column, so that it did not move even when the cavity was rotated. The thermocouple leads were taken out of the cavity through a hole near the bottom wall. The hole was connected to a flexible tube whose other end was elevated above the altitude of the test cell, thus avoiding leads and providing space for expansion upon freezing. The thermocouples yielded information on the evolution of the temperature distribution.

The procedure for the collection of the experimental data was as follows: The cavity was filled with a NH_4Cl-H_2O mixture of uniform concentration and positioned at a desired angle with respect to the gravity vector. Enough time was allowed for all fluid motion in the cavity to diminish. For all experiments in this study, the initial temperature of the fluid was room temperature. The coolant in the refrigerator circulator, which was precooled and kept at $-25°C$ throughout the experiments, was suddenly circulated through the heat exchanger machined into the stainless steel wall of the apparatus. As a result, natural convection and, eventually, solidification took place inside the cavity. The phase change interfaces and the double-diffusive interfaces in the liquid phase were observed and photographed at desired time intervals. Temperature measurements were recorded at two minute time intervals.

Initial solute concentrations of 5, 15, 20, and 25 wt % were used in the experiments. The solution filled the entire cavity (no free surface existed). The mixtures of 5 wt % and 15 wt % are located to the left of the eutectic point (Fig. 2a) and are termed "hypoeutectic." In this region, the dendrites in the mushy zone consist of ice. Water-lean fluid is rejected in the liquid phase. The 20 wt % concentration is very close to the eutectic composition of 19.7 wt %. Increasing the initial concentration to 25 wt % (inside the hypereutectic region located to the right of the eutectic point of Fig. 2a) yields

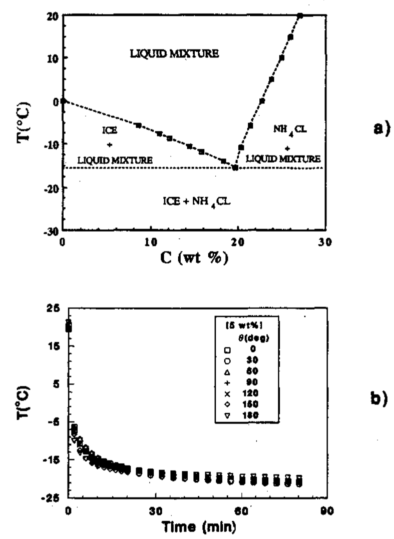

Fig. 2 a) The equilibrium phase diagram for a water–NH_4Cl mixture; b) the variation of the cold wall temperature with time for C_i = 0.05.

a water-rich interdendritic fluid rejected from the mushy region.

For each concentration, the experiments were performed by rotating the cavity from $\theta = 0$ deg to 180 deg at 30-deg intervals. The accuracy of the temperature measurements was estimated to be within three percent and was dictated by the accuracy of the software that converted the voltage measurements to temperature measurements. It is worth clarifying that the terminology "ice volume" defines the volume of the completely solid region. Whenever the volume of the mushy zone is measured it includes the volume of liquid saturating the solid "matrix" of the mushy zone. All volume measurements were performed from photographs by using a digital planimeter. Each measurement was performed five times and the average measurement was reported. The maximum deviation from the average in the course of the study was less than eight percent of the value of the measured quantity and defines the measurement error.[20]

The duration of each experiment was about 80 min. Note that steady state cannot be reached before complete solidification of the solution because all walls of the enclosure are adiabatic, except for the cold wall. However, after 60–70 min with freezing, the process slows down considerably.

III. Results and Discussion

As stated in the previous section, the coolant temperature as well as the initial temperature of the mixture were kept constant in all experiments in this study. Hence, the results illustrate the effects of the initial concentration and of the inclination angle on the freezing process. Representative measurements of the temperature of the cold wall are shown in Fig. 2b. These measurements correspond to the initial concentration of five percent. All other concentrations in the present study exhibited similar behavior. The wall temperature decreases rapidly and reaches a plateau at later times. The effect of the inclination angle on the cold wall temperature is minor.

Figure 3 illustrates the evolution of the temperature distribution at the centerline of the cavity at different inclination angles for $C_i = 0.05$. The symbols correspond to the experimental data and the lines correspond to the predictions of a simple theoretical model. This model will not be presented here because it follows the basic principles of one-dimensional conduction-dominated phase change existing in the literature.[20–23] In addition, the model resembles closely the one described in Cao and Poulikakos.[23] As shown in Fig. 3 for 0 deg $< \theta <$ 120 deg convection develops fast in the liquid region. As a result, away from the mush/liquid interface, the temperature distribution for the above mentioned inclination angles is practically uniform.

