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ONSET OF CONVECTION DURING VERTICAL
DIRECTIONAL DENDRITIC SOLIDIFICATION AND THE
STABILIZING EFFECT OF THE TWO-PHASE REGION

A Dissertation

Presented for the

Doctor of Philosophy Degree

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JOHN A. HOPKINS

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Dedicated to my loving wife, Terri.

Live joyfully with the wife whom thou lovest all the days of the life of thy vanity, which he hath given thee under the sun, all the days of thy vanity: for that is thy portion in this life, and in thy labor under the sun. Whatsoever thy hand findeth to do, do it with thy might; for there is no work, nor device, nor knowledge, nor wisdom, in the grave, whither thou goest.

Ecclesiastes 9:9-10

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ABSTRACT

The onset of convection during vertical directional dendritic solidification has been examined experimentally and numerically. The effects of the two-phase region are significant in determining the breakdown of the buoyantly unstable solutal diffusion layer produced during solidification of an $\text{NH}_4\text{Cl-H}_2\text{O}$ solution.

The transition to convection was experimentally characterized by a combined solutal and thermal Rayleigh number using interferometry. The critical combined Rayleigh number is dependent on the two-phase effects of the dendritic mushy zone. A linear stability model used measured values for effective primary dendrite arm spacings to model the experiments and yielded the same exponential dependence for critical combined Rayleigh number versus two-phase region height. The model includes an allowance for shrinkage flow and two-phase inertial effects. These terms have a significant influence on breakdown at high growth rates.

The two-phase effects were characterized by the height of the mushy zone. The critical combined Rayleigh number is an exponential function of this distance. Application of the model to the eutectic growth of $\text{NH}_4\text{Cl-H}_2\text{O}$ show that growth is stable for reduced levels of gravity and also for high growth rates.

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NOMENCLATURE

A	Discretization LHS, Constant
A_o	Constant
a	Wavenumber (1/cm)
a_c	Wavenumber at onset of convection (1/cm)
α	Thermal Diffusivity (cm ² /sec)
B	Discretization RHS, Stretching Function, Constant
B_T	Coefficient of thermal expansion (1/C)
B_C	Coefficient of solutal expansion (1/wt%)
b_1	Curve Fit Coefficient
C	Solutal concentration (wt%)
C_o	Initial solutal concentration (wt%)
C_l	Liquid solutal concentration (wt%)
C_{l2}	Liquid solutal concentration at T_{s1} (wt%)
C_s	Base state solutal concentration (wt%)
C_{s1}	Solid solutal concentration at T_{s1} (wt%)
C_{s2}	Solid solutal concentration at T_{s2} (wt%)
C_∞	Solutal concentration in far field (wt%)
C_{Tip}	Dendrite tip solutal concentration (wt%)
C'	Perturbed solute concentration (wt%)
ΔC	Concentration difference (wt%)

ΔC_{zt}	Concentration difference in two-phase region (wt%)
ΔC_{dl}	Concentration difference in fluid diffusion layer (wt%)
C_p	Specific Heat (J/g C)
CR	Cooling rate (C/min)
D	Solutal diffusivity (cm ² /sec)
d	Characteristic length for Forchheimer inertia term (cm)
d1	Primary dendrite arm spacing (cm)
dl	Extent of solutal diffusion layer in all liquid region (cm)
F	Forchheimer inertia term (1/cm)
F_s	Base state Forchheimer inertia term (1/cm)
f'	General function expanded in terms of Fourier components
$F(z)$	General perturbation solution function
G1	Temperature gradient at solid-liquid interface (C/cm)
$G1_c$	Temperature gradient at convective breakdown (C/cm)
G	Temperature gradient input to experiments (C/cm)
g	Gravitational acceleration (cm/sec ²)
g_e	Earth's gravitational acceleration (980 cm/sec ²)
γ	Solidification shrinkage parameter
H _{zbd}	Height of two-phase region at convective breakdown (cm)
I	Intensity of Light (W/cm ²)
κ	Permeability (cm ²)
κ_s	Base state permeability (cm ²)

k	Partition Coefficient
L	Characteristic length (cm)
L_f	Latent heat (J/g)
λ^*	Convective cell size (cm)
λ	Wavelength of Laser Light (cm)
m	Liquidus slope (C/wt%)
n	Index of refraction
n_o	Index of refraction of air
Δn_{dl}	Index of refraction difference in fluid diffusion layer
η	Computational domain
ν	Kinematic viscosity (cm ² /sec)
Ω	Stretching Function
π	Pi (≈ 3.14159)
Φ	Fraction of liquid or porosity
ϕ	Phase angle
P	Pressure (dynes/cm ²)
P_s	Base state pressure (dynes/cm ²)
P'	Perturbation pressure (dynes/cm ²)
Ra_T	Thermal Rayleigh number
Ra_S	Solutal Rayleigh number
Ra_C	Combined thermal and solutal Rayleigh number
Ra_o	Curve Fit Coefficient

R	Thermal Rayleigh number for case 5.5.2
R^*	Combined thermal and solutal Rayleigh number for case 5.5.2
ρ	Density (g/cm^3)
ρ_l	Liquid density (g/cm^3)
ρ_s	Solid density (g/cm^3)
ρ_o	Reference density (g/cm^3)
ρ'	Perturbation density (g/cm^3)
S	Solutal Rayleigh number for case 5.5.2
S'	Solute Gradient ($\text{wt\%/cm}$)
S	Stretching Parameter
σ	Temporal Growth Parameter ($1/\text{sec}$)
T, T_1, T_2	Temperature (C)
T_l	Liquid temperature (C)
T_s	Base state temperature (C)
T'	Perturbation temperature (C)
T_∞	Far Field Temperature (C)
ΔT	Temperature difference (C)
ΔT_{zt}	Temperature difference in two-phase region (C)
ΔT_{dl}	Temperature difference in fluid diffusion layer (C)
t	Time (sec)
τ	Distance Laser Travels Through Cuvette Test Section (cm)
U	Electric field (volts/cm)

U	Fluid velocity in x direction (cm/sec)
u	Fluid velocity in x direction (cm/sec)
u'	perturbation fluid velocity in x direction (cm/sec)
V	Solidification growth rate (cm/sec)
$\bar{V}$	Fluid velocity vector (cm/sec)
W	Fluid velocity in z direction (cm/sec)
w	Fluid velocity in z direction (cm/sec)
w'	Perturbation fluid velocity in z direction (cm/sec)
w_s	Base state fluid velocity in z direction (cm/sec)
x	Horizontal coordinate normal to beam propagation (cm)
$\bar{x}$	Solution vector W,T,C
y	Horizontal coordinate parallel to beam propagation (cm)
z	Vertical coordinate normal to beam propagation (cm)
z_t	Height of two-phase region (cm)
z_l	Far-field distance (cm)

1. INTRODUCTION

This study examines the effects of the porous two-phase region on the onset of convection, i.e convective breakdown, occurring during vertical directional dendritic solidification of a binary eutectic alloy. This problem involves several areas of heat and mass transfer, including convective stability, two-phase flow, and directional solidification. A numerical linear perturbation analysis was used to predict convective breakdown, while interferometry was used to experimentally characterize the breakdown.

Directional solidification is an important research topic for several reasons, the most notable being its relevance to materials processing. Accurate modelling of processing physics is needed to obtain acceptable finished components, but the problem is difficult since the conservation equations for mass, momentum, energy, and solute have to be satisfied, and the relations expressing phase fractions as functions of local thermosolutal conditions and material properties have to be included in the solution. Disparities in local length scales for these relations tend to make the problem numerically intractable.

The first analytical models for solidification were derived for planar front solidification¹, and early stability analyses were concerned with determining the solid growth front transition from a planar morphology to either a cellular or a dendritic (two-phase) morphology.² This research deals only with the dendritic morphology,

hence a two-phase region and the associated porous media effects exist for the solidification problems examined.

The occurrence of flows during solidification has a strong influence on the compositional structure of finished parts.³⁻⁷ The distribution of alloying constituents is important since it determines the solid material properties, and the capability to predict the final distributions is desirable.

For multicomponent systems, the temperature dependence of solid solubility for each constituent results in solute rejection during solidification. The solute transport that occurs creates a solutal diffusion layer ahead of the solidifying interface. If the solute is less dense than the solvent, this layer is subject to buoyancy driven convection. Therefore, the prediction of buoyant (natural) convection is of primary interest to the materials scientist.

Previous studies of horizontal and vertical directional solidification (VDS) have shown the gravity dependence of the flows that occur under these conditions.⁷⁻¹⁰ For the case of VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$, the convective flows follow a statically stable period of dendritic growth for which there are no perceptible flows.^{11,12} The onset of flow is dependent on the sum of the thermal and solutal Rayleigh numbers defined over the solutal diffusion layer.^{11,12} The convective flows appear as a transition from a diffusion-dominated solidification problem with a low velocity, uniform flow base state, to one that is dominated by convection. This research will attempt to define the transition in terms of the local conditions at breakdown, motivated by the fact that the flows occurring after transition contribute significantly to the mass transfer detrimental

to solid material properties. Hence, if the flows were suppressed, solidification could proceed without the associated convection generated macro-scale constituent segregation, giving a more uniform solid structure and a higher quality finished part.

Fluid flows are fundamental to many transport processes in nature, and heat and mass transfer are generally enhanced by the convective transport due to these flows. Convection is often marked by a transition from either a non-convective or nominal flow case to a fully developed flow situation. There are, in general, two cases in which transition occurs, the transition from a quiescent state to a convecting state and the transition from laminar or uniform convection to a turbulent or non-uniform convection. These transitions are significant due to the differences in transport properties for the two states. The significance is clearly evident in the case of flow over a sphere where the coefficient of drag undergoes a dramatic change when the flow transitions from laminar to turbulent.¹³ The designers of aircraft, automobiles, and other aerodynamically motivated objects are very interested in knowing under what conditions these transitions occur and the flow characteristics for each of the regions.¹⁴ Flow transitions are also significant in global weather patterns and the formation of clouds, and better predictions of weather trends are possible with knowledge of these transitions.¹⁵

When the problem in question is that of a fluid in a saturated porous material, additional factors need to be considered. The additional resistance to flow due to a porous media is typically included in the governing conservation equations as a Darcy force term in the momentum equation. This term originated from a force balance for

the pressure drop of a nominally laminar flow through a saturated porous medium.¹⁶ The pressure drop for low velocities was found to be directly proportional to the flow velocity, with the constant of proportionality being the permeability, κ . The relationship between permeability and the porosity, i.e. fraction of fluid, has been characterized for a variety of flow and material conditions,¹⁷⁻¹⁹ but the relation given by Blake, Kozeny, Carmen, and Hatch, often referred to as the Kozeny-Carmen or Blake-Kozeny relation is the most widely used for solidification studies.²⁰⁻²⁴

For certain flow conditions, the Darcy form of the momentum equation is found lacking, and the influence of the convective terms (the Forchheimer extension) and the influence of viscous terms (the Brinkman extension) are included in the momentum equation formulation.²⁵⁻²⁷ These extensions were used in this work and are discussed in Section 5.

The analysis of stability is most often accomplished through the use of perturbation methods.²⁸ The purpose of these methods is to determine, in a problem dependent parameter space, whether perturbations to a defined base state will grow with time. In a first order analysis such as in this study, the perturbations are assumed to be infinitesimal. The perturbations will either grow (transition has occurred), decay (transition has not occurred), or remain constant (the system is neutrally stable).

This study analyzes the transition to convective flow for VDS of the transparent binary eutectic system $\text{NH}_4\text{Cl-H}_2\text{O}$. This system is a transparent metal model that has been used extensively in previous solidification studies.^{4,6,7,11,29-32}

Because it is transparent, optical methods can be used to experimentally determine not only the occurrence of the transition to convection, but the thermal and solutal fields characterizing its occurrence. The thermal and solutal fields in the liquid create density differences which drive the buoyancy driven flows. Measurements of the thermal and solutal profiles yield parameters of which breakdown is a function. These parameters can be compared with the results of a permeability-dependent linear stability analysis to characterize the effects that the porous two-phase region has on the occurrence of breakdown.

The goal of this study was to characterize convective breakdown in terms of known parameters for the $\text{NH}_4\text{Cl-H}_2\text{O}$ system undergoing VDS in a one gravity environment. The dependence of the breakdown on solidification parameters such as solidification velocity and temperature gradient was examined, as was the dependence on gravity level. It is desirable to process materials under conditions in which there are no convective flows, and this analysis defined solidification and/or environment characteristics for which this is possible. Extrapolation to other systems can be accomplished, provided an accurate relation for permeability as a function of fraction of liquid is available.

In the following pages, Section 2 introduces background material including solidification and convective stability, and Section 3 defines the VDS convective stability problem analyzed in this study. Section 4 examines the experimental approach and results, and Section 5 discusses the numerical approach and results.

Section 6 gives a comparison of experimental and numerical results, and Section 7 states conclusions.

2. BACKGROUND

2.1 SOLIDIFICATION PROCESSING

Solidification is an important part of virtually all materials processing activities and is, therefore, an integral step in the manufacture of almost every man-made item. There are several sources in the literature that discuss the process and its relation to part formation.^{33,34} The compositional make-up of finished parts is very important, especially for critical applications such as turbine blades. Compositional segregation occurs in the part when constituents in a material are distributed in a non-uniform manner. The homogeneity is thus dependent on the solidification process used in its manufacture, and knowledge of solidification fundamentals and the accompanying mass transfer phenomena are crucial for the processing of acceptable components.

Solidification is characterized by a change of phase due to the removal of energy from a substance in a liquid state. Since the process is thermodynamic, it is determined by the minimization of Gibb's free energy. The relation between state values of temperature, concentration, and pressure and the local value for phase fraction is given by the equilibrium condition for free energy between phases. Most studies involving a solid-liquid phase change use experimental data for identifying the relationships between temperature, concentration, and phase fraction. This information is usually in the form of an equilibrium phase diagram, an example of which is shown in Figure 2.1. This particular diagram shows the characteristics of the solid to liquid (S-L) and liquid

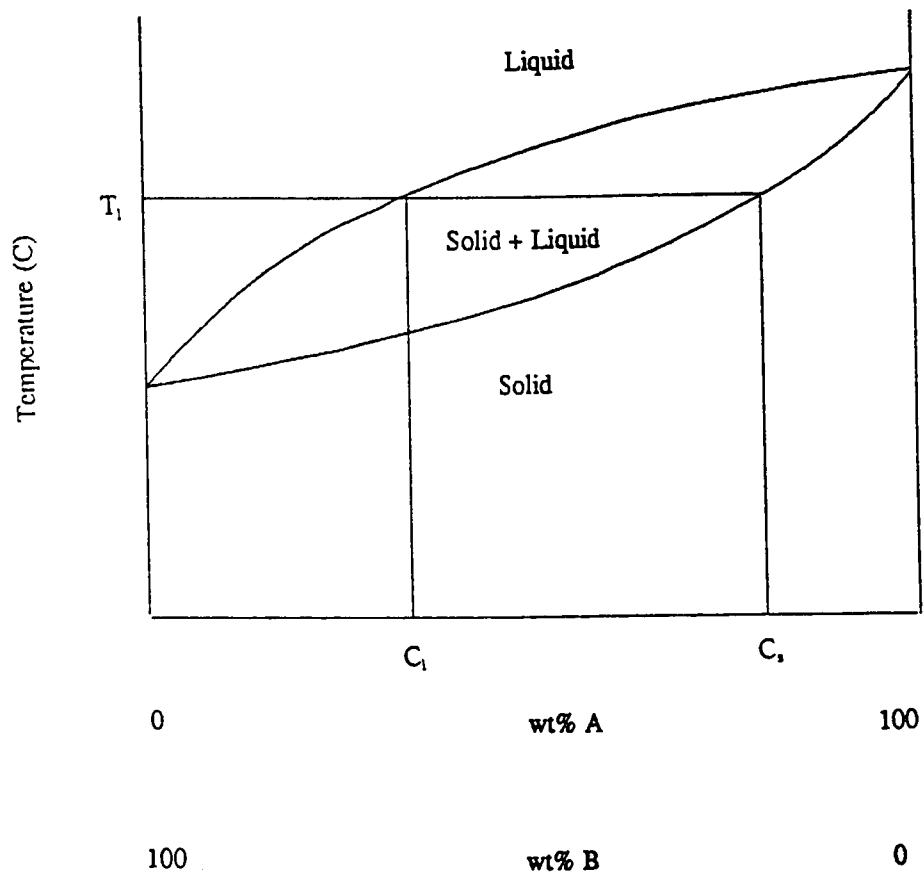


Figure 2.1 Binary Phase Diagram.

to solid (L-S) phase change processes that occur for this two component material over various temperature ranges.

The solidification process for a binary eutectic system is illustrated in Figure 2.2. Constituent A is the solute and B is the solvent. If a liquid of initial concentration C_0 has its temperature lowered to temperature T_1 , solidification will begin. At this temperature, solid forms with a concentration of C_{s1} . As the temperature is lowered further to T_2 , the solid formed has a concentration of C_{s2} , and the liquid has a concentration of C_{l2} . The equilibrium liquid concentration increases as the temperature decreases, necessitating a solute redistribution in the solidifying region. This also yields microsegregation in the solid. In the case of directional solidification, a solutal diffusion layer is produced ahead of the solidifying interface. Figure 2.3 shows a typical solutal diffusion field in front of a directionally solidifying dendritic growth front. The presence and evolution of this diffusion field has a marked effect on the solidification process. The local solidification conditions are dependent on the rate of diffusive solute transport occurring in this layer.

A solidifying material can take on different forms or morphologies. The morphology is either planar or cellular and is dictated by local thermophysical properties and solidification parameters.³⁴ Figure 2.4 shows examples of dendritic and equiaxed growth, which are special cases of the cellular growth morphology and important in the casting of multi-component alloys. Figure 2.4 also shows the temperature distributions for typical solidification.

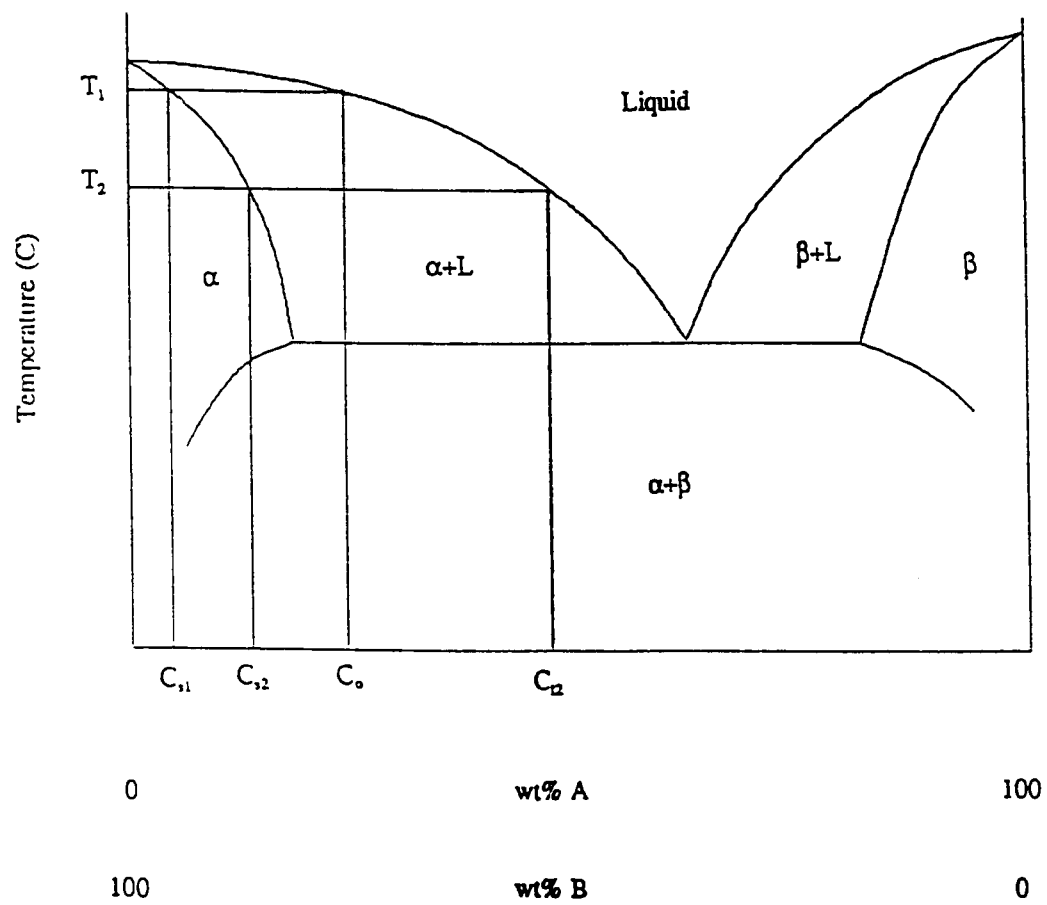


Figure 2.2 Binary Eutectic Phase Diagram.