In most cases, the temperature increases monotonically as we move away from the cold wall. Exceptions to this are the early time temperature distributions for θ = 30, 60, 90 deg. In this case, the temperature increases as we move away from the mushy region, reaches a maximum, and then decreases to a plateau.

As θ increases to θ = 150 deg and eventually θ = 180 deg where the cold wall is located on the underside and the system is, for most part, stably stratified (thermally), the temperature distribution indicates that the convection effect weakens and conduction is the main heat transfer mode.

The theoretical model performs reasonably well in predicting the temperature distribution in the solid and mushy region. Convection apparently is not the dominant mechanism in the mushy zone in our study. On the other hand, the model does not perform well in predicting the temperature in the liquid region when convection is important (θ = 0–120 deg), especially at early times when the driving temperature differences are large and the convection phenomenon is strong. As the temperature field becomes conduction dominated (θ = 150 deg, 180 deg) the predictions of the theoretical model improve. The model performs the best when θ = 180 deg.

Figure 4 presents photographs of the solidifying system. All three regions (solid, mushy, and liquid) were clearly discernable in the photographs. Two typical inclinations are presented (θ = 30 deg, 90 deg) and two typical times (t = 30 min, t = 75 min). In all cases, both the solid and the mushy regions grow with time. The solid region occupies the near-wall vicinity. The shape of the mush/liquid interface is seriously affected by the presence of the double diffusive convection. The two-dimensional nature of the mush/liquid interface affects, in turn, the shape of the solid/mush interface. Upon solidification in the mushy zone, ice crystallizes "out" of the solution. As a result, the liquid concentration in the mushy zone increases. As this heavier, "high" concentration mixture enters the liquid region through the mush/liquid interface, it falls downward. Hence, a high-density region exists near the bottom of the cavity. Similar behavior has been observed by other investigators.[1] In Fig. 4b, for θ = 90 deg, a "recession" on the mush/liquid interface exists in the vicinity of the bottom wall. The reason for the presence of this recession is speculated as follows: The solute-rich liquid issued out of the mushy zone is heavy, it is swept downward by the flow and it is accumulated at the bottom of the test cell. The resulting increase in concentration lowers the liquidus temperature near the bottom wall. This decreases the solidification rate (locally) and causes the above mentioned recession. As time progresses, the amount of solute-rich liquid accumulating at the bottom wall vicinity increases and the recession moves upward along the mush/liquid interface. On the other hand, for θ = 30 deg, the recession of the mush/liquid interface is less visible. This is due to the fact that for this inclination angle, the high-density fluid accumulates by gravity to the vicinity of the lower corner opposite the mush/liquid interface and is hardly in contact with it. Additional discussion on the above phenomena is included later in this section when the observations of the density field in the liquid phase will be presented.

Figure 4c shows photographs of the solidification process for an initial concentration C_i = 0.25, higher than the eutectic concentration (Fig. 2). In this case, the solid matrix in the mushy zone consists of crystalized ammonium chloride. Crystallization of ammonium chloride yields a structurally "fragile" matrix. As shown in Fig. 4c, NH_4Cl crystals "break off" at the solid/mush interface and after they are carried by the fluid (Fig. 4c, t = 5 min), they settle at the vicinity of the bottom wall (Fig. 4c, t = 75 min). In addition, such crystals are created in the undercooled liquid phase, away from the mushy zone. The solid/mush interface is highly dendritic compared to the rather smooth interface of Figs. 4a and 4b.

The photographs of Fig. 5 present a visualization of the sharp density interfaces in the liquid phase. These interfaces separate well-mixed regions of uniform temperature and concentration. Note that because we do not perform detailed temperature and concentration measurements in the liquid, the term "well-mixed regions" is used in a speculative sense. However, previous studies is double diffusion[24,25] have shown definitively that regions of the type observed in this study separated by sharp density interfaces are indeed well mixed. In Fig. 5a starting from a single interface, a multiple interface structure develops at late times. Figure 5b, on the other hand, corresponds to θ = 120 deg. In this case, the high concentration fluid is in contact with the solid/mush interface and causes a recession of this interface as discussed earlier. Interestingly, the double diffusive interfaces in Fig. 5c appear near the top of the cavity and advance downward with time. The reason for this phenomenon stems from the fact that Fig. 5c corresponds to an initial concentration C_i = 0.25, higher than that of the eutectic point of Fig. 2a. Here, the solid matrix of the mushy zone consists of crystallized ammonium chloride. The density of the liquid in the mush decreases as the ammonium chloride crystallizes "out." When this "low density" fluid enters the liquid region through the mush/liquid interface it moves upward and occupies the upper part of the

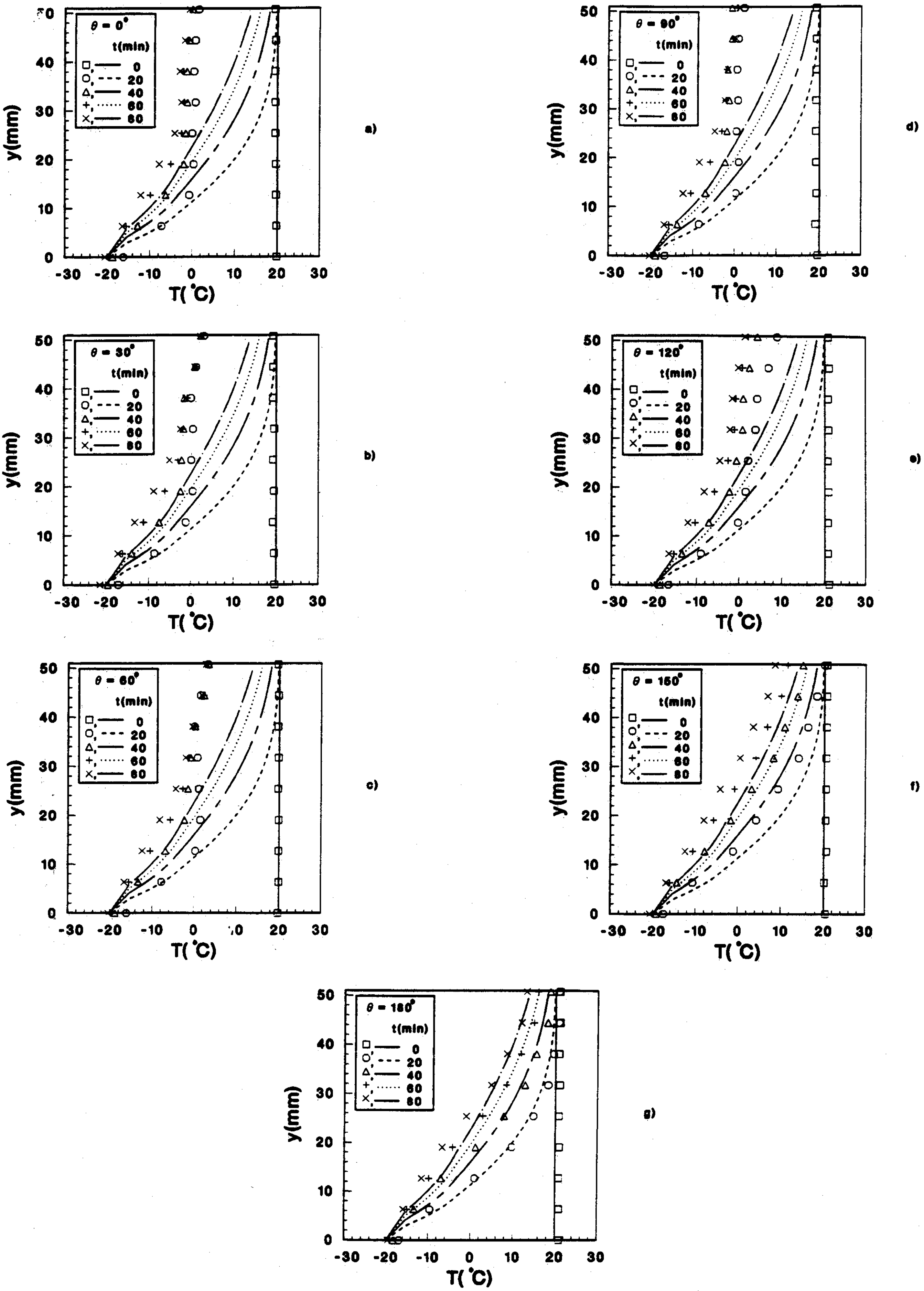

Fig. 3 Temperature distribution at the centerline of the cavity for $C_i = 0.05$.