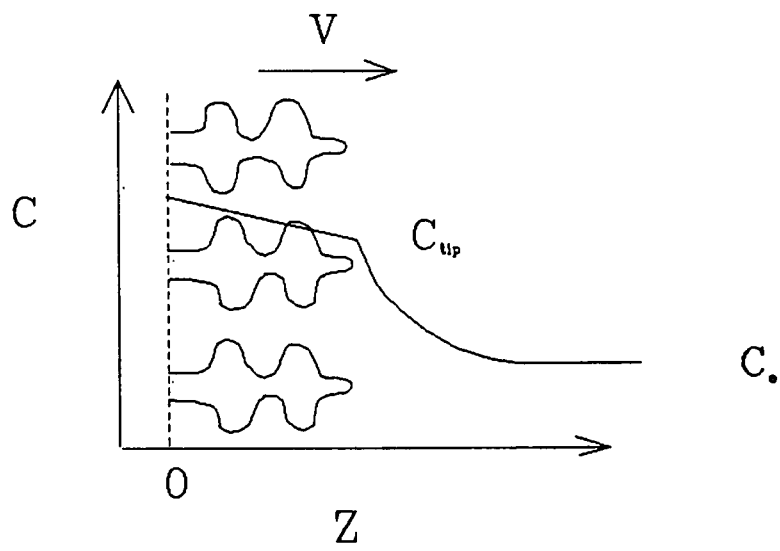
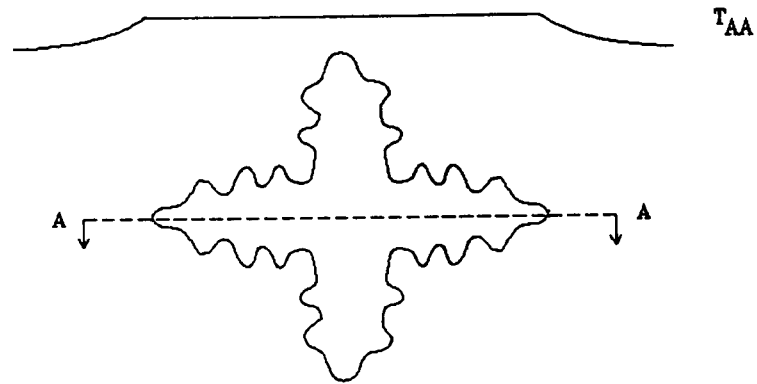


Figure 2.3 Solute Profile for Directional Solidification.

Equiaxed



Dendritic

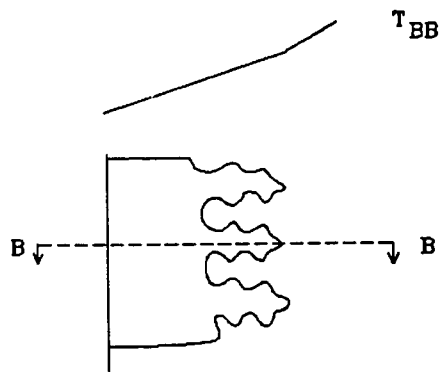


Figure 2.4 Comparison of Equiaxed and Dendritic Growth.

The local thermal and solutal fields are important in determining the solidification process. Directional solidification is one technique that attempts to control the thermal field so that the solidification process is as uniform as possible. By limiting growth of the solid-liquid interface to one direction, the material is made uniform in the plane normal to the solidification direction. This process aligns the grains or interfaces between solid zones and is used in the manufacture of turbine blades and, in variant forms, for crystal growth.^{34,35} It is, however, subject to non-uniform mass transfer due to convective fluid flows occurring during solidification.³⁵ These flows can create large scale constituent redistribution known as macrosegregation. Consider Figure 2.2. If constituent A is of a different density than constituent B, the potential for density gradients and buoyancy driven flows exist. The fluid in the diffusion layer will move in the direction of gravity if it is more dense and will move in the opposite direction if it is less dense. To help minimize the macrosegregation caused by these flows, the solidification process is made to occur in the vertical plane in a direction opposite the gravity vector. This is particularly effective for the rejection of a solute that is more dense than the original liquid material. In the case of the rejection of a light solute, the diffusion layer can become unstable and result in convection.^{11,12}

An important factor in determining the character of these convective flows is the influence of the two-phase region, i.e. mushy zone. This region, denoted as S+L in Figure 2.2, is comprised of solutal diffusion layer liquid and dendritic solid. It is a saturated porous media and, via momentum exchange, can have profound effects on the convective flows occurring in the system.

Most fluid dynamic models for this region use a Darcy term in the conservation of momentum equations to account for the additional momentum dissipation due to the interspersed solid material.^{20-23,36-38} The Darcy term requires knowledge of the porous media in terms of a permeability function. Several models have been developed which define the two-phase region permeability in terms of the phase fraction and dendritic structure.^{17,39,40} However, the models are limited in range of applicability. More recently, the importance of additional terms in properly modelling the effects of porous media had also been shown.^{25,41-43} Viscous diffusion and inertial effects due to interacting boundary layers have been shown to be significant contributors to momentum exchange in porous layers for some types of flows.^{23,25,41-43} These terms are referred to as the Brinkman and Forchheimer extensions, respectively.

These topics are important in the production of superalloy parts such as turbine blades. Superalloys are material with high strength-to-weight ratios and good temperature resistant properties. In general, they have a strong and relatively heavy base metal alloyed with metals that are either lighter, harder, or more resistant to temperature. The number of alloying metals in any particular superalloy is large. Several of the superalloys are processed using vertical directional solidification (VDS) and have two-phase regions and a diffusion layer that are subject to buoyancy driven flows. Because these materials are used in safety-critical applications, the structure of the part is very important, and components that are free of flaws due to segregation are very desirable. The expense of creating these components has given rise to speculation of possible

space processing in which the gravity levels would be low enough such that the buoyancy driven flows and resulting segregation would not exist.

2.1.1 *SOLIDIFICATION EXPERIMENTS*

Experiments using actual metals, e.g. Pb-Sn and Al-Cu, have been successful at identifying the importance of convective flows in creating macrosegregation.^{3,5,7,44,45} These studies have shown that buoyancy is primarily responsible for these flows.

Flemings et al examined the solidification of Al-4.5 wt% Cu and showed that macrosegregation is influenced by convection.⁴⁵ Mehrabian et al also studied convective effects during solidification of Al-Cu and using x-ray techniques.⁴⁴ Giamei and Kear also noted segregation effects in several Nickel-based superalloys. They examined the directional solidification of Ni-Al to identify the levels and types of segregation.³ These experiments attempted to correlate compositional segregation with the presence of defects in the solid. These defects were called freckles due to their physical appearance. Streat and Weinberg used radioactive tracers to identify the effects of buoyancy driven flows on segregation during VDS of Pb-Sn.⁵ Macrosegregation was shown to be due to the buoyancy driven flows occurring in that system.

The inherent difficulties associated with using post-processing and radioactive tracers for metal alloy studies have resulted in the extensive use of metal models in solidification experiments.^{4,6,7,11,12,29-32,46} $\text{NH}_4\text{Cl-H}_2\text{O}$ is a common metal model and has the advantage that it is transparent. Copley et al used VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ to visualize buoyancy driven liquid jets originating from the mushy zone.⁴ Asai and Muchi used a dye tracer during VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ to examine the formation of channel type

segregation.⁶ A qualitative pattern for the fluid motion was obtained in this manner, and a mathematical model was derived to predict segregation. In an attempt to prevent channel segregation, Sample and Hellawell examined the effects of centripetal accelerations on the solidification of Pb-Sn and $\text{NH}_4\text{Cl-H}_2\text{O}$.⁷ They concluded that the formation of the channels is preceded by a period during which the growth of the mushy zone increases to a stable value. They concluded that the origin of fluid creating the channels was ahead of the mushy zone rather than within it. Szekely and Jassal used a variety of techniques to study the horizontal (normal to the direction of gravity) directional solidification of $\text{NH}_4\text{Cl-H}_2\text{O}$.⁴⁶ The flows in these experiments extended into the mushy-zone, and were strong enough to break dendrite arms. Based on these studies, the effects of fluid motion on the solidification process have been well-documented in both metallic and non-metallic systems.

The influence of the two-phase region on convection has resulted in the characterization of the permeability of this region as a function of the fraction of liquid and geometry of the dendrites.^{17-19,39} Recent studies in dendritic systems measured permeability for Pb-Sn and Al-Cu metallic systems, and recommended using a permeability model based on either an empirical form of the Blake-Kozeny relation or the model of Drummond and Tahir for cylinders.^{17,39} The use of these models will be discussed in section 5.

The convective flows occurring during solidification have a significant effect on the resulting thermal and solutal fields. Several experiments have utilized transparent metal models to measure the solutal fields present during solidification.^{21,47} Intrusive

refractometry⁴⁷ and the withdrawal of sample fluid⁹ have both been used in lateral solidification studies to make spatial solute measurements. These studies were designed to supplement modelling efforts, and good qualitative agreement between experimental and numerical results was obtained.

2.1.2 THE CAST SOLIDIFICATION EXPERIMENTS

The Casting and Solidification Technology (CAST) experiments performed in support of the January 1992 International Microgravity Laboratory spacelab mission have closely examined the VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ in a one-gravity environment. Through the implementation of a system which carefully controls solidification and the use of several optical techniques, studies of the events preceding convective flow have been possible.^{22,11,12,30,32,48,49} It has been shown that for VDS under certain conditions, the flows do not immediately occur, but develop from a stable growing diffusion layer to, first stationary convection and then to fully developed convective flows.^{11,12} The transition to these stable stationary convective cells, in the form of Rayleigh-Benard cells, is influenced by the character of the two-phase region.⁴⁸

During the growth stage preceding the cellular convection, quantitative measurements of temperature and concentration can be made through the use of the index of refraction variations of $\text{NH}_4\text{Cl-H}_2\text{O}$. While other studies have typically used either probes or removal of material, the CAST experiments use layer averaging to non-intrusively measure the solutal field ahead of the mushy zone.^{31,32}

2.1.3 *SOLIDIFICATION MODELLING*

Analytical and mathematical modelling of solidification has progressed dramatically during the last 20 years. The first models were based on planar solidification with no convection,¹ while current models take into account dendritic growth and convective flows.^{21-23,37,50} In the immediate future, microscopic modelling will be added to capture the physics of the microstructure, with the realization that the microscopic and macroscopic models must be linked appropriately.^{22,51-53}

The first solidification models were relatively simple. Convective flows were usually ignored, and uncoupled equation sets were used to describe a one-dimensional plane front solidification process.¹ The first application of uncoupled momentum and energy equations was performed by Szekely,⁴⁶ who solved the solute equation using assumptions concerning the solidification front. Coupled, multi-equation models followed, usually treating the solidification velocity as a known quantity.⁵⁴⁻⁵⁸ The latest models use coupled energy, momentum, and solute equations as well as relations for phase fraction and density to characterize the process.^{20-22,37,50}

Current models are also able to handle two-phase regions more accurately than earlier models through the inclusion of extensions to the Darcy model.²¹ Although the phase fraction is dictated by the equilibrium phase diagram in most of the models, it has been shown that non-equilibrium effects are needed to accurately match experiments.²²

Most current modelling efforts use a set of either continuum or averaged conservation equations. Continuum type equations as in Bennon and Incropera^{20,59} treat individual phases separately while Beckermann et al²¹ use averages for conserved

quantities for given computational volumes. Although there are differences in the resulting equation sets,⁶⁰ the numerical implementation of both approaches is similar. These models have been shown to be adequate to qualitatively describe various solidification processes, but lacking in quantitative detail. Lowry has shown that additional relations are necessary for macroscopic modelling of solidification in which there are significant contributions from micro-scale phenomena.²² Ni and Beckermann have also stated the need for additional micro-scale relations and recommended procedures to incorporate them into existing macro-modelling efforts.⁵¹

2.2 CONVECTIVE STABILITY

The stability of fluid systems is of importance in many engineering fields. A body that is subjected to aerodynamic drag forces, for example, will be designed with thought of the flow characteristics. Cloud formation,¹⁵ wind and ocean currents,⁶¹ and electronics cooling⁶² are a few of the many examples in which convective stability is very important. The characteristics of these flows are likewise important, and stability analyses are often performed to aid in identifying them.

The solution to the conservation equations and boundary conditions for a given problem may not be unique. This gives rise to situations in which several solutions and flow characteristics are possible for a particular set of conditions. A stability analysis determines which of the solutions is most likely to occur. The difference between the flow characteristics for the possible solutions can be significant. For example, the

difference between laminar and turbulent flow is significant when the drag forces on a given body are compared. Similarly, the difference between a quiescent and a dynamic fluid layer is significant in terms of the difference in energy transport for each system.

A stability analysis seeks to determine whether or not a base state solution is stable to perturbations from that state.^{28,63,64} If another solution is more likely, the perturbations will result in that solution, otherwise the perturbations will decay and the base solution will persist. The classic example of this is the visual idea of a ball that is either on top of a hill, at the bottom of a valley, or on a horizontal surface. A ball at rest on top of a hill is currently in a satisfactory state of non-motion, but if the ball is perturbed, it will move to another state of equilibrium. A ball at rest at the bottom of a valley is stable to perturbations in its position and a ball on a horizontal surface idealizes neutral stability, i.e. the separation between the states at the top and bottom of the hill.

The base state solution for fluid analysis is usually either quiescent or uniform and laminar. A quiescent base case is usually subject to alternate dynamic solutions because of external accelerations such as either body forces or surface tension forces. The stratification of fluid layers is the most common quiescent base case. Consider a fluid layer that is less dense at the bottom (with the gravity vector pointed down) and more dense at the top. If displaced vertically, the light fluid wants to rise due to the difference in hydrostatic pressure arising from the density difference. If the density difference is large enough, a light element of fluid perturbed upward will continue to rise, representing another solution for this system.

If the base state is a laminar flow, the alternate solution of interest is usually a turbulent flow. This is evidenced by Kelvin-Helmholtz shear flows, flows over bodies, flows in wakes and jets, Taylor instabilities, and stratified layers with flow.^{28,63} The subject of turbulence is important in a variety of topics in fluid dynamics, most of which, unfortunately, are not well documented in terms of physical understanding.¹³ The fact that turbulence is somewhat a random phenomena makes finding solutions difficult and in many instances the emphasis is placed on identifying when a turbulent solution will predominate over a laminar one. The turbulence problem is thus particularly well suited to a stability analysis, since the nature of the alternate solution is not required.

An interesting example of hydrodynamic stability is the case of thermosolutal convection.^{65,66} This process arises when a multicomponent system has a large difference between thermal and mass diffusivities. Consider a solution that is subjected to a stabilizing temperature gradient and a destabilizing solute gradient such that the combined effect is that of a stable density gradient. Even though the system would seem to be stable with regard to perturbations, the difference in diffusivities can give rise to fluid motion. If a fluid element is displaced upward, it will quickly gain thermal energy, and will therefore be subjected to the effects of the unstable solute gradient. Convective motion will ensue. This type of phenomena occurs in the ocean currents and in other geophysical problems.

2.2.1 STABILITY EXPERIMENTS

The transition from one flow regime to another has been the subject of significant experimental work, especially in the area of flow over bodies.¹³ Wind tunnels have been used extensively to identify the transition from laminar to turbulent flow for various bodies. Reynolds' early work for the transitions occurring in internal flows resulted in the parameter set governing the transition being given his name.⁶⁷ Benard reported on the transitions occurring in a stratified fluid also subjected to surface tension gradients.⁶⁸ A subsequent analysis by Rayleigh led to the famous parameter governing buoyancy driven convection which now bears his name.⁶⁹ Others have performed experiments to examine the effects of horizontal density gradients, non-steady boundary conditions, and non-constant property values on the convective stability of stratified fluid layers.⁷⁰⁻⁷³

As discussed in Section 2.1.2, experiments concerning VDS have also been performed. The identification of the onset of buoyancy driven flows during solidification was made by McCay et al, and Hopkins.^{11,12} These studies showed the importance of the effects of the two-phase region on the transition process.

2.2.2 STABILITY MODELLING

Stability analyses have also been applied to directional solidification processes for identifying the transition from planar to dendritic growth morphologies.^{2,54,55,74} The equations for these problems were written in terms of interface energy and shape and were solved analytically.² The problems were also written in terms of conservation equations and solved numerically.^{54,55,74}

Because of the importance of convective flows in creating segregation, stability analyses have been used to identify when such flows occur.^{38,54} The analysis is similar to that of stratified fluid layers because the solute profile in the diffusion layer yields density profiles similar to those for stratified fluid layers.⁷⁵ Coriell et al modelled a planar solidification front and showed the dependence of onset of convection on the initial concentration in the melt.⁵⁴ Nandapurkar et al were the first to use the full set of coupled equations for dendritic solidification in analyzing the onset of convective flows in a Pb-Sn system.³⁸ These equations have provisions for accounting for momentum losses in the two-phase region. The equations of Nandapurkar, however, do not account for either solidification shrinkage/expansion or Forchheimer inertial effects in the two-phase region. The importance of these two effects was investigated in this dissertation research.

Stability analyses are most easily performed numerically when a linear approach is used. In such an analysis the perturbations are assumed to be infinitesimal and non-linear perturbation terms are dropped. This approach has been used successfully with experiments for both stratified layers and the transition from laminar to turbulent flow over bodies.^{28,63} Coriell et al used this approach to identify the transition to convective flow for the planar VDS of Pb-Sn.⁵³ Nandapurkar et al also used this approach for the dendritic VDS of Pb-Sn.³⁸

The importance of constituent segregation occurring during VDS and its relation to convective stability have been shown. The purpose of this research was to use linear

stability theory to model the flow transitions occurring in experiments involving VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$.

3. PROBLEM DEFINITION

The transparent metal model $\text{NH}_4\text{Cl-H}_2\text{O}$ was chosen to characterize the onset of convection occurring during VDS. It is a binary eutectic material that solidifies dendritically under nearly all conditions.⁷⁶ The solute redistribution effects which give rise to a density inversion and the porous media effects which inhibit breakdown are both characteristic of this system.^{11,12} The experimental identification of flows which develop during solidification was originally accomplished using the $\text{NH}_4\text{Cl-H}_2\text{O}$ system.⁴ It has also been used for experiments using several orientations with respect to external acceleration forces, and in experiments with unsteady acceleration forces.⁷ A variety of optical techniques have previously been utilized to quantify the $\text{NH}_4\text{Cl-H}_2\text{O}$ solidification process,⁷⁷ but perhaps the most intensive set of experiments are those ongoing in support of low-gravity experiments to study the effects of flow on dendritic growth, the NASA Casting and Solidification Technology (CAST) experiments. These experiments culminated in a space shuttle experiment aboard the International Microgravity Laboratory (IML-1) in January 1992. The optical techniques used in this project to measure concentration, temperature, and growth characteristics during VDS include the schlieren and central dark ground methods of phase contrast, interferometry, and particle tracking. Holography was used in conjunction with the first three.^{77,49}

This dissertation research extends the work previously performed under the CAST project by precise experimental quantification of those properties associated with the convective breakdown and providing interpretation of those results through the use

of a linear stability model (LSM). The LSM was specifically formulated and developed for this doctoral research. The experimental approach of the dissertation research was similar to that of previous CAST studies.^{11,30,48} A quartz cuvette was filled with a NH_4Cl -72wt% H_2O solution and then heated or cooled at the top and bottom surfaces to produce a temperature gradient. After a hold period to allow equilibration, the temperatures were lowered at a constant rate, thus initiating solidification in the vertical direction opposite the acceleration of gravity. The cuvette was placed in a Mach-Zehnder interferometer during the process to allow real-time measurement of the index of refraction of the solutal diffusion layer. From this data, the temperature and concentration changes across the layer were obtained, and the height of the two-phase region was recorded. Critical Rayleigh numbers, similar to those used by McCay et al¹¹ and Hopkins,¹² were calculated and correlated with the height of the two-phase region.

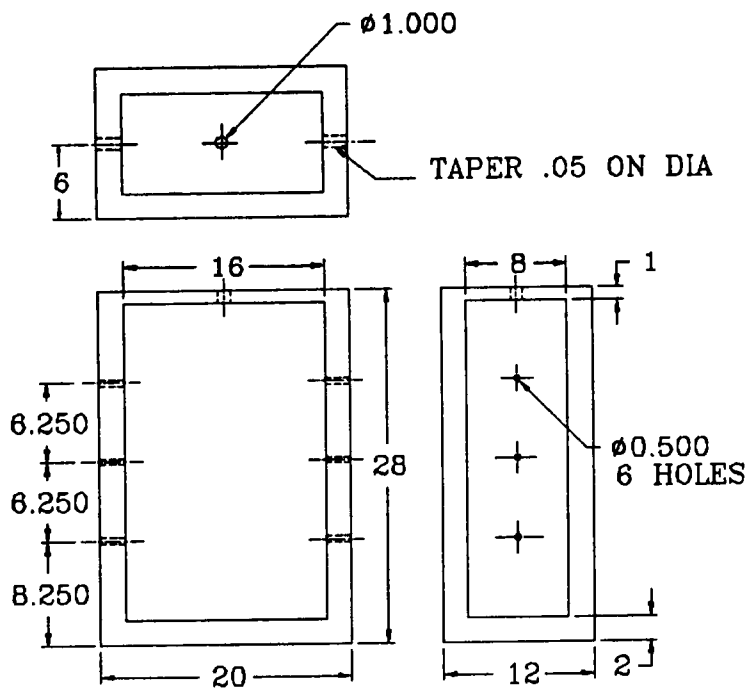
The LSM, a linear perturbation analysis, was used to predict critical Rayleigh numbers for the various growth conditions in order to more precisely quantify the effects the two-phase region has on the breakdown. This allowed evaluation of several existing permeability models for predicting the influence of the two-phase region on the breakdown process.