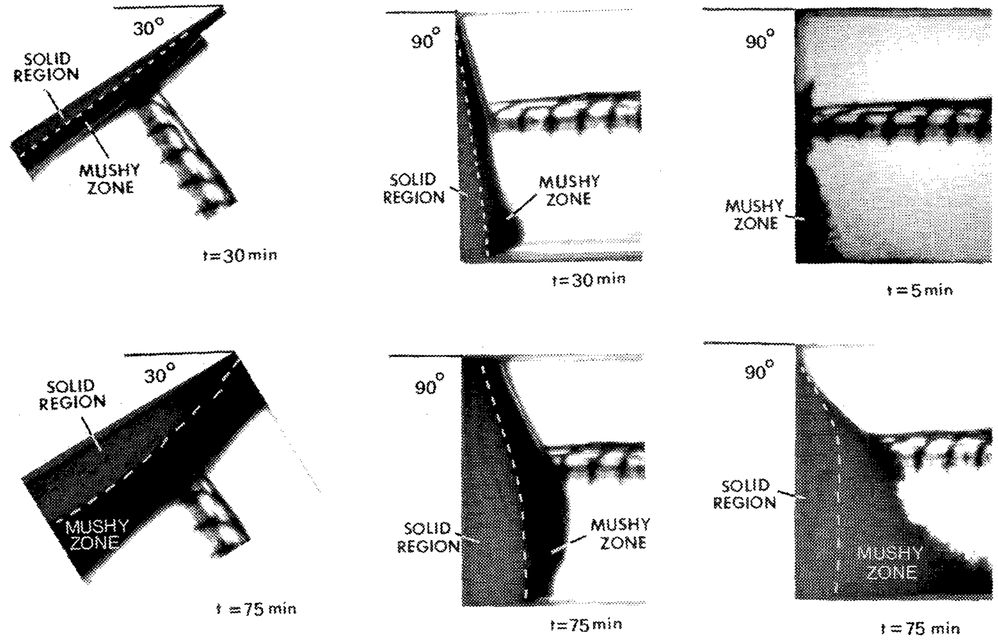

Fig. 4 Photographic documentation of the solidification process for characteristic and angles and initial concentrations. a) $C_i = 0.15$, $\theta = 30$ deg; b) $C_i = 0.15$, $\theta = 90$ deg; c) $C_i = 0.25$, $\theta = 90$ deg.

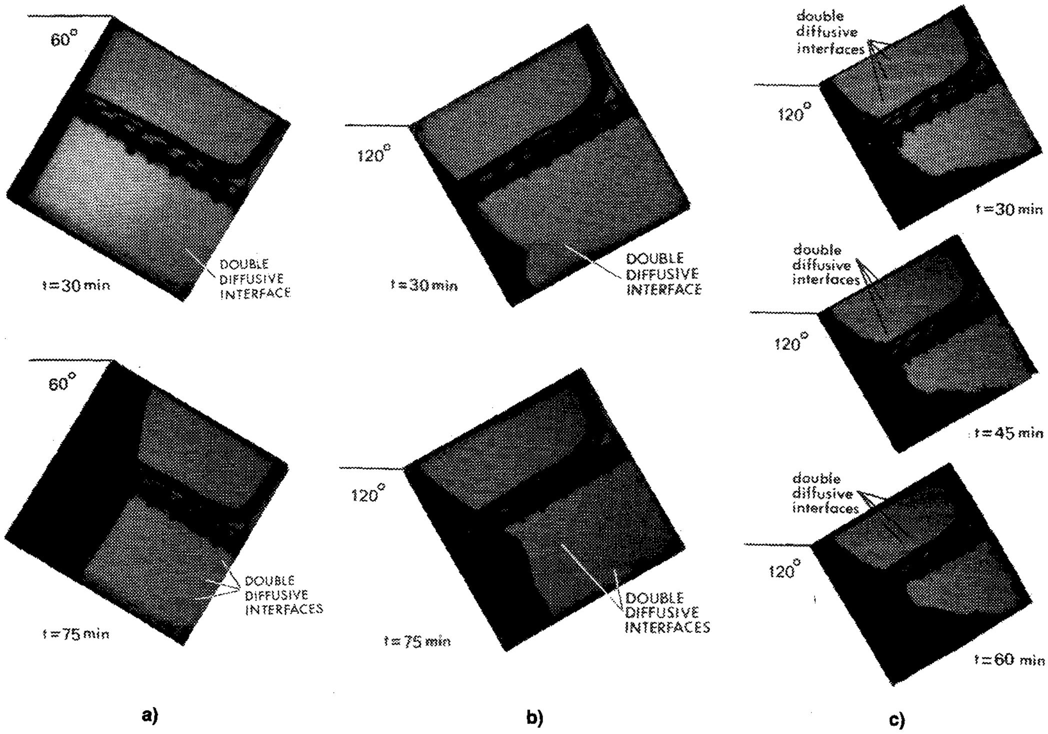

Fig. 5 Examples of the double diffusive interfaces in the liquid region. a) $C_i = 0.15$; b) $C_i = 0.2$; c) $C_i = 0.25$.