4. EXPERIMENTS

4.1 EXPERIMENT EQUIPMENT AND MATERIALS

To experimentally characterize the convective breakdown, a NH_4Cl -72wt% H_2O solution was placed in a quartz cuvette that was either heated or cooled as required at its top and bottom surfaces with thermoelectric devices. The cuvette and accompanying assembly were placed in a Mach-Zehnder interferometer to characterize the density changes occurring in the fluid due to the solidification process.

The cuvette, shown in Figure 4.1, was a high optical quality, rugged container appropriate for use with interferometry. Figure 4.2 shows the cuvette assembly. Thermoelectric devices (TEDs) provided the desired temperatures at the top and bottom cuvette surfaces. K-type control thermocouples of .003" diameter were placed within a slot in copper plates sandwiched between the cuvette and the thermoelectric devices. Zinc-coated water-cooled copper heat sinks were placed on opposite sides of the thermoelectric devices to aid dissipation of residual energy. A high thermal conductivity paste was used between all heated/cooled surfaces to ensure uniform thermal profiles across all areas through which energy was transferred. The paste, in conjunction with the slotted copper plates, allowed accurate measurement of the cuvette top and bottom temperatures with the thermocouples located in the slots. Figure 4.3 is an exploded view showing the specifics of the sandwiched components. Thermocouples identical to those at the top and bottom surfaces were also placed in



Dimensions in mm

Figure 4.1 Three View Drawing of Cuvette.

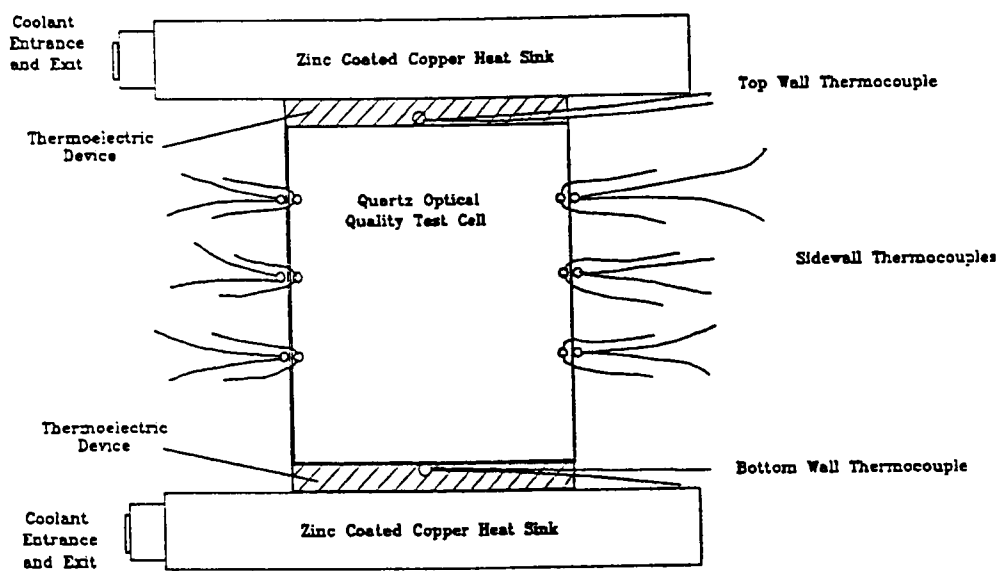


Figure 4.2 Cuvette and Solidification Assembly.

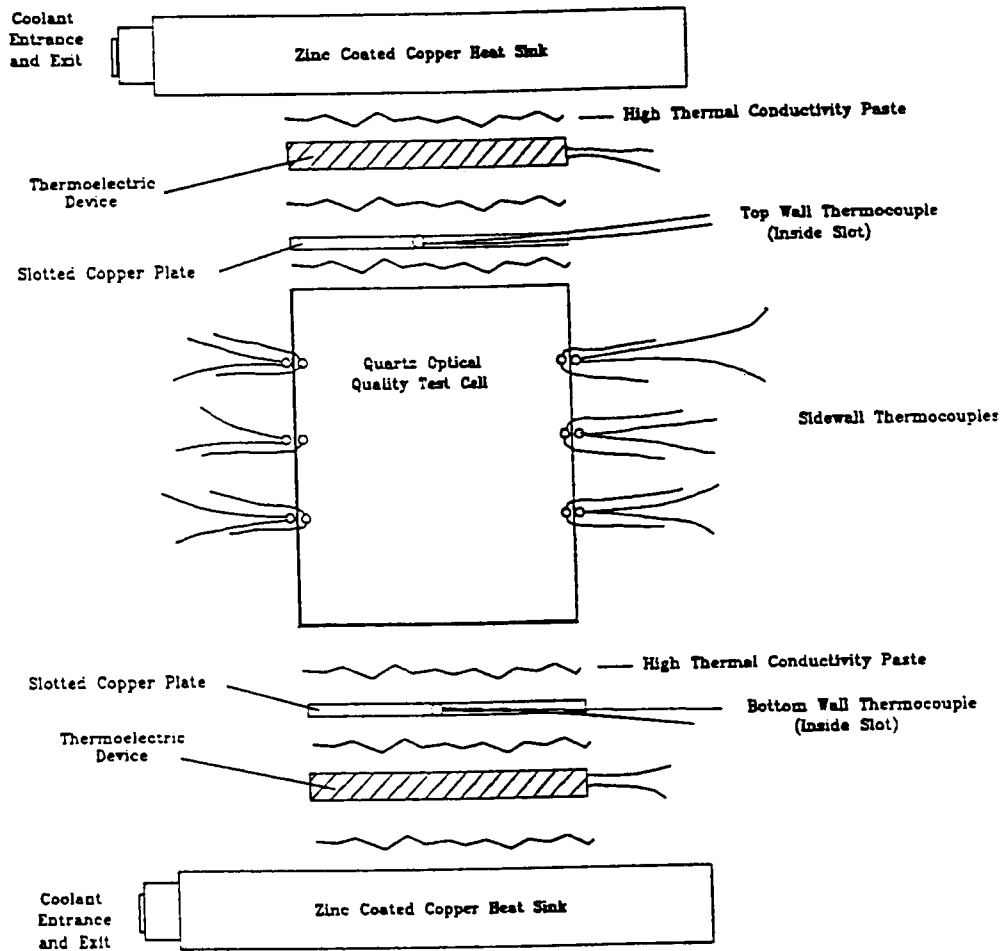


Figure 4.3 Exploded View of Cuvette and Solidification Assembly.

holes in the side walls and sealed with epoxy. These measured the temperature profile in the solution independent of the interferometer results.

A personal computer based control system was used to measure and record the thermocouple responses and send voltages to the thermoelectric devices based on these values and user requirements. A diagram of the control system is shown in Figure 4.4. The voltages were amplified using an operational amplifier circuit with 15V dc power supplies. A model 500 Keithley data acquisition system was used for A/D and D/A conversion, while an IBM PC-XT was used for the host computer. The controlling BASIC program⁷⁸ allowed the user to input desired top and bottom temperatures, time duration of set points, and cooling rates for the solidification portion of the experiment. With these user inputs, the program used measured temperatures to calculate the correct voltages for the thermoelectric devices.

The cuvette assembly was placed in a Mach-Zehnder interferometer to observe the solidification process as it occurred. The system is shown in Figure 4.5. The light source, a collimated beam from a 25 milliwatt He-Ne laser was split into two beams of approximately equal optical length, one of which passed through the cuvette, while the other was not altered. These beams were then recombined and the resulting image was recorded with a video camera. In this manner, a time history of the process was recorded for post-experiment analysis. An equipment parts list is given in Appendix A (adapted from reference 49). The optical table was mounted on vibration isolation mounts to reduce the influence of extraneous accelerations.

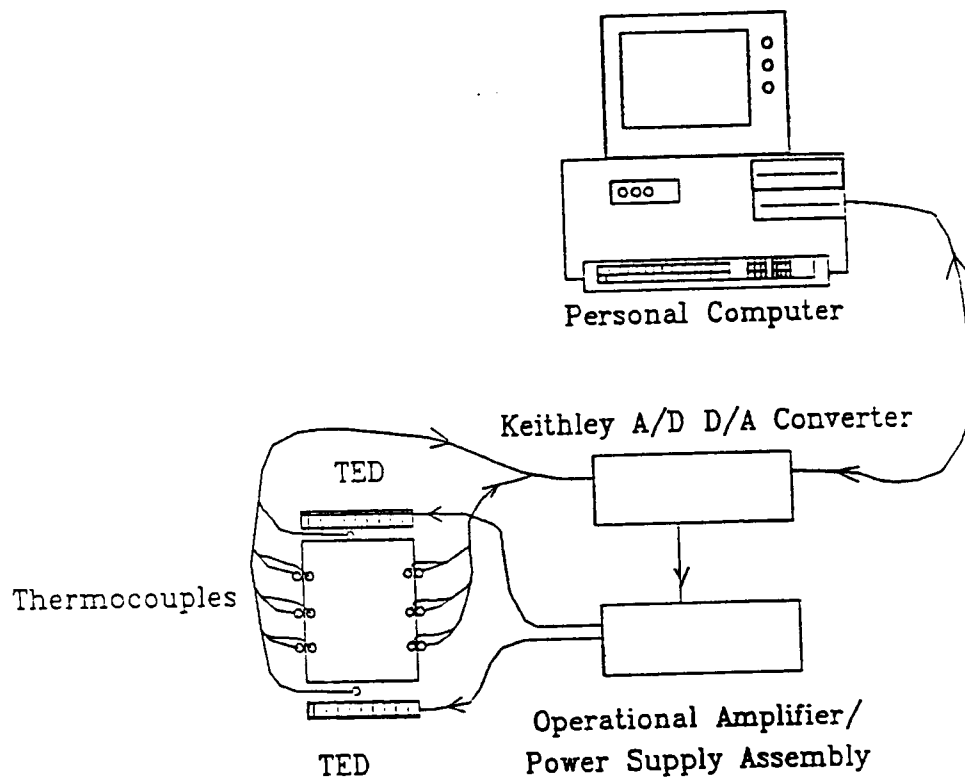


Figure 4.4 Solidification Control System.

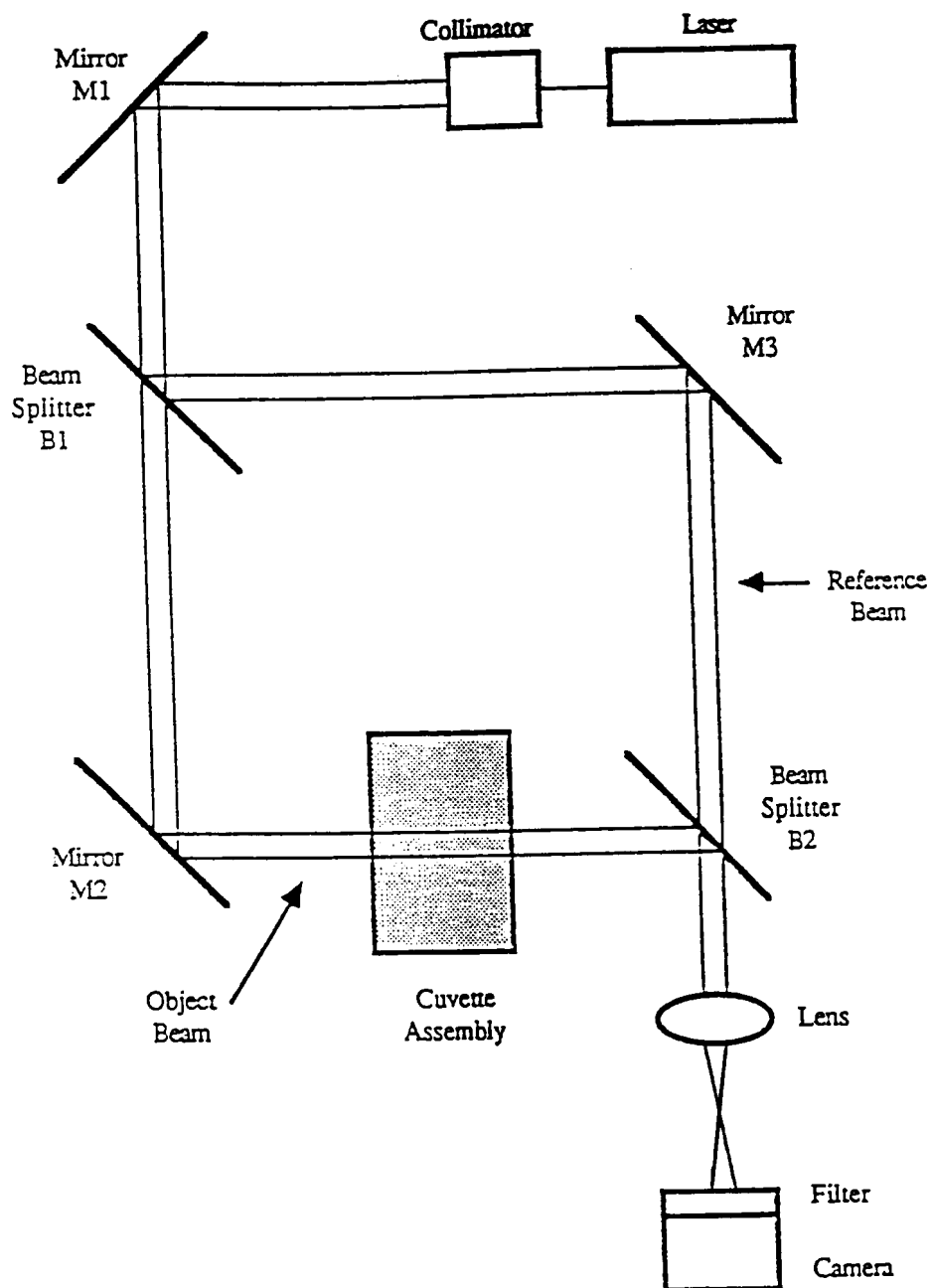


Figure 4.5 Interferometry System.⁴⁹

4.2 EXPERIMENTAL PROCEDURES

The NH_4Cl -72wt% H_2O solution was prepared by mixing appropriate amounts of Fisher deionized and ultrafiltered H_2O (W-4) and Fisher reagent grade NH_4Cl (A-649) in a 250 ml plastic laboratory bottle. Because the melting point of the solution is higher than room temperature, the solution was heated to completely dissolve any solid material before filling the cuvette. The solution was also vigorously shaken to ensure homogeneous mixture. A hypodermic syringe was then used to transfer the solution from the bottle into the cuvette through a fill hole in the top. The top was sealed with electrical tape to prevent leakage from the fill hole, and the cuvette was then placed in the cuvette assembly in the interferometer.

There were three operational modes used during the experiments: preheat, gradient hold, and cool down. Figure 4.6 shows a typical user-selected time history of the top and bottom temperatures for the experiments. The preheat mode was used at the beginning of some experiments to aid in obtaining an equilibrium status prior to cool down. This consisted of holding top and bottom temperatures at 40 °C for approximately 5 minutes. The gradient hold mode was used in all experiments for much the same purpose, to obtain an equilibrium starting point for the cool down mode. After experimentation, it was found that a gradient hold of approximately 3 hours was usually sufficient to obtain a stable two-phase region size. This was true with or without the preheat mode, so the preheat was not generally performed.

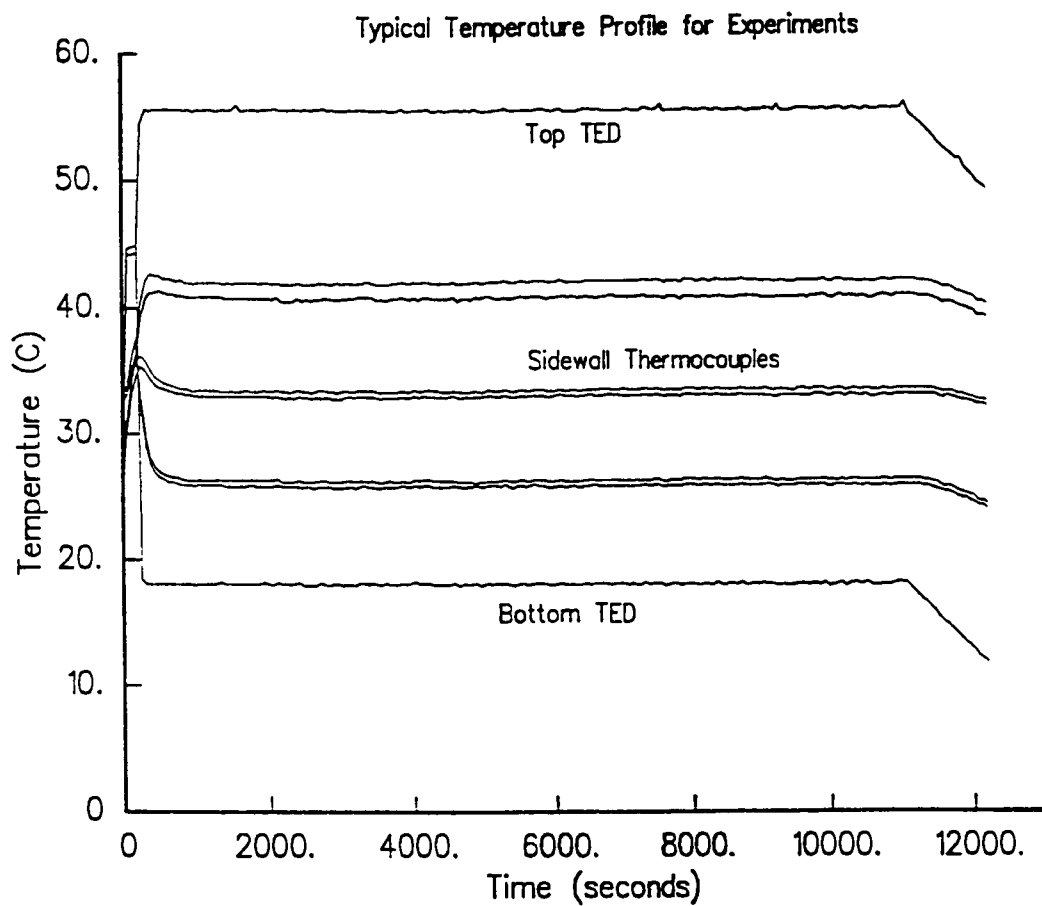


Figure 4.6 Typical Temperature Profile for Experiments.

Table 4.1 gives the number of experiments performed for each set of conditions. Four cooling rates and four bottom temperatures make up the primary variables. Ten experiments of each case were performed for a temperature gradient of $13.4\text{ }^{\circ}\text{C/cm}$. In addition, a set of experiments were performed for one of the cooling rates at each of the bottom temperatures for a temperature gradient of $25\text{ }^{\circ}\text{C/cm}$. This yielded a total of 200 experiments.

During each experiment, temperatures were recorded by the computer, and video data was recorded using a camera and a VHS format video cassette recorder. The length scale for the images was obtained from calibration marks on the cuvette walls. Figure 4.7 shows the steps performed in each experiment.

4.3 DATA ANALYSIS

4.3.1 INTRODUCTION

The primary data from the experiments were the video recordings of the interferometry images. These data were analyzed post-experiment with the aid of a digital image processor. Images at the beginning of the cool down mode and at the onset of convection were digitized for analysis. Onset of convection was identified when an undulation in the fringes in the diffusion layer appeared. This represents the transport of less dense liquid upward into a region of higher density. When this occurred, the average density for horizontal layers in that region was changed, modifying the phase shift occurring as the laser beam passed through that region.

Table 4.1 Number of Cases for Each Set of Experiment Conditions.

		G=13.4 °C/cm	G=25 °C/cm
CR=3 °C/min	Tbot=22 °C	10	-
	Tbot=21 °C	10	-
	Tbot=20 °C	10	-
	Tbot=19 °C	10	-
CR=2 °C/min	Tbot=22 °C	10	10
	Tbot=21 °C	10	10
	Tbot=20 °C	10	10
	Tbot=19 °C	10	10
CR=1 °C/min	Tbot=22 °C	10	-
	Tbot=21 °C	10	-
	Tbot=20 °C	10	-
	Tbot=19 °C	10	-
CR=0.3 °C/min	Tbot=22 °C	10	-
	Tbot=21 °C	10	-
	Tbot=20 °C	10	-
	Tbot=19 °C	10	-

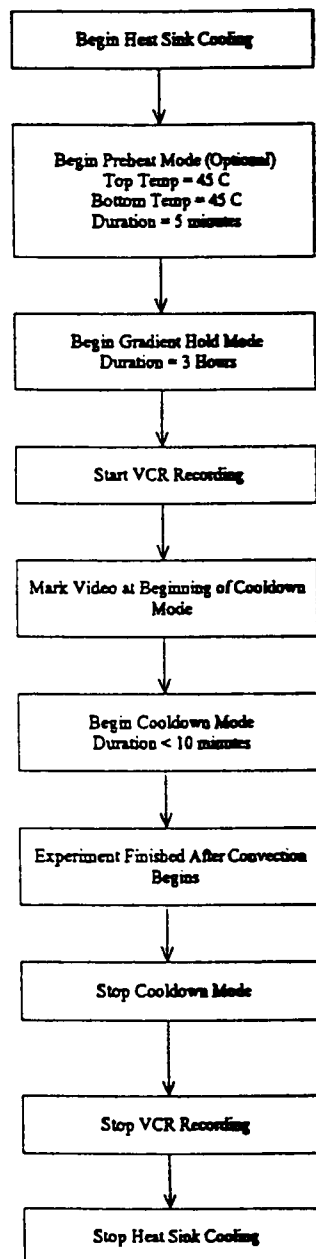


Figure 4.7 Experimental Procedure.

Figure 4.8 shows a composite (each section separated by approximately 3 seconds) interferogram during breakdown. Both the extent and vertical location of the undulation of the fringes near the solid-liquid interface increases with time due to convection.

The image at the beginning of the cool down mode was used to obtain the initial two-phase (mushy-zone) region height. The image at breakdown was used to obtain two-phase region height, the extent of the solutal diffusion layer, and the temperature and concentration differences from the bottom of the layer to the top of the diffusion layer. Figure 4.9 is a time composite of images from a typical experiment showing the evolution of the solutal diffusion layer near the solid-liquid interface prior to the onset of convection.

4.3.2 INTERFEROMETRIC DATA REDUCTION

The interferometer used in these experiments consisted of two coherent beams that initially travel along (ideally) identical optical paths. One of the beams is passed through the test region and in so experiences a shift in phase angle, ϕ , due to the different index of refraction of this region. Figure 4.10 illustrates this effect. The electric field component exiting from the test section can be written in complex time-independent notation as

$$U_o(x,z) = A_o e^{i\phi(x,z)} \quad 4.1$$

where A_o is a constant. The spatial phase shift is due to the variation of index of refraction in the test section integrated in the direction of the beam and is given as

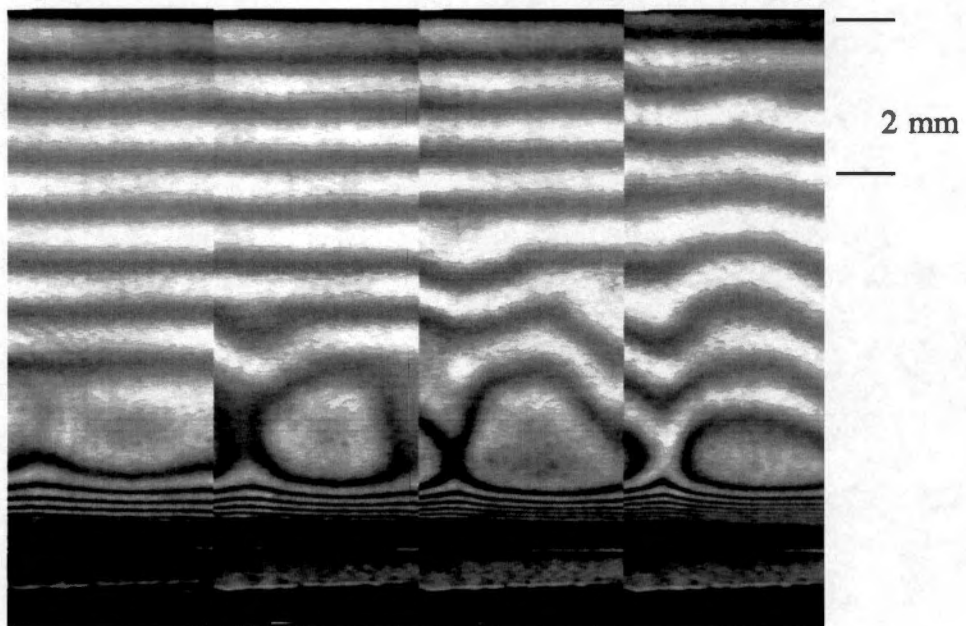


Figure 4.8 Time Composite Interferogram Illustrating Convective Breakdown.

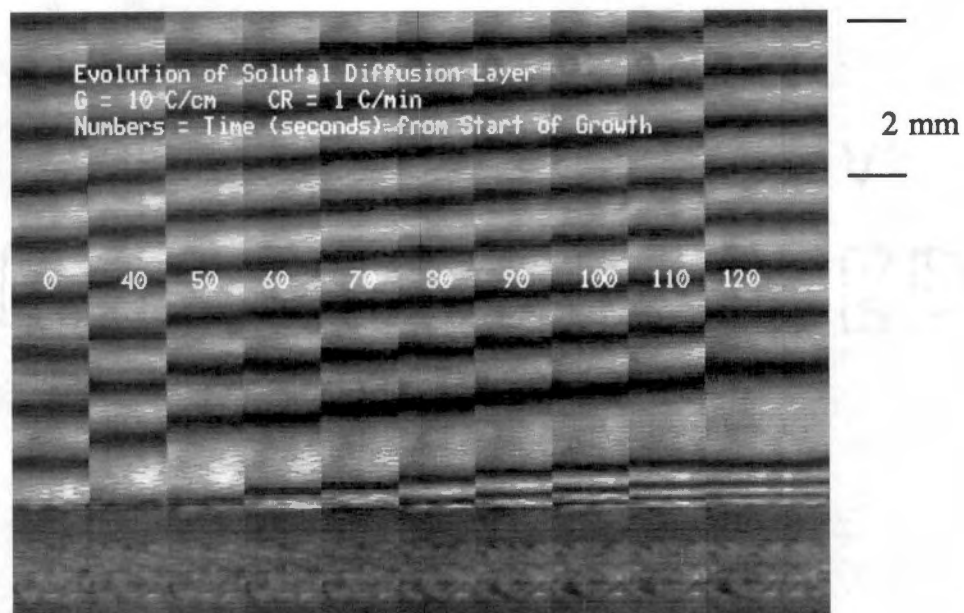


Figure 4.9 Time Composite Interferogram.

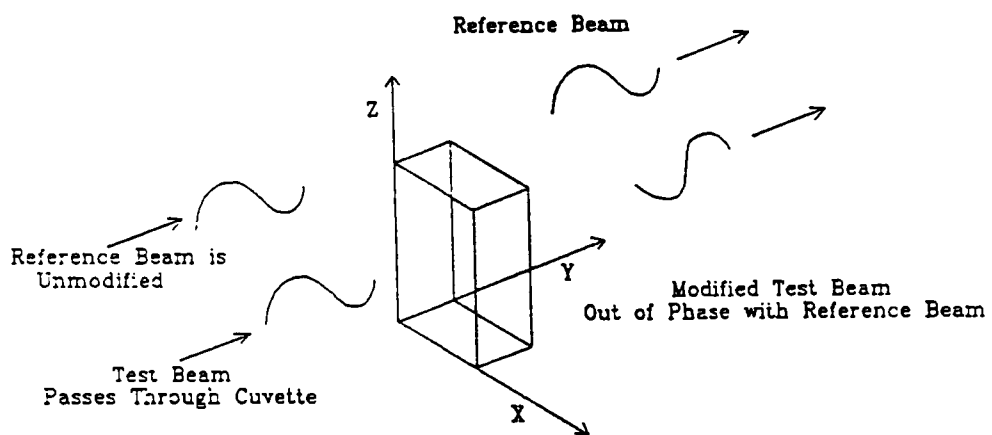


Figure 4.10 Phase Modification of Light Passing Through the Test Section.

$$\phi(x,z) - \phi_o = \frac{2\pi}{\lambda} \int_0^{\tau} [n(x,y,z) - n_o] dy \quad 4.2$$

where n_o and ϕ_o are values of index of refraction and phase angle for the reference beam. If the solidification process yields uniform variations in the plane normal to growth, the index of refraction variations in the y direction (beam propagation direction) are negligible, and the phase shift of the test beam is

$$\phi(x,z) - \phi_o = \frac{2\pi \tau}{\lambda} n(x,z) - n_o \quad 4.3$$

Upon combination with the reference beam, the resulting image intensity is (assuming equal amplitude beams)

$$I(x,z) = 2A_o \left(1 + \cos[\phi(x,z) - \phi_o] \right) \quad 4.4$$

Every 2π phase shift produces intensity extrema values, i.e. fringes, due to constructive and destructive interference between the beams. The location of the fringes, superposed on the image of the test section, gives the relative spatial phase field. This field can be used to calculate index of refraction variations in the test section using

$$\Delta n = \frac{\lambda}{2\pi \tau} \Delta \phi \quad 4.5$$

The index of refraction of the solution as a function of temperature and concentration has been measured for temperatures and concentrations of interest for

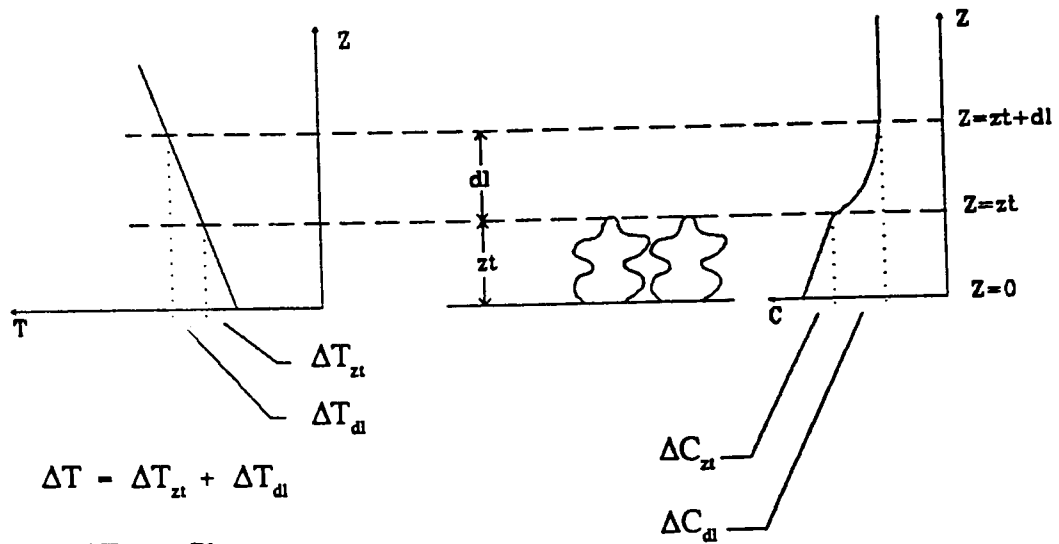
this study and is given by^{23,11}

$$\begin{aligned} n(T,C) = & 1.337 - 1.73 \times 10^{-4} (T - 22 \text{ }^{\circ}\text{C}) \\ & + 1.63 \times 10^{-3} (C - 72.93 \text{ wt}\%) \end{aligned} \quad 4.6$$

To characterize the breakdown in terms of the temperature and concentration profiles and the height of the two-phase region, the phase distribution determined from the interference fringes was used to calculate the temperature and solutal profiles over the extent of the density inversion. Figure 4.11 shows the contribution that both the two-phase and the fluid regions have on the concentration and temperature differences used to characterize the breakdown.

Equation 4.5 was applied to the fluid above the solutal diffusion layer, where only temperature is varying, to obtain a temperature gradient. This was accomplished by interactively measuring the distances between visually identified fringe centers using the digital image processor and employing equations 4.5 and 4.6. These equations give a temperature difference between fringes (a 2π phase shift) of 0.4572 $^{\circ}\text{C}$. This temperature gradient was assumed valid for the region below the density inversion since the thermal diffusivity is high and the diffusion layer heights were small ($< 0.1 \text{ cm}$) for this study.

Concentration differences in the solutal diffusion layer were then obtained by using visually identified fringes near the interface to calculate the index of refraction change in this region and then removing temperature effects using the previously measured temperature gradient. The point of reference for the index of refraction



$$\Delta T = \Delta T_{zt} + \Delta T_{dl}$$

$$\Delta T_{zt} = -Gl \, zt$$

$$\Delta T_{dl} = -Gl \, dl$$

$$\Delta C = \Delta C_{zt} + \Delta C_{dl}$$

$$\Delta C_{zt} = \frac{1}{m} \Delta T_{zt} = -\frac{1}{m} Gl \, zt$$

$$\Delta n_{dl} = (\# \text{ fringes})_{dl} \frac{\lambda}{\tau} = -1.73 \times 10^{-4} \Delta T_{dl} + 1.63 \times 10^{-3} \Delta C_{dl}$$

$$\Delta C_{dl} = \frac{1}{1.63 \times 10^{-3}} \left[(\# \text{ fringes})_{dl} \frac{\lambda}{\tau} + 1.73 \times 10^{-4} \Delta T_{dl} \right]$$

Figure 4.11 Temperature and Concentration Differences in Experiments.

difference was the maximum density point, the center of the widest fringe. The difference was obtained by counting the number of opposite intensity fringes located between this point and the solid-liquid interface, determining the phase shift as a discrete (integer multiples of π) quantity. In the two-phase region, thermodynamic equilibrium was assumed and the concentration difference was calculated using the temperature gradient and the equilibrium phase diagram. These differences were used to calculate critical Rayleigh numbers for the system.

The thermal Rayleigh number is given as

$$Ra_T = \frac{g B_T \Delta T L^3}{\nu \alpha} \quad 4.7$$

and the solutal Rayleigh number is

$$Ra_S = \frac{g B_C \Delta C L^3}{\nu D} \quad 4.8$$

The stability of the system is dependent on the sum of these individual numbers. The effects of the two-phase region were shown by correlating the combined Rayleigh number with the height of the two-phase region at breakdown.

4.3.3 MEASUREMENT UNCERTAINTY

The uncertainty in measuring phase angle and distances was dependent on several factors. These included the resolution from the video recordings, optical imperfections and noise, and errors in human judgement. Because the measurements are dependent on human judgement, estimating the measurement uncertainty is difficult. Two approaches were used. In one, measurement values were selected

which would give a bound for the maximum error, and in the other, uncertainty percentages were calculated based on the estimated measurement error relative to an appropriate mean value for that measurement.

The estimated worst case value for length measurements was based on the vertical resolution of the camera of 242 lines which was displayed and analyzed on a 512 pixel grid. The worst case uncertainty in measuring lengths was estimated to be bounded by ± 5 pixels. The minimum value for characteristic length, L , was 0.083 cm. This corresponded to approximately 75 pixels on the digitized video image. This gives an estimated error in measuring L of ± 0.0055 cm (approximately 6.7 %). The minimum length associated with calculating the temperature gradient was approximately 0.035 cm. Substituting the estimated error in distance measurements yields an estimated error of ± 1.75 °C/cm (approximately 13%) in measuring temperature gradient, hence, the uncertainty of measuring ΔT was 20%. The error in measuring total phase difference in the fluid diffusion layer was dependent on accurately counting the number of fringes, the error of which is difficult to determine precisely. A crude estimate was ± 0.5 fringes with a minimum fringe count of 1.5. Substituting these values for # fringes and ΔT , the uncertainty in measurement of ΔC_{dl} was 24%. The uncertainty in ΔC_{zt} was the same as for ΔT_{zt} . Substituting these values into equations 4.5-4.8 yielded a total worst-case estimated error in measuring the critical combined Rayleigh number of 53%.

Engineering estimates of the probable measurement uncertainties yielded a more reasonable value for the total error. A more representative value of ± 2 pixels

was used as the estimated resolution in identifying fringe locations. The average value for the characteristic length, L , was approximately 0.1 cm. This corresponded to approximately 90 pixels on the digitized video image. The estimated error of ± 2 pixels in locating screen objects gives an error in distance measurements of ± 0.02 mm (approximately 2%). The average length associated with calculating the temperature gradient was approximately 0.045 cm. Substituting the estimated error in distance measurements gives an estimated error of ± 0.5 °C/cm (approximately 5%) in measuring temperature gradient, hence, the uncertainty in measuring ΔT was 7%. The error in counting the number of fringes was again estimated to be 0.5 fringes with a typical fringe count of 3.5. Substituting these values for # fringes and ΔT , the uncertainty in measurement of ΔC_{dl} was 13%. The uncertainty in ΔC_{zl} was the same as that for ΔT_{zl} . Substituting these values into equations 4.5-4.8 yielded a total estimated error in measuring the critical combined Rayleigh numbers of approximately $\pm 10\%$.

4.4 RESULTS

The combined Rayleigh number at breakdown was determined as a function of the two-phase region for each set of experimental conditions. The data were fitted to an exponential fit in the form

$$Ra_c = Ra_o e^{(b_l h_{mbd})} \quad 4.9$$

where Ra_o and b_l were determined from least squares fitting. Figures 4.12- 4.16 present the data for different cooling rates. The effect of the mushy zone on critical combined Rayleigh number is quite evident. The critical combined Rayleigh number increases exponentially with the mushy zone height.

Although some of the spread in data can be attributed to measurement errors, the makeup of the mushy zone is believed to be the most significant contributor to the variation in experimental measurements. The mushy zone is a complicated structure of solid members which are thermodynamically active and continually evolving with time.³⁵ Thus two-phase regions of the same extent (height), but with different mushy zone characteristics, could exhibit different resistances to flow. Hence the structure of the dendrites in the two-phase region is not constant with respect to time, contrary to the common assumption in most solidification studies.^{4,6,7,11,12,79} These factors become significant when characterizing the effects of size on the two-phase region's permeability.

The two-phase structure, due to coarsening effects, is not in true equilibrium for any short duration experiment, and it is believed that the variation in porous media effects due to structure differences is the primary cause of the scatter in the data. The experimental values for two-phase region height and Rayleigh numbers are given in tabular form in Appendix B.

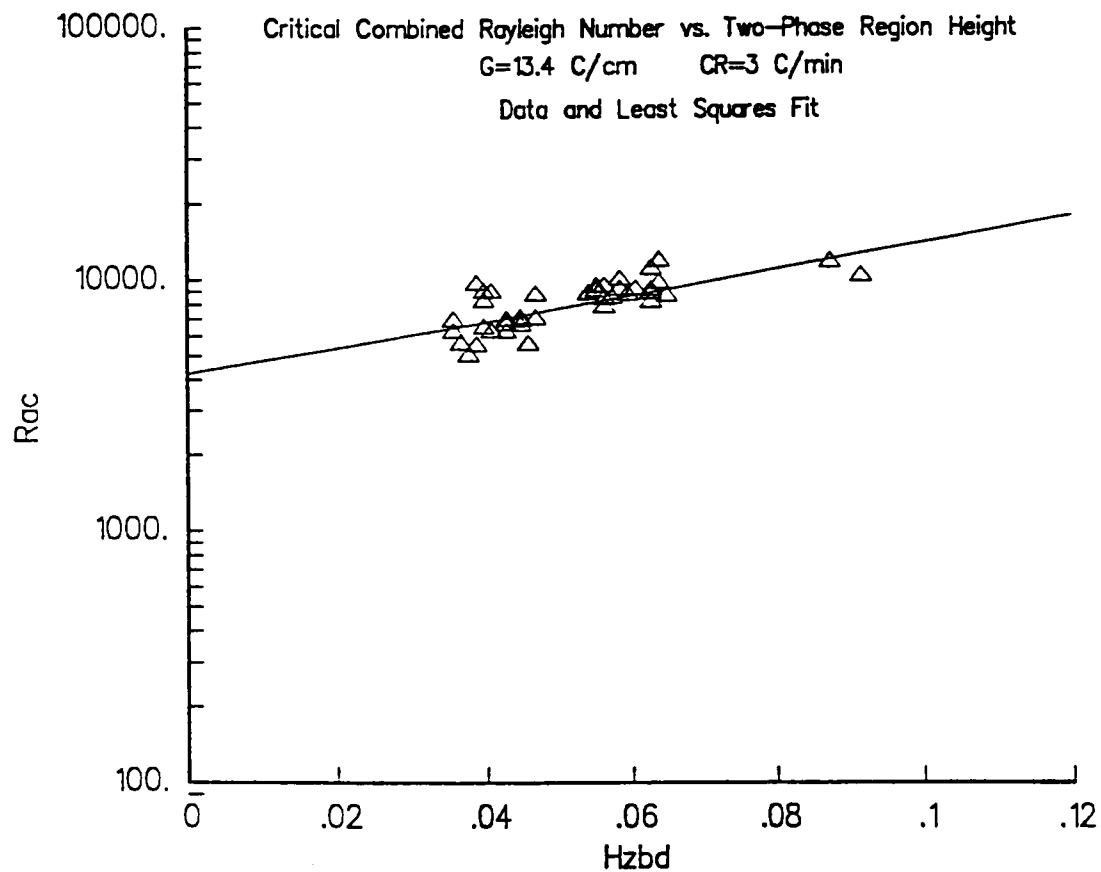


Figure 4.12 Experimentally Measured Critical Rayleigh Numbers vs. Two-Phase Region Height $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=3 \text{ } ^\circ\text{C/min}$.