cavity, eventually creating a multiplicity of well-mixed regions of uniform temperature and concentration separated by sharp interfaces as shown in Fig. 5c.

Photographs of the type shown in Fig. 4 were used to obtain tracings of the solid/mush and mush/liquid interfaces at characteristic times, angles, and concentrations. These tracings are shown in Figs. 6 and 7. Of interest, again, is the fact that in several cases (Figs. 6b, 6c, 7b, 7c) the mush/liquid interface distance from the cold wall increases as we move away from the top wall, reaches a maximum and decreases thereafter. The reason for the decrease is the presence of flow near the bottom wall as indicated by the presence of the diffusive interfaces discussed earlier. For most part, the solid/mush interface is less affected by the flow and the solid region thickness as we move away from the top wall. As shown in Figs. 6 and 7, there are instances in which the cold wall of the cavity is covered partly by the solid region and partly by the mushy region. In other instances, the cold wall is covered by all three regions, solid, mushy and liquid. Cases where the cold wall is lined by mush and liquid only, also exist. This behavior is a direct result of the effect of the double diffusion flow on the solidification process. In cases where this flow was weak[26]

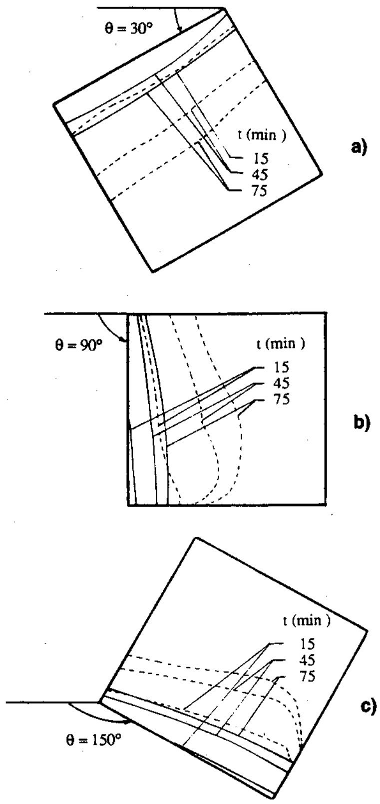

Fig. 6 The evolution of the solid/mush and the mush/liquid interfaces in time for C_i = 0.05. a) θ = 30 deg; b) θ = 90 deg; c) θ = 150 deg.

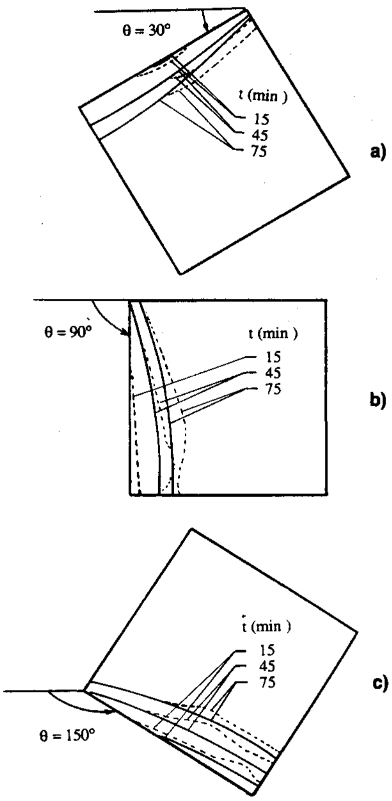

Fig. 7 The evolution of the solid/mush and the mush/liquid interfaces in time for C_i = 0.2. a) θ = 30 deg; b) θ = 90 deg; c) θ = 150 deg.