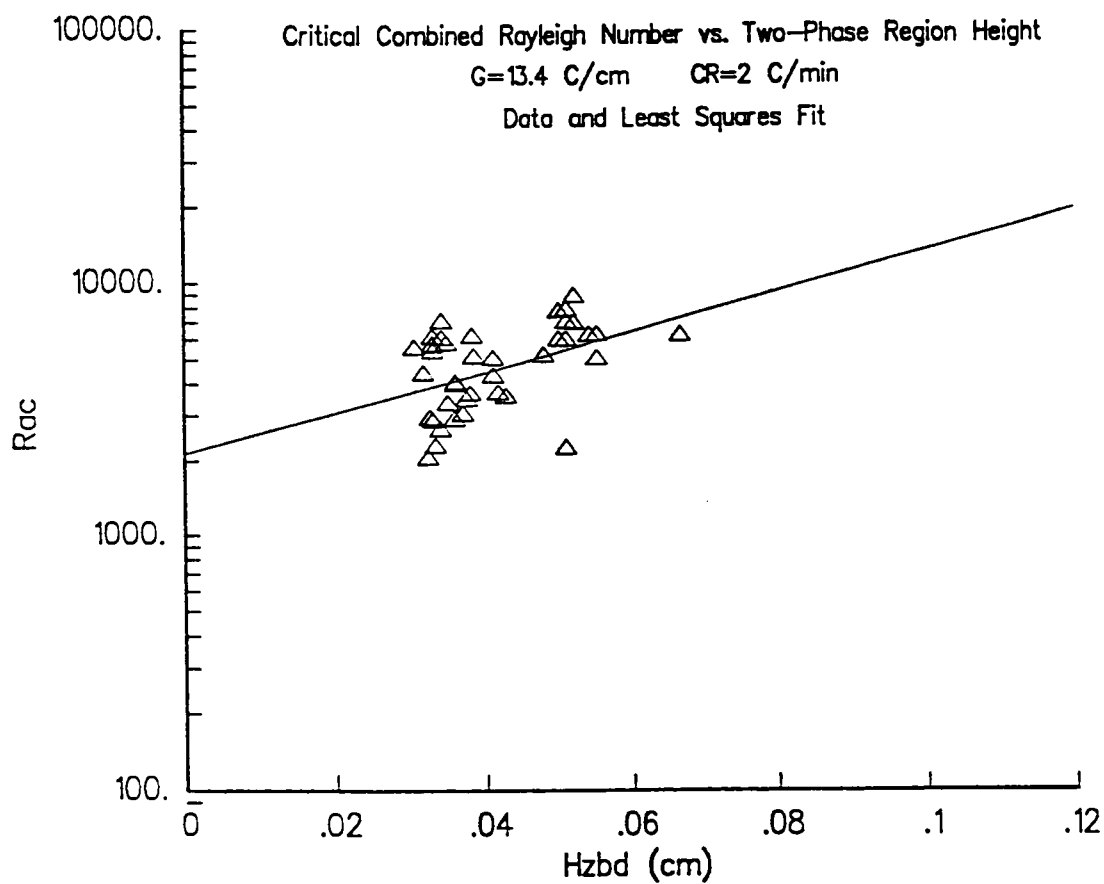


Figure 4.13 Experimentally Measured Critical Rayleigh Numbers vs. Two-Phase Region Height $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=2 \text{ } ^\circ\text{C/min}$.

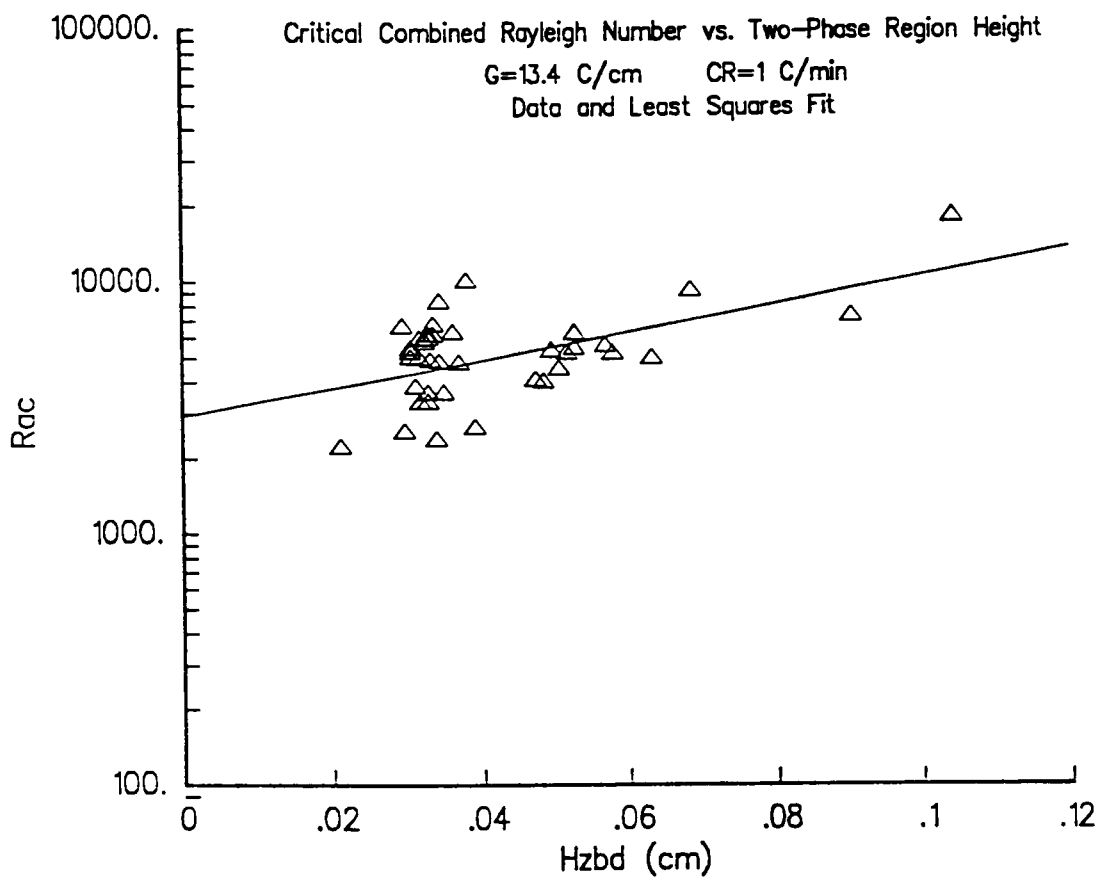


Figure 4.14 Experimentally Measured Critical Rayleigh Numbers vs. Two-Phase Region Height $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=1 \text{ } ^\circ\text{C/min}$.

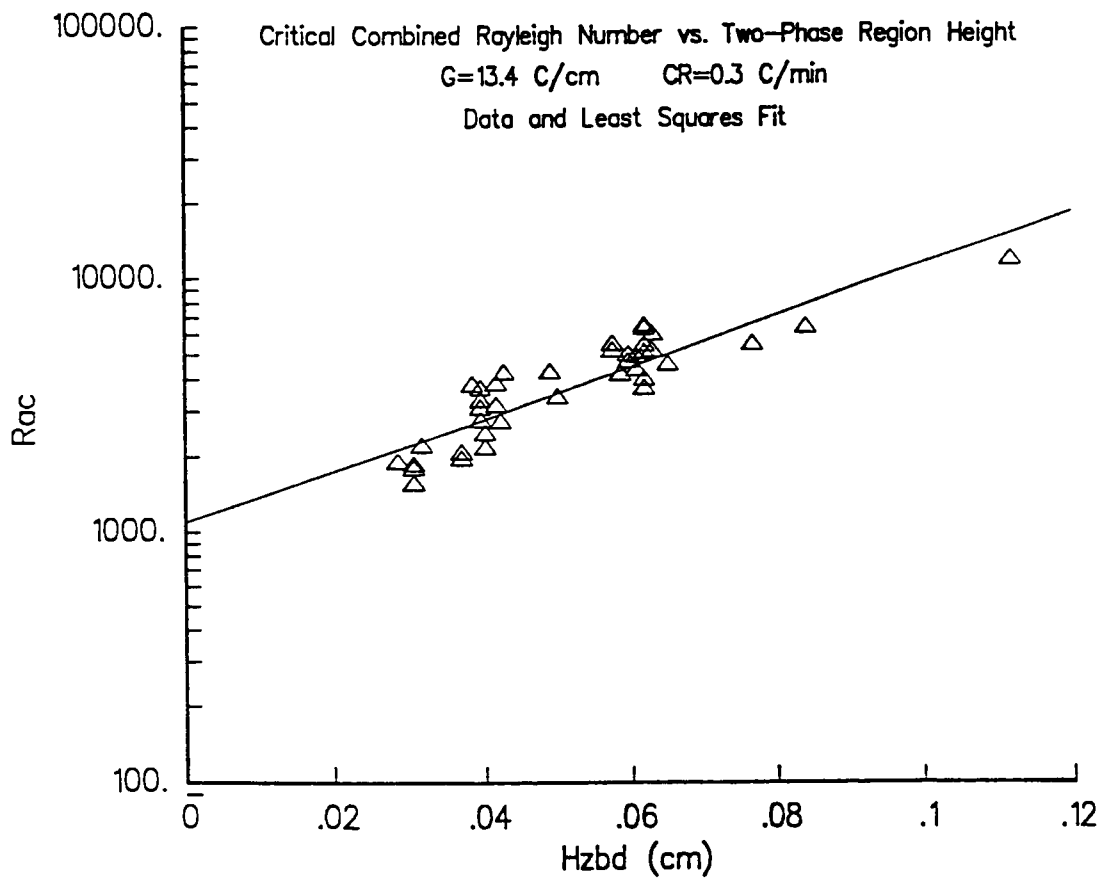


Figure 4.15 Experimentally Measured Critical Rayleigh Numbers vs. Two-Phase Region Height $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=0.3 \text{ } ^\circ\text{C/min}$.

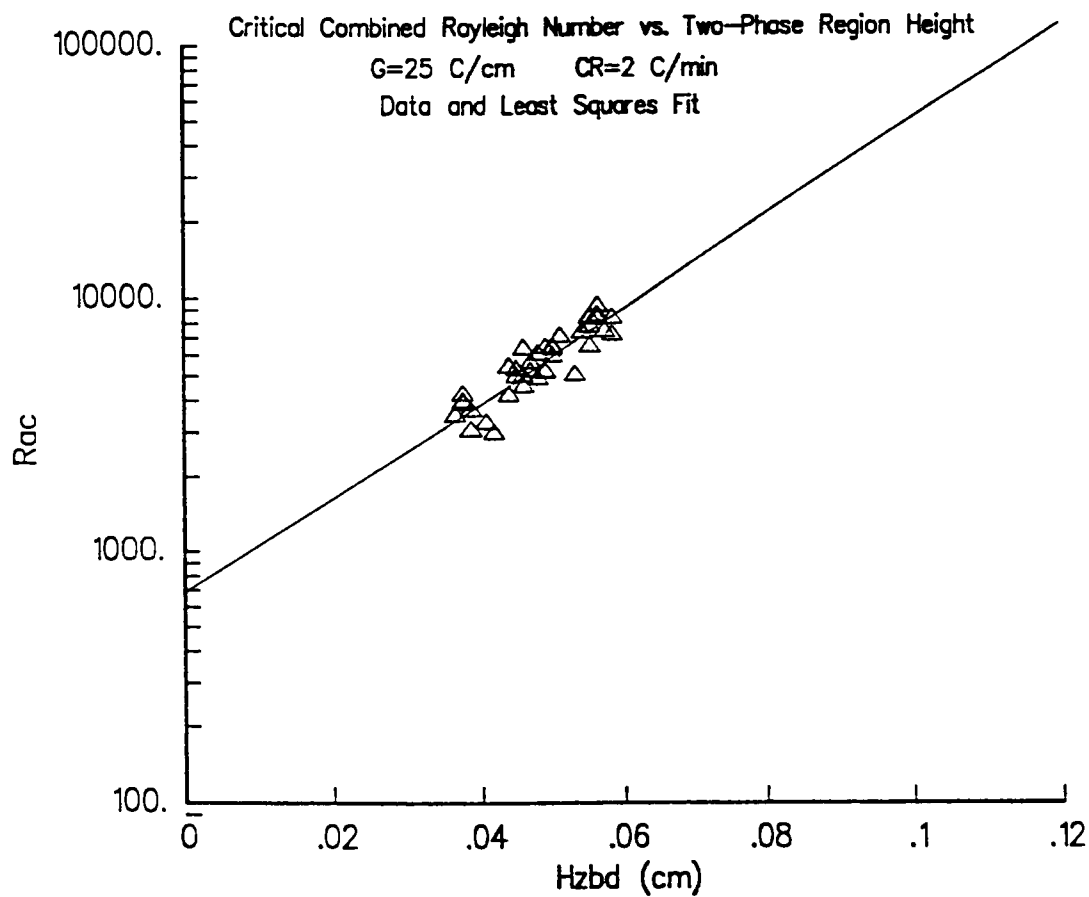


Figure 4.16 Experimentally Measured Critical Rayleigh Numbers vs. Two-Phase Region Height $G=25$ °C/cm $CR=2$ °C/min.

5. NUMERICAL STUDY

5.1 CONSERVATION EQUATIONS

A linear perturbation analysis was used to predict the stability of the VDS process. The conservation equations used in this study are similar to those given by Ganesan and Poirier⁶⁰ and include both an allowance for a shrinkage flow due to density differences between the liquid and solid phases and a first order approximation of the Forchheimer extension to the Darcy-Brinkman momentum equation. The governing equations consist of conservation equations for mass, momentum, energy, and solute and have been used in various forms for specific applications.^{38,50,80-82} The derivations presented in this chapter follow approximately those in Nandapurkar et al³⁸ but the differences will be emphasized.

The problem domain used in this study is shown in Figure 5.1. The equations are written as being two-dimensional with infinite x boundaries, while z also approaches infinity (far field conditions) in the positive direction. The gravity vector is aligned in the negative z direction and solidification proceeds at a constant rate in the positive z direction. The height of the two-phase region is given by z_t , the extent of the solutal diffusion layer is dl , and the fluid layer height is z_l . Boundary conditions are specified for $z=0$, $z=z_t$, and $z=z_t+z_l$, where z_t and z_l are problem dependent.

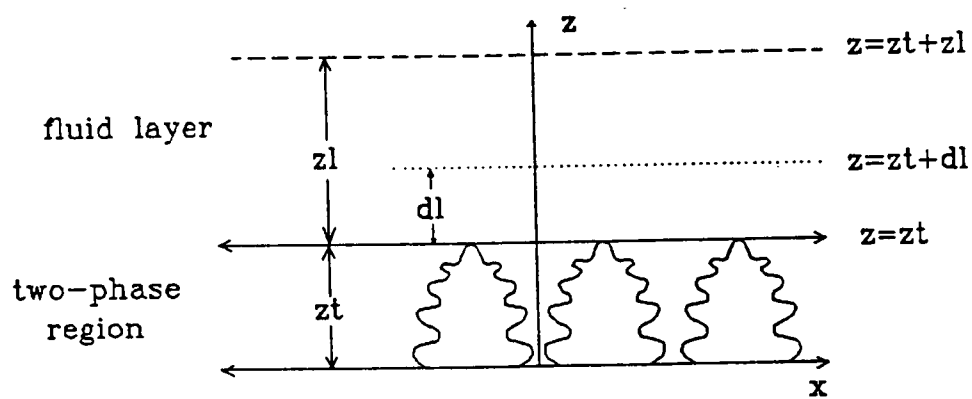


Figure 5.1 Domain Definition.

In order to model the solidification of $\text{NH}_4\text{Cl-H}_2\text{O}$, the conservation of mass (continuity) equation has an additional term which accounts for the difference in the liquid and solid densities.³³ Thus the continuity equation becomes

$$\frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} = \gamma \frac{\partial \Phi}{\partial t} \quad 5.1$$

where

$$\gamma = \frac{\rho_s - \rho_l}{\rho_s} \quad 5.2$$

This gives rise to fluid flow if the fraction of liquid is changing with time.

The momentum equation is written in terms of velocities that are volume averaged quantities for the two-phase region and approach true velocity values as the fraction of liquid approaches one in the fluid layer. To account for inertia losses in the two-phase region, a modified Ergun model was used for the Forchheimer extension.⁴³ The equation for the x momentum is given by

$$\begin{aligned} \frac{\partial u}{\partial t} + \frac{1}{\Phi} \left(u \frac{\partial u}{\partial x} + w \frac{\partial u}{\partial z} \right) = & - \frac{\Phi}{\rho_o} \frac{\partial P}{\partial x} + \nu \nabla^2 u \\ & - \frac{\nu \Phi}{\kappa} u - F \Phi \left| \bar{V} \right| u \end{aligned} \quad 5.3$$

where the left-hand side consists of the convective terms and the right-hand side consists of terms for pressure, viscous (Brinkman), Darcy, and inertia (Forchheimer) forces. The z momentum equation is similar with an additional term for the buoyancy

force. The equation is given as

$$\begin{aligned} \frac{\partial w}{\partial t} + \frac{1}{\Phi} \left(u \frac{\partial w}{\partial x} + w \frac{\partial w}{\partial z} \right) = & - \frac{\Phi}{\rho_o} \frac{\partial P}{\partial z} + \nu \nabla^2 w \\ & - \frac{\nu \Phi}{\kappa} w - \Phi \frac{\rho}{\rho_o} g \\ & - F \Phi \left| \bar{V} \right| w \end{aligned} \quad 5.4$$

F is given as a simplified representation of the Ergun model in which it is a function of a characteristic length associated with the geometry of the porous medium, d, and the fraction of liquid ϕ .⁸³ F is defined as

$$F = \frac{1.75(1-\Phi)}{\Phi^3 d} \quad 5.5$$

The fraction of liquid is a significant factor in these equations since the permeability is a function of the fraction liquid. Various functional forms for this dependence will be discussed for each of the two-phase problems solved. The equations are equally valid for both the two-phase and all liquid regions since the fraction of liquid becomes one, and the permeability approaches infinity in the fluid layer.

The energy equation is written in terms of temperature and is given by

$$\frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + w \frac{\partial T}{\partial z} = \alpha \nabla^2 T - \frac{L_f}{c_p} \frac{\partial \Phi}{\partial t} \quad 5.6$$

The time rate of change of energy in a differential volume is balanced by the energy

leaving due to convection and conduction and by the latent energy associated with the phase change process.

The solute equation is an extension of the solute redistribution model of Flemings.⁴⁵ Diffusion of material in the liquid phase is added using Fick's law. Note that this equation also contains the difference in phase densities. The equation is

$$\begin{aligned} \Phi \frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} + w \frac{\partial C}{\partial z} - D \left[\frac{\partial}{\partial x} \left(\Phi \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial z} \left(\Phi \frac{\partial C}{\partial z} \right) \right] \\ - \frac{(1-k)}{(1-\gamma)} \frac{\partial \Phi}{\partial t} C \end{aligned} \quad 5.7$$

Equations 5.1 - 5.7 represent the conservation principles necessary to determine the convective stability of a solidifying system. The solidification velocity is held constant and to make the equations more tractable, they are transformed to a coordinate system moving with the solidifying interface at the solidification velocity V . The differential form of the transformation is given by

$$\frac{\partial}{\partial z} = \frac{\partial}{\partial z'} \quad ; \quad \frac{\partial}{\partial t} = \frac{\partial}{\partial t'} - V \frac{\partial}{\partial z'} \quad 5.8$$

where the primes represent the moving coordinate system. Substituting into 5.1 - 5.7 and dropping the primes for convenience yields the following equations for the moving coordinate system

$$\frac{\partial u}{\partial x} + \frac{\partial w}{\partial z} - \gamma \frac{\partial \Phi}{\partial t} - \gamma V \frac{\partial \Phi}{\partial z} \quad 5.9$$

$$\begin{aligned} \frac{\partial u}{\partial t} - V \frac{\partial u}{\partial z} + \frac{1}{\Phi} \left(u \frac{\partial u}{\partial x} + w \frac{\partial u}{\partial z} \right) - \frac{\Phi}{\rho_o} \frac{\partial P}{\partial x} + \nu \nabla^2 u \\ - \frac{\nu \Phi}{\kappa} u - F \Phi \left| \bar{V} \right| u \end{aligned} \quad 5.10$$

$$\begin{aligned} \frac{\partial w}{\partial t} - V \frac{\partial w}{\partial z} + \frac{1}{\Phi} \left(u \frac{\partial w}{\partial x} + w \frac{\partial w}{\partial z} \right) - \frac{\Phi}{\rho_o} \frac{\partial P}{\partial z} + \nu \nabla^2 w \\ - \frac{\nu \Phi}{\kappa} w - \Phi \frac{\rho}{\rho_o} g \\ - F \Phi \left| \bar{V} \right| w \end{aligned} \quad 5.11$$

$$\frac{\partial T}{\partial t} - V \frac{\partial T}{\partial z} + u \frac{\partial T}{\partial x} + w \frac{\partial T}{\partial z} - \alpha \nabla^2 T - \frac{L_f}{c_p} \frac{\partial \Phi}{\partial t} + V \frac{L_f}{c_p} \frac{\partial \Phi}{\partial z} \quad 5.12$$

$$\Phi \frac{\partial C}{\partial t} - V \Phi \frac{\partial C}{\partial z} + u \frac{\partial C}{\partial x} + w \frac{\partial C}{\partial z} -$$

$$\begin{aligned} D \left[\frac{\partial}{\partial x} \left(\Phi \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial z} \left(\Phi \frac{\partial C}{\partial z} \right) \right] \\ - \frac{(1-k)}{(1-\gamma)} \frac{\partial \Phi}{\partial t} C + V \frac{(1-k)}{(1-\gamma)} \frac{\partial \Phi}{\partial z} C \end{aligned} \quad 5.13$$

The general boundary conditions for equations 5.9-5.13 are

$$\begin{aligned}
z = 0, \quad T = T_o, \quad C = C_o, \quad u = w = 0 \\
z = z_t + z_l, \quad T = T_\infty, \quad C = C_\infty, \quad u = w = 0
\end{aligned}
\tag{5.14}$$

Specific modifications to the boundary conditions will be given for each application.

The approach in a linear stability analysis is to express certain variables in the fundamental equations in terms of a sum of a steady (time independent) base solution and a time varying perturbation solution. The base solution is obtained and extracted from the perturbation equations, and it is determined whether the perturbation solution is growing, constant, or decaying with time. This latter step is usually accomplished by assuming the form of the perturbation solution and solving for the case in which the solution is marginally stable, i.e. neither growing nor decaying with time.

5.2 BASE STATE SOLUTION

The perturbation variables in this study were velocity, pressure, temperature, concentration, and density. The base solution was defined as having spatial variations in the z direction only. With this assumption the equations governing the base solution are obtained from equations 5.9 - 5.13 yielding the following for conservation of mass, x momentum, z momentum, energy, and solute, respectively

$$\frac{dw_s}{dz} = -\gamma V \frac{d\Phi_s}{dz}
\tag{5.15}$$

$$\frac{\partial P_s}{\partial x} = 0 \quad 5.16$$

$$\begin{aligned} -V \frac{dw_s}{dz} + \frac{w_s}{\Phi_s} \frac{dw_s}{dz} - \frac{\Phi_s}{\rho_o} \frac{dP_s}{dz} + v \frac{d^2 w_s}{dz^2} - \frac{v \Phi_s}{\kappa} w_s \\ - \Phi_s \frac{\rho}{\rho_s} g - F \Phi (w_s)^2 \end{aligned} \quad 5.17$$

$$\frac{d^2 T_s}{dz^2} + \frac{(w_s - V)}{\alpha} \frac{dT_s}{dz} + \frac{VL_f}{c_p \alpha} \frac{d\Phi_s}{dz} = 0 \quad 5.18$$

$$\begin{aligned} \frac{d^2 C_s}{dz^2} + \frac{(w_s - V)}{D} \frac{dC_s}{dz} + \frac{1}{\Phi_s} \frac{d\Phi_s}{dz} \frac{dC_s}{dz} \\ + \frac{(1-k)}{(1-\gamma)} \frac{V}{\Phi_s D} \frac{d\Phi_s}{dz} C_s = 0 \end{aligned} \quad 5.19$$

The inclusion of the shrinkage term in equation 5.15 means that there will be a finite base state velocity w_s if γ is non-zero. The energy equation can be rewritten in the two-phase region by using the liquidus slope from the equilibrium phase diagram

$$m = \frac{dT}{dC} \quad 5.20$$

This yields from equations 5.18 and 5.19, respectively

$$\frac{d\Phi_s}{dz} = \frac{-mc_p\alpha}{VL_f} \left(\frac{d^2C_s}{dz^2} + \frac{(V-w_s)}{\alpha} \frac{dC_s}{dz} \right) \quad 5.21$$

$$\frac{d^2C_s}{dz^2} = \frac{-\left[\frac{V-w_s}{D} - \frac{(1-k)}{(1-\gamma)} \frac{(V-w_s)mc_p}{\Phi_s DL_f} C - \frac{(V-w_s)mc_p}{V\Phi_s L_f} \frac{dC_s}{dz} \right] \frac{dC_s}{dz}}{\left[1 - \frac{mc_p\alpha}{L_f V \Phi_s} \frac{dC_s}{dz} - \frac{(1-k)}{(1-\gamma)} \frac{mc_p\alpha}{\Phi_s DL_f} C \right]} \quad 5.22$$

Equations 5.15, 5.21, and 5.22 can be solved simultaneously for contraction flow velocity, concentration, fraction liquid, and, using the equilibrium phase diagram, temperature in the two-phase region. The boundary conditions for these equations are

$$\Phi_s(zl) = 1, \quad C_s(zl) = C_l, \quad \left(\frac{dC_s}{dz} \right)_{zl} = \frac{1}{m} G_l, \quad w_s(0) = 0 \quad 5.23$$

The base state temperature and solutal profiles in the all fluid region are given by the following equations and boundary conditions

$$\frac{d^2T_s}{dz^2} + \frac{v-w_s}{\alpha} \frac{dT_s}{dz} = 0 \quad T_s(zl) = T_l, \quad \left(\frac{dT_s}{dz} \right)_{zl} = G_l \quad 5.24$$

$$\frac{d^2C_s}{dz^2} + \frac{v-w_s}{D} \frac{dC_s}{dz} = 0 \quad C_s(zl) = C_l, \quad C_s(zl+zl) = C_{\infty} \quad 5.25$$

Equation 5.24 can be integrated to yield

$$T_s(z) = T_i - \frac{G_i \alpha}{V - w_s} \left[\exp \left(\frac{V - w_s}{\alpha} (zt - z) \right) - 1 \right] \quad 5.26$$

The boundary conditions of equation 5.25 require the use of a solute balance at the solid-liquid interface.³³ These relations are

$$(C_i - C_\infty) (V - w_s(zt)) = -D \left(\frac{dC_s}{dz} \right)_u, \quad \left(\frac{dC_s}{dz} \right)_u = \frac{G_i}{m} \quad 5.27$$

which give the following for the dendrite tip solutal boundary condition

$$C_i = C_\infty - \frac{DG_i}{(V - w_s)m} \quad 5.28$$

Using this relation, equation 5.25 can be integrated to yield

$$C_s(z) = C_\infty - \frac{G_i D}{(V - w_s)m} \exp \left(\frac{V - w_s}{D} (zt - z) \right) \quad 5.29$$

The base state solution is thus known for both the two-phase and fluid regions.

5.3 PERTURBATION EQUATIONS

The approach taken in a stability analysis is to identify the effects of perturbations on the base solution. Selected variables are written by superposing the base state solution and perturbation solution as follows

$$\begin{aligned}
u(x,z,t) &= u'(x,z,t) \\
w(x,z,t) &= w_s(x,z) + w'(x,z,t) \\
P(x,z,t) &= P_s(x,z) + P'(x,z,t) \\
T(x,z,t) &= T_s(x,z) + T'(x,z,t) \\
C(x,z,t) &= C_s(x,z) + C'(x,z,t) \\
\rho(x,z,t) &= \rho_s(x,z) + \rho'(x,z,t)
\end{aligned}
\tag{5.30}$$

These expressions are now substituted into equations 5.9-5.13. The perturbed quantities are assumed to be small enough that non-linear terms can be neglected. The base solution terms are then removed to yield the following equations governing the perturbation solutions for mass, x momentum, z momentum, energy, and solute, respectively

$$\frac{\partial u'}{\partial x} + \frac{\partial w'}{\partial z} = 0 \tag{5.31}$$

$$\frac{\partial u'}{\partial t} = V \frac{\partial u'}{\partial z} - \frac{\Phi}{\rho_o} \frac{\partial P'}{\partial x} + \nu \nabla^2 u' - \frac{\nu \Phi_s}{\kappa_s} u' - F_s \Phi w_s u' \tag{5.32}$$

$$\begin{aligned}
\frac{\partial w'}{\partial t} &= V \frac{\partial w'}{\partial z} - \frac{\Phi}{\rho_o} \frac{\partial P'}{\partial z} + \nu \nabla^2 w' - \frac{\nu \Phi_s}{\kappa_s} w' \\
&- \Phi \frac{\rho'}{\rho_o} g - F_s \Phi w_s w'
\end{aligned}
\tag{5.33}$$

$$\frac{\partial T'}{\partial t} + (w_s - V) \frac{\partial T'}{\partial z} + w' \frac{dT_s}{dz} = \alpha \nabla^2 T' \tag{5.34}$$

$$\Phi \frac{\partial C'}{\partial t} + (w_s - V\Phi_s) \frac{\partial C'}{\partial z} + w' \frac{dC_s}{dz} - D\Phi_s \nabla^2 C' \quad 5.35$$

$$+ D \frac{d\Phi_s}{dz} \frac{\partial C'}{\partial z} + V \left(\frac{1-k}{1-\gamma} \right) \frac{d\Phi_s}{dz} C'$$

By taking the curl of equations 5.32 and 5.33 and using 5.31, the pressure term in equations 5.32 and 5.33 can be eliminated. Assumptions concerning the form of the perturbation solutions are also made. Fourier type representations are used to account for the full range of perturbations, i.e. instead of examining random perturbation functions, individual Fourier components are examined individually. If one component is found to be unstable, then the system is unstable. The perturbations are represented by Fourier type expansions as

$$f'(x,z,t) = F(z) \exp[\sigma t + iax] \quad 5.36$$

where σ is the temporal growth term and a is the wavenumber of the individual Fourier components. It is the sign of σ that determines whether the perturbations are growing or decaying with time. This form is substituted for u' , w' , T' , and C' in equations 5.32-5.35. The perturbation density is written in terms of perturbed temperature and concentration using the relation

$$\rho' = -\rho_o (B_T T' + B_C C') \quad 5.37$$

The final perturbation equations can then be written as a set of ordinary differential equations in the z domain for W , T , and C

$$\begin{aligned}
& \frac{d^4 W}{dz^4} + \left(\frac{V}{v} - \frac{1}{\Phi_s} \frac{d\Phi_s}{dz} \right) \frac{d^3 W}{dz^3} \\
& + \left(\frac{F_s \Phi_s}{v} w_s - \frac{V}{v \Phi_s} \frac{d\Phi_s}{dz} - 2a^2 - \frac{\Phi_s}{\kappa_s} \right) \frac{d^2 W}{dz^2} \\
& + \left(\frac{1}{v \Phi_s} \frac{dF_s w_s \Phi_s}{dz} + \frac{F_s w_s}{v} \frac{d\Phi_s}{dz} + \frac{a^2}{\Phi_s} \frac{d\Phi_s}{dz} \right) \\
& + \left(\frac{1}{\kappa_s} \frac{d\Phi_s}{dz} - \frac{Va^2}{v} - \frac{d\frac{\Phi_s}{\kappa_s}}{dz} \right) \frac{dW}{dz} \\
& + \left(a^4 + \frac{F_s \Phi_s w_s a^2}{v} + \frac{\Phi_s a^2}{\kappa_s} \right) W - \frac{g \Phi_s a^2}{v} (B_c C + B_T T) \\
& - \sigma \left(\frac{-1}{v} \frac{d^2 W}{dz^2} + \frac{1}{v \Phi_s} \frac{dW}{dz} - \frac{a^2}{v} W \right)
\end{aligned} \tag{5.38}$$

$$\frac{d^2 T}{dz^2} + \left(\frac{w_s - V}{\alpha} \right) \frac{dT}{dz} - a^2 T - \frac{1}{\alpha} \frac{dT_s}{dz} W = \sigma T \tag{5.39}$$

$$\begin{aligned}
& \frac{d^2 C}{dz^2} + \left(\frac{\frac{w_s - V}{\Phi_s}}{D} + \frac{1}{\Phi_s} \frac{d\Phi_s}{dz} \right) \frac{dC}{dz} + \left(\frac{(1-k)}{(1-\gamma)} \frac{V}{D \Phi_s} \frac{d\Phi_s}{dz} - a^2 \right) C \\
& - \frac{1}{D \Phi_s} \frac{dC_s}{dz} W = \sigma C
\end{aligned} \tag{5.40}$$

This equation set has retained the dependence on the wavenumber, a , and the temporal growth parameter σ . The momentum equation is similar but not identical to that given

by Nandapurkar et al.³⁸ However, it can be shown that the Nandapurkar et al equation is dimensionally incorrect. This point is discussed further for the Pb-Sn test case.

5.4 SOLUTION NUMERICS

Equations 5.38 - 5.40 represent a full eigenvalue problem which can be solved by finite difference approximation. A non-uniform grid was used to resolve the spatial dependence of the coefficients of these equations. Grid points were clustered around the solid-liquid interface ($z=z_t$). The permeability in this region has a very high spatial dependence, and small grid spacings are needed to resolve this dependence. The z derivatives are discretized over a non-uniform grid using uniform stencils in computational space in conjunction with grid metric terms resulting from chain rule substitutions. The problem is thus transformed from the physical domain z to the computational domain η . The chain rule substitutions for the z derivatives, using velocity as an example, are

$$\frac{dW}{dZ} = \frac{dW}{d\eta} \frac{d\eta}{dz} \quad 5.41$$

$$\frac{d^2W}{dZ^2} = \frac{dW}{d\eta} \frac{d^2\eta}{dZ^2} + \frac{d^2W}{d\eta^2} \left(\frac{d\eta}{dZ} \right)^2 \quad 5.42$$

$$\begin{aligned} \frac{d^3W}{dZ^3} = & \frac{dW}{d\eta} \frac{d^3W}{dZ^3} + \frac{d^2W}{d\eta^2} \frac{d\eta}{dZ} \frac{d^2\eta}{dZ^2} \\ & + 2 \frac{d^2W}{d\eta^2} \frac{d\eta}{dZ} \frac{d^2\eta}{dZ^2} + \frac{d^3W}{d\eta^3} \left(\frac{d\eta}{dZ} \right)^3 \end{aligned} \quad 5.43$$

$$\begin{aligned} \frac{d^4W}{dZ^4} = & \frac{dW}{d\eta} \frac{d^4\eta}{dZ^4} + 4 \frac{d^2W}{d\eta^2} \frac{d\eta}{dZ} \frac{d^3\eta}{dZ^3} \\ & + 3 \frac{d^2W}{d\eta^2} \left(\frac{d^2\eta}{dZ^2} \right)^2 + 6 \frac{d^3W}{d\eta^3} \left(\frac{d\eta}{dZ} \right)^2 \frac{d^2\eta}{dZ^2} \\ & + \frac{d^4W}{d\eta^4} \left(\frac{d\eta}{dZ} \right)^4 \end{aligned} \quad 5.44$$

The transformation function was taken from Anderson et al.⁸⁴ It clusters points around a given location, and, in this case, the grid is made fine in the vicinity of the solid-liquid interface and becomes increasingly coarse for $z=0$ and $z=zl$. The term s determines the degree of clustering or stretching. The transformation is given as

$$\begin{aligned} \eta = & B + \frac{1}{s} \sinh^{-1} \left[\left(\frac{z}{zt} - 1 \right) \sinh(sB) \right] \\ B = & \frac{1}{2s} \ln \left[\frac{1 + (e^s - 1) \frac{zt}{zt + zl}}{1 + (e^{-s} - 1) \frac{zt}{zt + zl}} \right] \end{aligned} \quad 5.45$$

and the corresponding metric derivatives are

$$\frac{d\eta}{dz} = \frac{\sinh(sB)}{s \, zt} \left[\Omega \right]^{\frac{1}{2}} \quad 5.46$$

$$\frac{d^2\eta}{dz^2} = \frac{-\sinh^3(sB)}{s z t} \left(\frac{z}{zt} - 1 \right) [\Omega]^{-\frac{3}{2}} \quad 5.47$$

$$\begin{aligned} \frac{d^3\eta}{dz^3} = & \frac{3 \sinh^5(sB)}{s z t^3} \left(\frac{z}{zt} - 1 \right)^2 [\Omega]^{-\frac{5}{2}} \\ & - \frac{\sinh^3(sB)}{s z t^3} [\Omega]^{-\frac{3}{2}} \end{aligned} \quad 5.48$$

$$\begin{aligned} \frac{d^4\eta}{dz^4} = & - \frac{15 \sinh^7(sB)}{s z t^4} \left(\frac{z}{zt} - 1 \right)^3 [\Omega]^{-\frac{7}{2}} \\ & + \frac{9 \sinh^5(sB)}{s z t^4} [\Omega]^{-\frac{5}{2}} \end{aligned} \quad 5.49$$

where

$$\Omega = 1 + \left(\frac{z}{zt} - 1 \right)^2 \sinh^2(sB) \quad 5.50$$

Equations 5.41 - 5.50 are used with a uniform differencing scheme for derivatives with respect to η to represent the z derivatives. The eigensystem is then written in matrix form as

$$A \bar{x} = \sigma B \bar{x} \quad 5.51$$

where A and B are square matrices and $\bar{x}$ is the solution W , T , and C .

Since the real part of the temporal growth parameter σ dictates if the perturbations are stable, it is set to zero to determine the neutral stability case. The

imaginary part of σ is likewise set to zero, limiting the analysis to non-oscillatory convection. This is known as satisfying the exchange of stabilities.⁶⁴ Experiments have shown that stationary convection is the type of breakdown for this problem.³⁰ The perturbations are neither growing nor decaying with time but are marginally stable. The solution for this condition is said to be at the onset of instability. Equation 5.50 can then be written as

$$A \bar{x} = 0 \quad 5.52$$

and a solution exists if the determinant of the matrix A is zero.

The determinant of the matrix A was calculated using the IMSL⁸⁵ routine LUDATF, which performs an L-U decomposition on A and returns the determinant. The IMSL routine DVERK⁸⁵ was used to solve equations 5.15, 5.21 and 5.22, the base state in the two-phase region. An IBM RS6000 Model 530 was used to execute the programs which were written in FORTRAN. The time required per iteration varied from approximately .5 CPU seconds for order A of 150 to approximately 10 CPU seconds for order A of 370.

5.5 RESULTS

To verify the numerical approach and its application, several test cases were solved before using the approach for the $\text{NH}_4\text{Cl-H}_2\text{O}$ system. The cases were performed in an ascending order of complexity, and the results are given here to demonstrate the effectiveness of the approach.

5.5.1 VALIDATION CASE 1: THERMAL CONVECTION

The problem of an enclosed fluid heated from below is a convective stability problem that is well known.^{63,69} The fluid is initially at rest with a linear temperature gradient, G_l , in effect across the top and bottom rigid walls. As the temperature gradient is increased, the potential for flow also increases as the density inversion steepens. There are no solutal effects in this problem and the governing equations simplify from 5.38 - 5.40 to

$$\frac{d^4 W}{dz^4} - 2a^2 \frac{d^2 W}{dz^2} + a^4 W - \frac{gB_r}{\nu} a^2 T = 0 \quad 5.53$$

$$\frac{d^2 T}{dz^2} - a^2 T + \frac{G_l}{\alpha} W = 0 \quad 5.54$$

with the boundary conditions

$$W = \frac{dW}{dz} = T = 0 \quad \text{at } z = 0 \quad \text{and } z = 1 \quad 5.55$$

The solution was obtained using both a uniform grid and the stretched grid with various degrees of stretching. The domain was defined as $z_t = z_l = 0.5$ and was divided into 101 points. Property values for Pb - 20 wt% Sn from Nandapurkar were chosen for the calculations.³⁸ These values are given in Table 5.1. This problem served as a fundamental test of the accuracy of the matrix coefficients including the metric terms for the stretched grid. It was originally formulated by Rayleigh⁶⁹ and the results for the neutral stability of this problem are given in graphical form by Chandrasekhar.⁶³ The theoretical critical Rayleigh number for this case is

Table 5.1 Pb-20wt% Sn Property Values.³⁸

Property	Value
ν Viscosity cm^2/sec	2.73×10^{-3}
D Solutal Diffusivity cm^2/sec	3.00×10^{-5}
α Thermal Diffusivity cm^2/sec	1.73×10^{-2}
C_p Specific Heat $\text{J}/(\text{g } ^\circ\text{C})$	0.08
L_f Latent Heat J/g	30.92
k Partition Coefficient	0.453
m Liquidus Slope $^\circ\text{C}/\text{wt}\% \text{H}_2\text{O}$	-2.33
ρ_o Reference Density g/cm^3	9.672
B_T Thermal Expansion Coef $1/^\circ\text{C}$	1.07×10^{-4}
B_C Solutal Expansion Coef $1/\text{wt}\% \text{H}_2\text{O}$	5.07×10^{-3}
γ Solidification Contraction Coefficient	0.0

$$Ra_c = \frac{gB_r GL^4}{\nu \alpha} = 1707.76 \quad 5.56$$

The problem was solved using the linear stability model (LSM) for various values of wavenumber and Gl . A comparison of the LSM and theoretical results are shown in Figure 5.2, indicating excellent agreement for both grids. The slight difference between $s=5$ and $s=1$ is due to the decreased accuracy in implementing the boundary conditions with the higher boundary grid spacings for the stretched grid. Table 5.2 gives the maximum and minimum mesh spacings for the grids used.

5.5.2 VALIDATION CASE 2: THERMOSOLUTAL CONVECTION

In the second validation case, a fluid layer is enclosed between two horizontal free surfaces and is subjected to both thermal and solutal linear gradients.⁸⁶ As in the thermal only case, the fluid is initially at rest. There are no two-phase effects for this problem and the governing equations are

$$\begin{aligned} \frac{d^4 W}{dz^4} - 2a^2 \frac{d^2 W}{dz^2} + a^4 W \\ - \frac{gB_r G_l T}{\alpha \nu \pi^4} - \frac{gB_c S'}{D\nu \pi^4} C = 0 \end{aligned} \quad 5.57$$

$$\frac{d^2 T}{dz^2} - a^2 T + W = 0 \quad 5.58$$

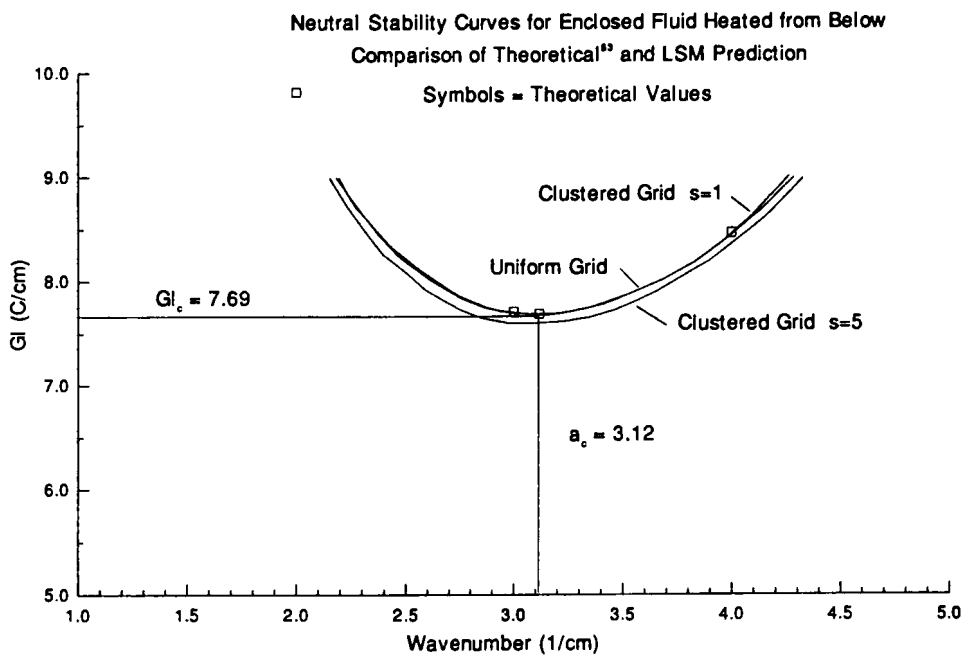


Figure 5.2 Thermal Stability Validation Case.