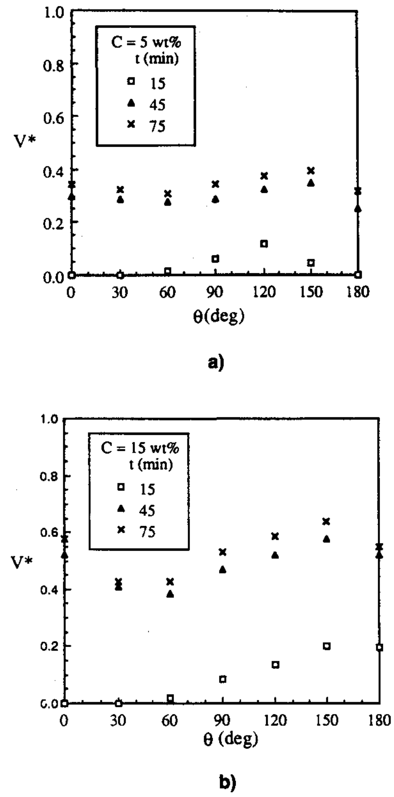

Fig. 8 The effect of the inclination angle on the ratio of the volume of the solid region to the volume of the entire region in which the solid phase exists (solid plus mush). a) C_i = 0.05; b) C_i = 0.15.

(θ = 180 deg, for example), such behavior was not observed and the cold wall was covered by a single region (mush or solid). It is important to note that for C = 20 wt % (Fig. 7) in several cases (Fig. 7a, t = 45 min, t = 75 min; Fig. 7b, t = 45 min; Fig. 7c, t = 45 min, t = 75 min), the mushy region covers only part of the solid region. As a result, both a solid/liquid and a mush/liquid interface coexist.

Figures 8a and 8b illustrate the effect of the inclination angle on the growth of the ratio of the volume of the solid region to the volume of the entire region in which the solid phase exists (solid plus mushy regions). Clearly, the effect of θ on V^* is rather weak. In just about all cases, however, V^* increases with time, indicating that the growth rate of the solid region is faster than that of the mushy region. Another indication that the data offer is that at greater times the dependence of V^* on θ features a maximum and a minimum. In Figs. 8a and 8b for example, for t = 75 min, this maximum occurs at approximately θ = 150 deg. The minimum occurs at approximately θ = 60 deg in Fig. 8a, and θ = 45 deg in Fig. 8b. The value of the initial concentration does not seem to have a serious qualitative impact on the dependence of V^* on time and on the inclination angle.

A result of engineering interest is the dependence of the total amount of solidified volume (in completely solid or mushy form) on time, inclination angle, and concentration.

Using a regression analysis[27] for the data collected in the study, the following correlation was obtained:

$$V^{**} = A\tau^B e^{D \cos(\pi/2 - \psi)} \tag{1}$$

where

$C_i = 0.05$; $\quad A = 4.711$, $B = 0.797$,

$D = 0.044$, $\psi = 0.941\,\pi$

$C_i = 0.15$; $\quad A = 7.221$, $B = 1.068$,

$D = 0.106$, $\psi = 0.93\,\pi$

The above correlation was plotted together with all the data points for comparison,[20] which is not shown here for brevity. However, it is felt that Eq. (1) performs rather well within the range $0 < \theta < \pi$, $0 < \tau < 0.05$ investigated in the present study.

Conclusions

In this paper, an experimental study on the phenomenon of solidification of a binary alloy in an inclined rectangular cavity was reported. It was found that the cavity was occupied by three distinct regions: a solid region, a mixed-phase region, and a liquid region. The presence of the double diffusive convection in the liquid region severely affected the shape of the mush/liquid interface and, indirectly, the shape of the solid/mush interface. The inclination angle had a paramount effect on the double diffusion flow and, therefore, on the solidification process.

The flow in the liquid region was characterized by a sequence of well-mixed regions of uniform temperature and concentrations situated one on top of the other and separated by sharp interfaces. This finding agrees with observations in classical double diffusion studies. The well-mixed regions and the double diffusive interfaces were initiated near the lower region of the cavity for initial concentrations smaller than the eutectic, and near the upper region of the cavity for initial concentrations higher than the eutectic.

A simple theoretical model based on conduction predicted the temperature distribution in the solid region acceptably. However, this model did not perform well in the liquid region. There, the agreement between the model and the experiments was good only for 150 deg $\leq \theta \leq$ 180 deg where the natural convection flow was weak and the heat transfer in the liquid, especially at late times, was conduction dominated.

Detailed results on the dependence of the total solid volume produced in the system on time, inclination angle, and initial concentration were obtained. These results were summarized with the help of an engineering correlation [Eq. (1)].

Acknowledgment

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