Table 5.2 Grid Spacing for Thermal Convection Validation Case.

Grid Type	$\Delta Z_{\min}$ (cm)	$\Delta Z_{\max}$ (cm)
Uniform Spacing	0.0100	0.0100
Stretched (s=1)	0.0096	0.0108
Stretched (s=5)	0.0041	0.0247

$$\frac{d^2C}{dz^2} - a^2C + W = 0 \quad 5.59$$

Where S' is the solute gradient and π appears because the equations have been non-dimensionalized. The appropriate boundary conditions are

$$W - \frac{d^2W}{dz^2} - T - C = 0 \quad \text{at } z = 0 \quad \text{and } z = \pi \quad 5.60$$

The solute gradient was arbitrarily set such that the solutal Rayleigh number

$$S = \frac{g_B c S'}{D \nu \pi^4} \quad 5.61$$

was equal to three. Selecting a value for either the solutal or thermal Rayleigh number was necessary because the solutal and thermal profiles are independent. Iterations in this case were performed on the thermal Rayleigh number

$$R = \frac{g_B \tau G_i}{\alpha \nu \pi^4} \quad 5.62$$

by changing the temperature gradient. Property values for Pb - 20 wt% Sn were again used. The solution was obtained using a uniform grid with the domain defined as $z_l = z_u = \pi/2$. The combined theoretical critical Rayleigh number, $R+S$, for this problem is 6.75 at a wavenumber of .707.⁸⁶ Figure 5.3 shows a comparison of LSM predictions and theoretical values. Once again the agreement is very good.

The ability to match these fundamental theoretical cases provided the necessary validation for the LSM. Unfortunately, further comparisons with predictions for cases including solidification and two-phase effects were hampered by a lack of such cases

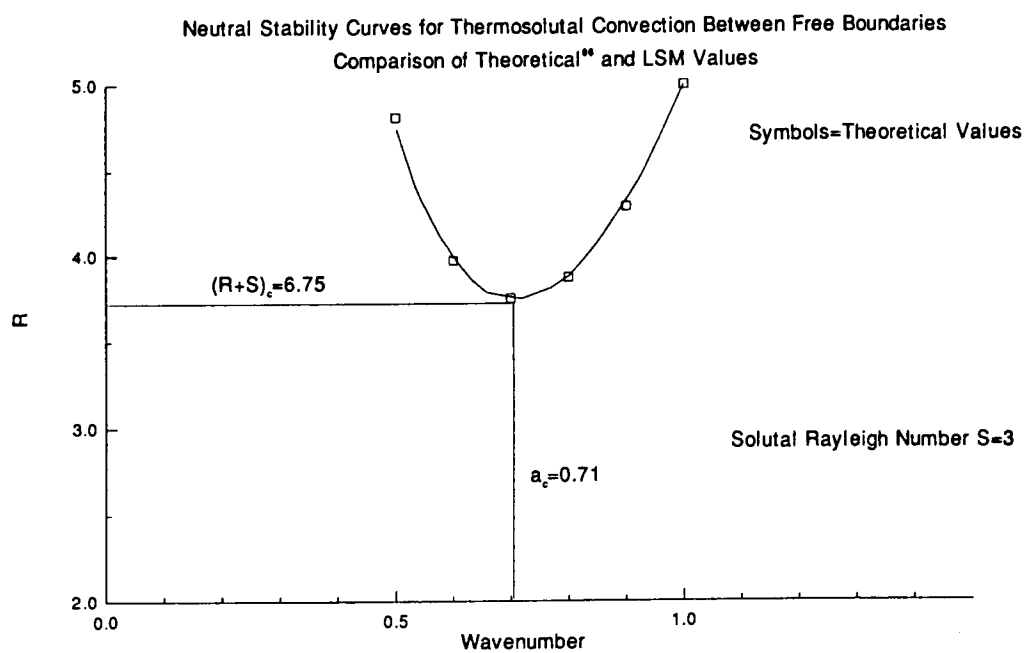


Figure 5.3 Thermosolutal Stability Validation Case.

in the literature and errors identified in those that have been published. One such case was chosen for investigation using LSM, however, and is discussed in the following section.

5.5.3 COMPARISON CASE: SOLIDIFICATION OF Pb-Sn

This case is that given by Nandapurkar et al.³⁸ The equations and domain are 5.38-5.40 without contraction flow and Forchheimer terms ($\gamma=0$, $F_s=0$). The equations are thus

$$\begin{aligned}
 & \frac{d^4 W}{dz^4} + \left(\frac{V}{v} - \frac{1}{\Phi_s} \frac{d\Phi_s}{dz} \right) \frac{d^3 W}{dz^3} \\
 & + \left(\frac{-V}{v \Phi_s} \frac{d\Phi_s}{dz} - 2a^2 - \frac{\Phi_s}{\kappa_s} \right) \frac{d^2 W}{dz^2} \\
 & + \left(\frac{a^2}{\Phi_s} \frac{d\Phi_s}{dz} + \frac{1}{\kappa_s} \frac{d\Phi_s}{dz} - \frac{Va^2}{v} - \frac{d\Phi_s}{\kappa_s dz} \right) \frac{dW}{dz} \\
 & + \left(a^4 + \frac{\Phi_s a^2}{\kappa_s} \right) W - \frac{g\Phi_s a^2}{v} (B_c C + B_T T) \\
 & = \sigma \left(\frac{-1}{v} \frac{d^2 W}{dz^2} + \frac{1}{v \Phi_s} \frac{dW}{dz} - \frac{a^2}{v} W \right)
 \end{aligned} \tag{5.63}$$

$$\frac{d^2 T}{dz^2} + \left(\frac{-V}{\alpha} \right) \frac{dT}{dz} - a^2 T - \frac{1}{\alpha} \frac{dT_s}{dz} W = \sigma T \tag{5.64}$$

$$\frac{d^2C}{dz^2} + \left(\frac{-V}{D} + \frac{1}{\Phi} \frac{d\Phi}{dz} \right) \frac{dC}{dz} + \left(\frac{(1-k)V}{D\Phi} \frac{d\Phi}{dz} - a^2 \right) C - \frac{1}{D\Phi} \frac{dC}{dz} W = \sigma C \quad 5.65$$

The boundary conditions are

$$W = \frac{d^2W}{dz^2} = T = C = 0 \text{ at } z = 0 \text{ and } z = z_t + z_l \quad 5.66$$

A modified Kozeny-Carmen permeability model with empirically based modifications for the Pb-Sn system was used. This model assumes the flow is parallel to the primary dendrites and that the primary arm spacing is given by³⁸

$$d_1 = 556 G_l^{-.311} \quad 5.67$$

The permeability is then written as⁵

$$K = \frac{4.70 \times 10^{-4} d_1^2 \Phi^3}{1 - \Phi} \quad 5.68$$

The solution steps for this problem are given in Figure 5.4. The program inputs and physical properties are entered and for each temperature gradient, G_l , the base state is obtained for a fixed grid using equations 5.21-5.22, 5.26, and 5.29. The $z=0$ boundary conditions are for a eutectic solid, therefore z_t must also be determined in the base state solution, since it is a function of temperature gradient. z_l was chosen as $20D/V$ as in Nandapurkar. With z_t and z_l known, a stretched grid was calculated.

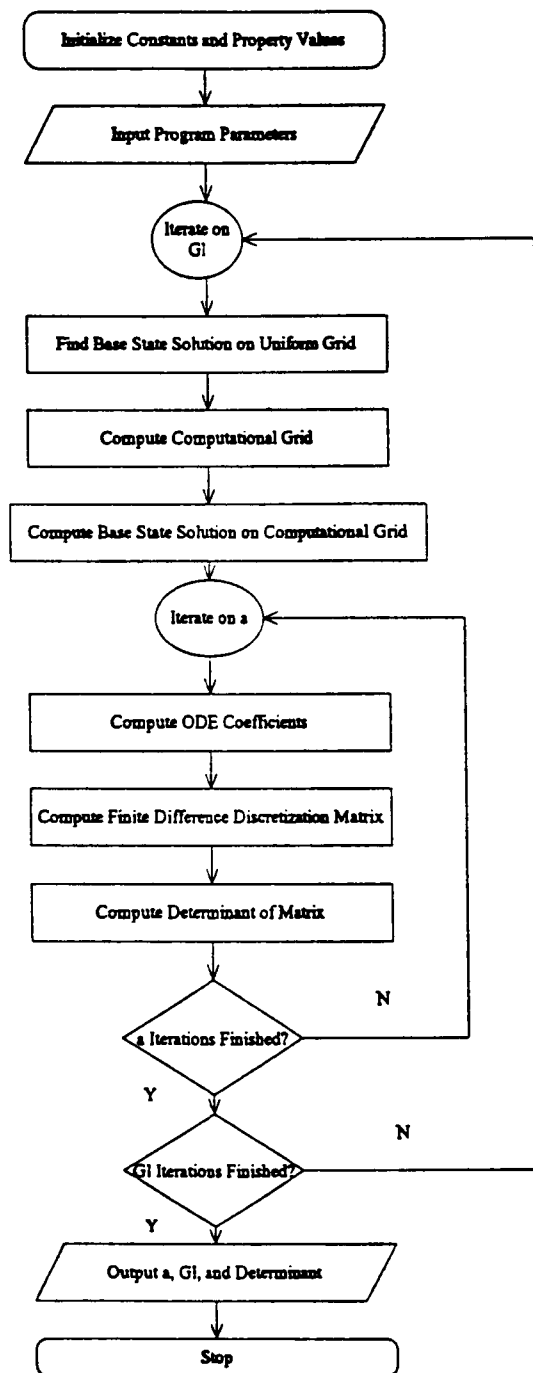


Figure 5.4 Program Steps for Pb-Sn Comparison Case.

Because D/V is small and z_t is as large as 20 cm, clustering around the interface is necessary to capture the influence of the mushy zone.

Figure 5.5 shows the base state liquid fraction values predicted by LSM. The values for z_t reported in Nandapurkar et al for given temperature gradients were duplicated by LSM predictions.³⁸ The ODE coefficients were calculated for given values of wavenumber, a , and the finite difference discretization matrix A was filled. Figures 5.6 and 5.7 show a comparison between LSM predictions with a stretched grid and $s=3$ and reported values for $g = 1$, and $.5 g_e$, respectively. Figure 5.8 shows the distribution of points for $Gr=10$ and 60 °C/cm. The results compare favorably at $1 g_e$, but show significant differences at $0.5 g_e$. The discrepancy is present in part because the momentum equation in Nandapurkar is incorrect (the third term in his equation 59 is dimensionally incorrect). Although the numerically dominant term in his equation 59,

$$\frac{\Phi, a^2}{K} \quad 5.69$$

is identical to the term in equation 5.63 in this study, the differences in the other terms contribute to the departure of solutions on a more detailed level. The implementation of the base state solution is also an area of possible discrepancy. In this study, first order at the boundary and second order otherwise finite differences were used to approximate derivatives of base state coefficients for which a closed form solution was not known. The method used by Nandapurkar is given.

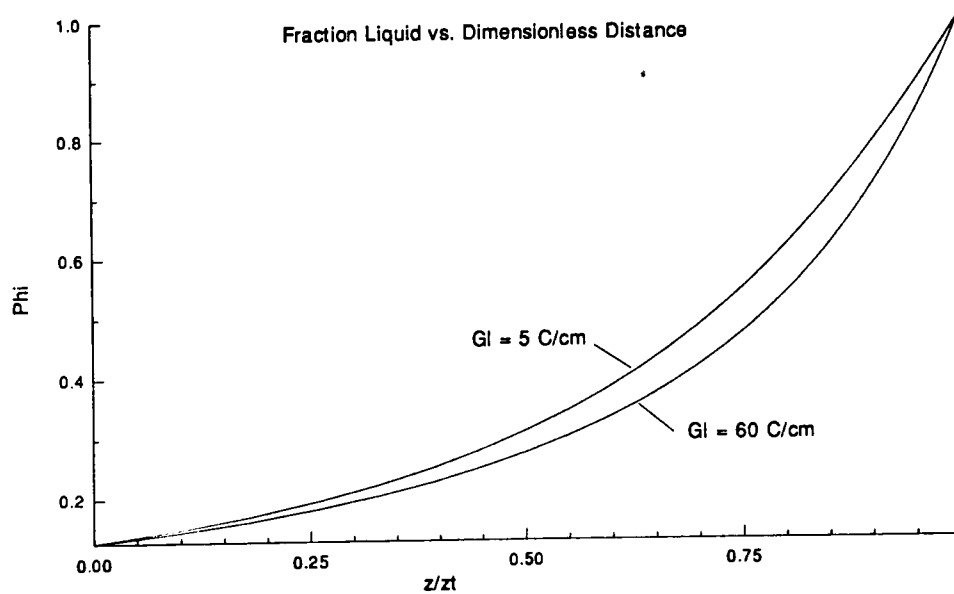


Figure 5.5 Fraction of Liquid Profiles for Pb-Sn Comparison Case.

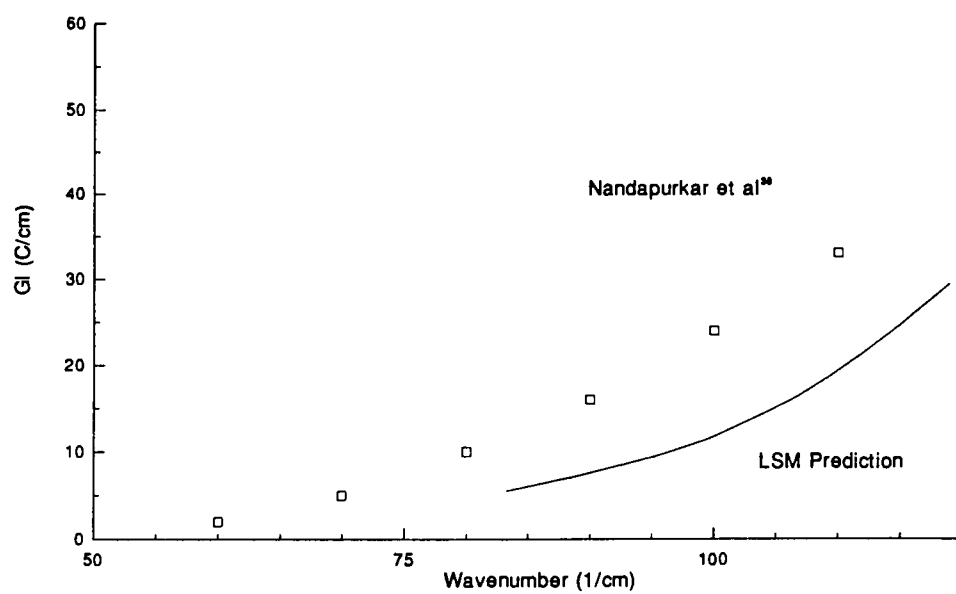


Figure 5.6 Pb-Sn Comparison Case $g=1g_e$.

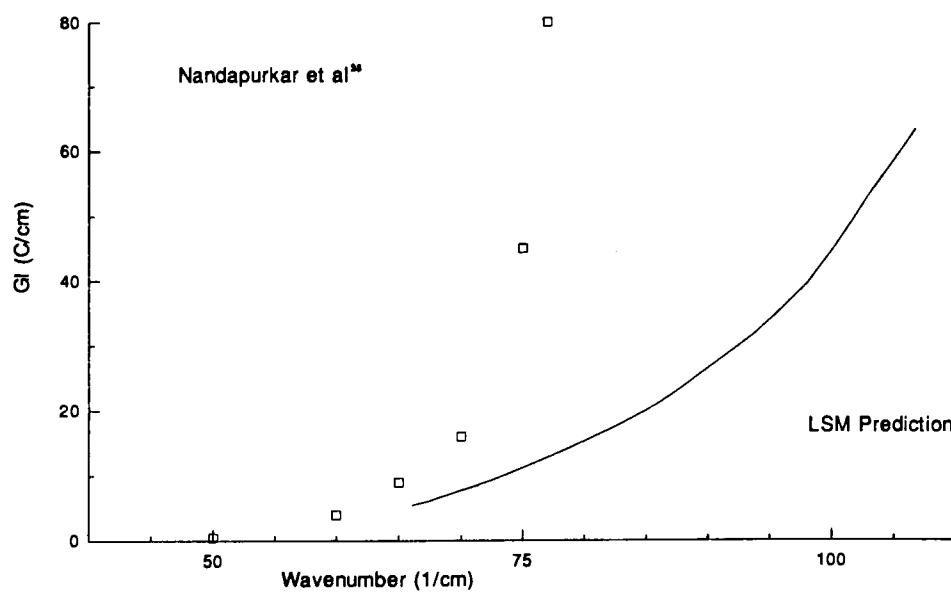


Figure 5.7 Pb-Sn Comparison Case $g=0.5g_c$.

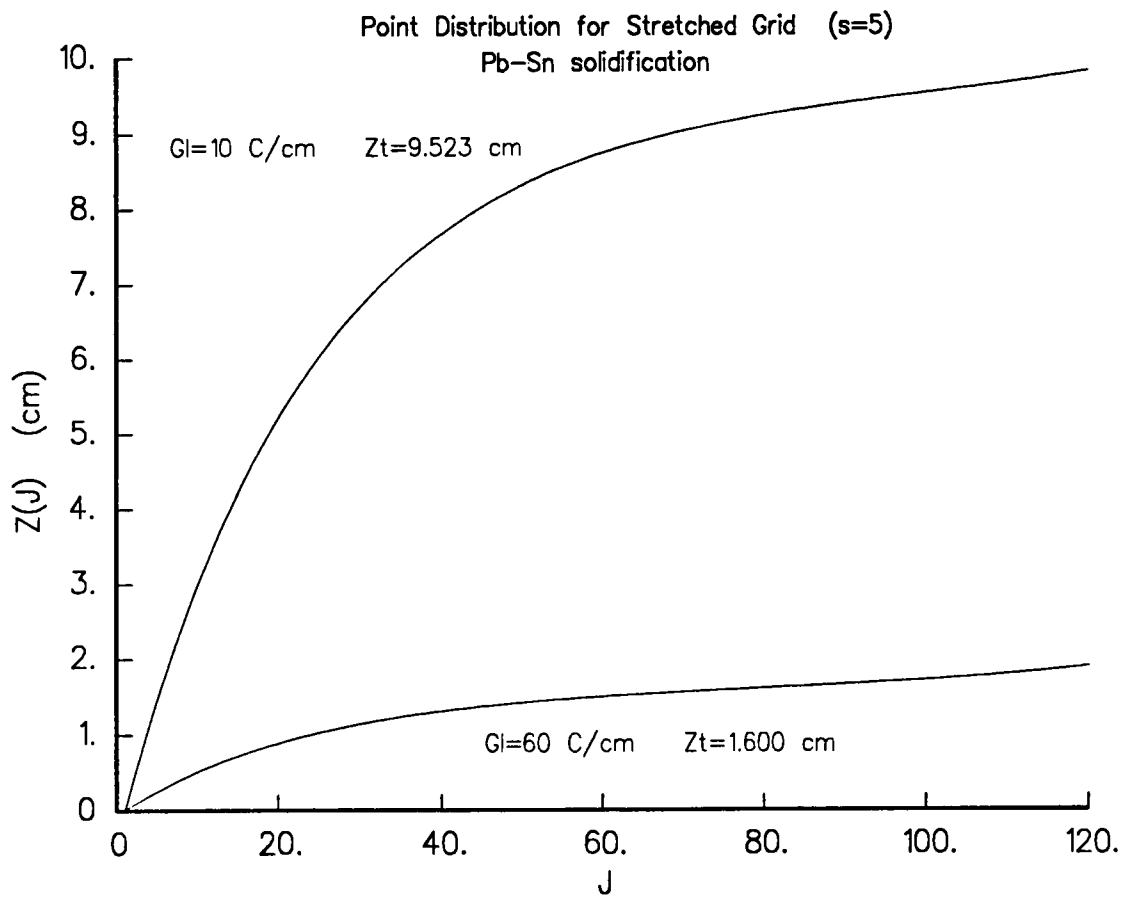


Figure 5.8 Grid Point Distribution for Pb-Sn Comparison Case.

5.5.4 SOLIDIFICATION OF $\text{NH}_4\text{Cl-H}_2\text{O}$: GENERAL RESULTS

The VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ is governed by equations 5.38-5.40 without modification. The perturbation boundary conditions are identical to those used in the PB-Sn test case, equation 5.66, but the base state boundary conditions must be written for a non-eutectic temperature at $z=0$ to model the experiments. z_t and z_l were part of the base state solution in the Pb-Sn case, but are not known a priori in the $\text{NH}_4\text{Cl-H}_2\text{O}$ case. In the absence of the submodels needed to provide these boundary conditions (which are beyond the scope of this work), experimentally determined values were used. As before, the temperature, concentration, and fraction of liquid values in the two-phase region are part of the solution for a given mushy zone height, z_t . z_l is determined through input of the extent of the diffusion layer, dl . In Section 5.5.3, dl was determined by the steady state solutal diffusion length D/V . In this case, z_l was set to twice this value, i.e. $2D/V$. This allows a study of the effects of the mushy zone independent of other inputs and also more closely approximates the experiments. Figure 5.9 gives the solution steps for this problem. The steady state solution in the mushy zone requires iteration when a simple integration solution such as Runge-Kutta is used since equations 5.15, and 5.21-5.23 constitute a two point boundary value problem. The problem is not very sensitive to initial guesses for $w_s(z_t)$ and a secant method was used for updating iteration values. Table 5.3 gives the property values for $\text{NH}_4\text{Cl- 72wt\% H}_2\text{O}$.²²

To examine the general effects of various solidification conditions, a matrix of LSM calculations were made. Table 5.4 gives the matrix and results. The effects of

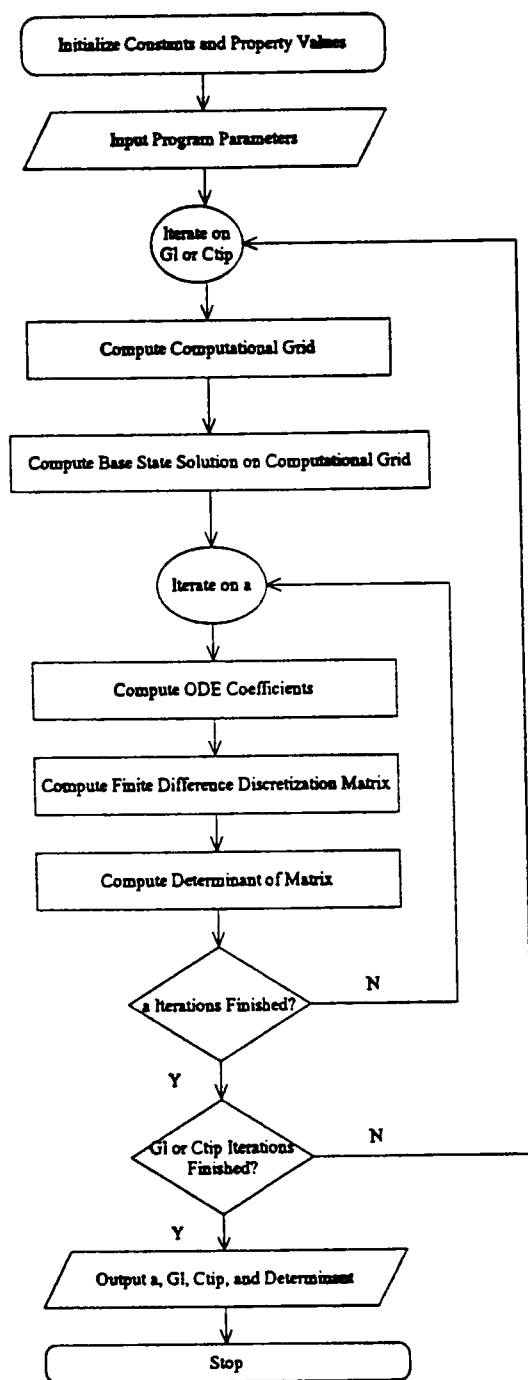


Figure 5.9 Program Steps for $\text{NH}_4\text{Cl-H}_2\text{O}$ Solution.

Table 5.3 NH_4Cl -72wt% H_2O Property Values.²²

Property	value
ν Viscosity cm^2/sec	9.82×10^{-3}
D Solutal Diffusivity cm^2/sec	2.30×10^{-5}
α Thermal Diffusivity cm^2/sec	1.50×10^{-3}
C_p Specific Heat $\text{J}/(\text{g } ^\circ\text{C})$	0.79
L_f Latent Heat J/g	66.04
k Partition Coefficient	0.004
m Liquidus Slope $^\circ\text{C}/\text{wt}\% \text{H}_2\text{O}$	-4.68
ρ_o Reference Density g/cm^3	1.067
B_T Thermal Expansion Coef $1/^\circ\text{C}$	2.95×10^{-4}
B_C Solutal Expansion Coef $1/\text{wt}\% \text{H}_2\text{O}$	2.81×10^{-3}
γ Solidification Contraction Coefficient	0.295

Table 5.4 NH₄Cl-H₂O General Results.

	V cm/sec	z cm	dl cm	rcon	g cm/sec ²	γ	K model	d cm	a _c 1/cm	Gl _c C/cm	Ra _c
dv1	2d-3	.05	.05	1d-8	1 g _o	.295	P	1d40	26	5.5	887
dv2	3d-3								28	9.5	1456
dv3	4d-3								30	15.5	2325
dv4	5d-3								34	29.5	4392
dz1	5d-3	.04	.06	1d-8	1 g _o	.295	P	1d40	36	62.5	7476
dz2		.045	.055						34	35.5	4763
dz3		.05	.05						34	29.5	4392
dz4		.06	.04						35	24.5	4206
dz5		.065	.035						37	27.5	5343
dz6		.067	.033						38	34.5	6917
dk1	5d-3	.05	.05	1d-8	1 g _o	.295	P	1d40	34	29.5	4392
dk2				.90					35	40.5	6031
dk3				.95					35	33.5	4839
dk4				10					30	14.5	2159
dk5				100					30	13.5	2010
dg1	5d-3	.05	.05	1d-8	1 g _o	.295	P	1d40	34	29.5	4392
dg2					.9				34	38.5	5446
dg3					1.1				33	21.5	3549
dg4					1.5				31	11.5	2569
dc1	5d-3	.05	.05	1d-8	1 g _o	.295	P	1d40	34	29.5	4392
dc2						.250			35	33.5	4983
dc3						.210			36	42.5	6316
dc4						.190			38	62.5	9284
dkc	5d-3	.05	.05	1d-8	1 g _o	.295	P	1d40	34	29.5	4392
dkc							KC		29	13.5	2010
dkc							DT		31	23.5	3499
df1	5d-3	.05	.05	1d-8	1 g _o	.295	P	1d40	34	29.5	4392
df2								1	34	29.5	4392
df3								.01	34	29.5	4392
df4								.002	34	33.5	4839
df5								.0013	33	38.5	5733

growth rate, permeability, acceleration, shrinkage flow and inertia terms on the convective stability were investigated.

The effects of permeability were examined in three ways. In one, the permeability was directly modified by multiplication by a constant. This gives a direct increase or decrease in the momentum dissipation due to the two-phase region. In addition, the ratio of two-phase layer height to fluid layer height was also examined since it has an indirect influence on the momentum dissipation in the two-phase region. This has been shown to be significant in terms of effect on the determination of stability for static base cases with no solidification.⁷⁵ Finally, three different models for permeability were investigated. One of the models is from Poirier,¹⁷ as used in the Pb-Sn test case, and one from Drummond and Tahir.^{39,50} These two models and the widely used Kozeny-Carmen expression were compared.

A stretched grid ($s=3$) with 61 points was used for all of the test cases. To aid comparison, a base set of inputs was defined for the study matrix with the domain defined as $z_t=dl=0.05$ cm, the permeability model of Poirier, $1 g_e$ acceleration level was , contraction flow enabled, and no Forchheimer term. z_l was set to $2dl$ after a comparison between it, $z_l=5dl$ and $z_l=10dl$ showed no difference in results. This set of inputs constituted the base case from which each of the above parameters was independently modified.

Figures 5.10-5.16 show the neutral stability curves predicted for each of the cases investigated. Figures 5.17-5.22 show the extracted critical combined Rayleigh number plotted as a function of the appropriate parameter for each specific case. In

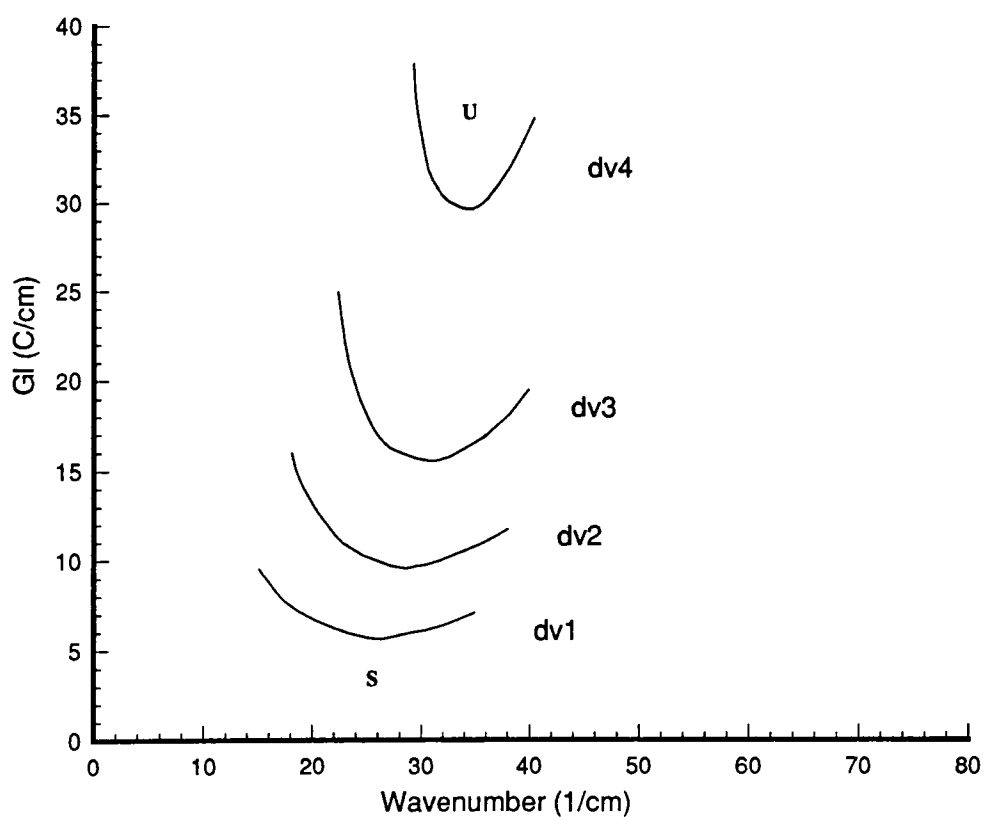


Figure 5.10 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of Growth Rate.

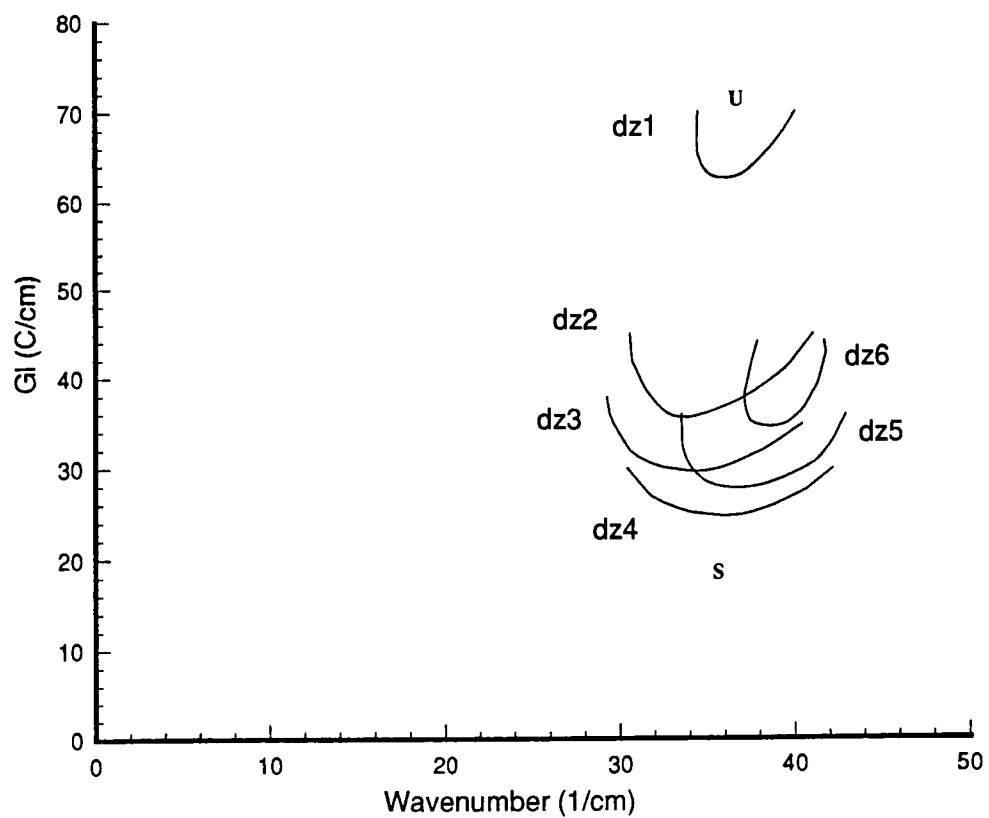


Figure 5.11 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of the Relative Height of the Two-Phase Region.

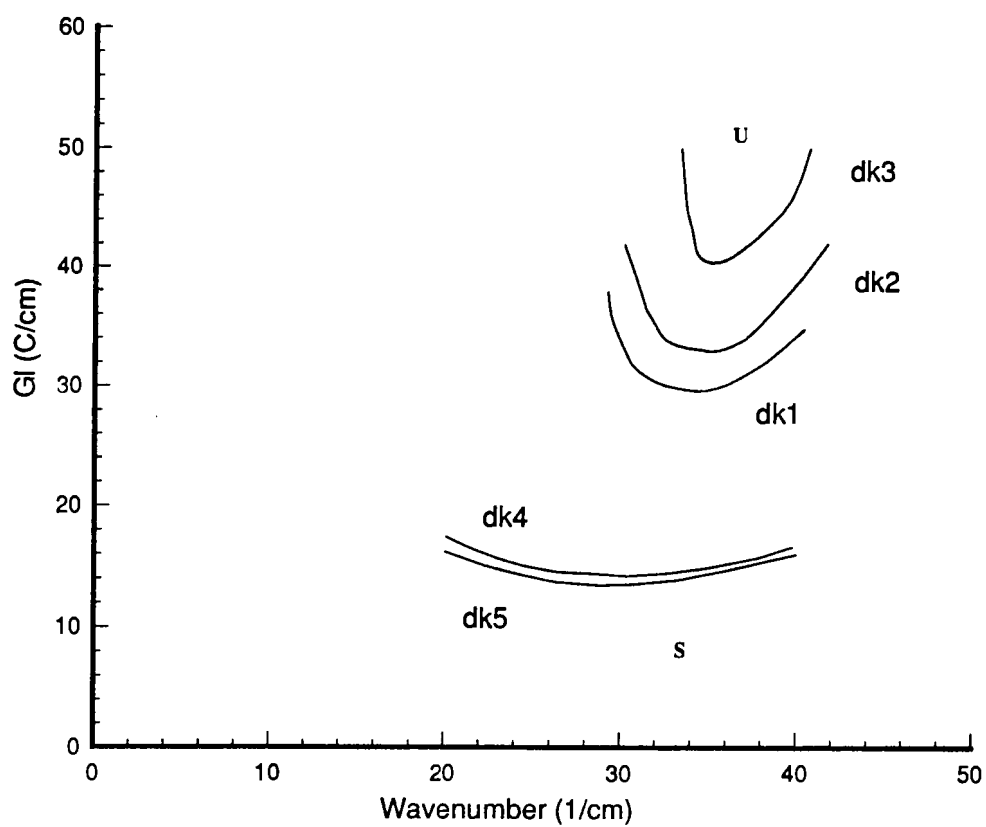


Figure 5.12 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of Permeability.

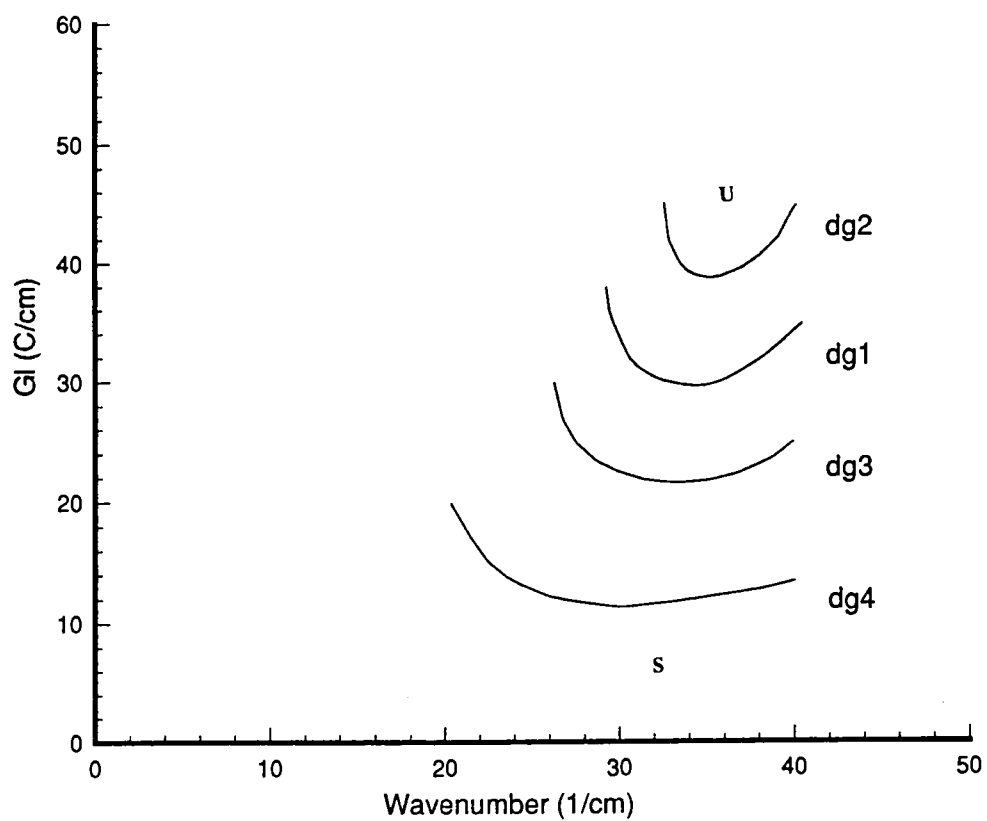


Figure 5.13 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of Gravity Level.

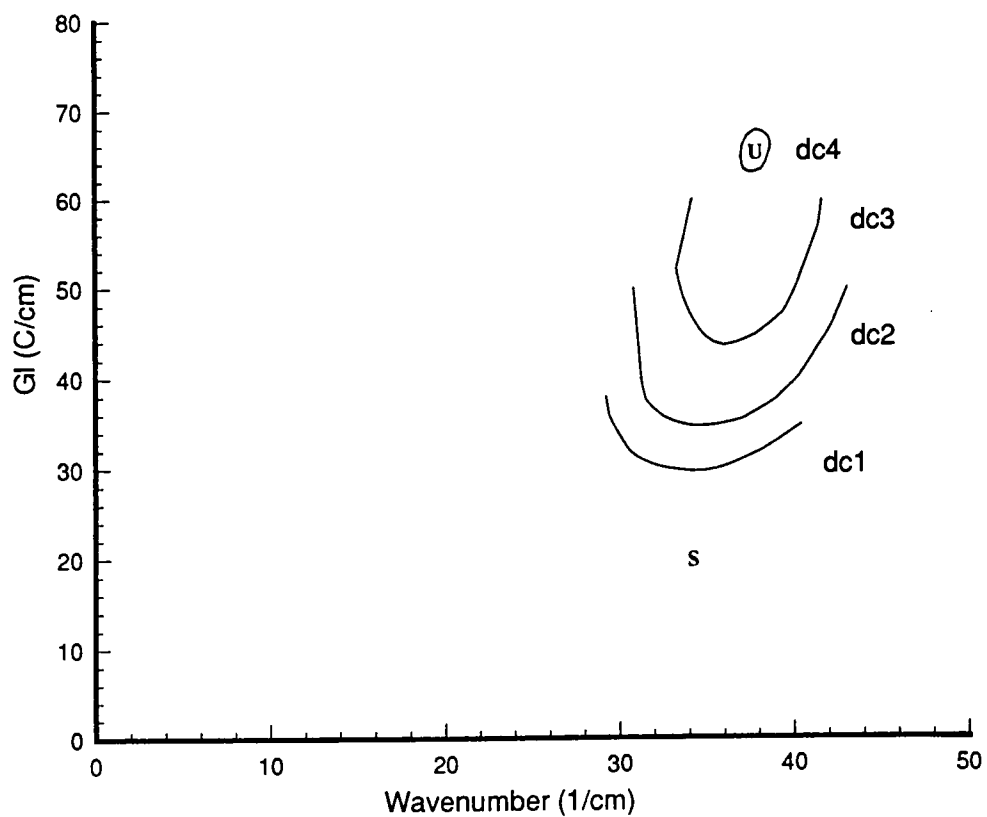


Figure 5.14 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of Contraction Flow.

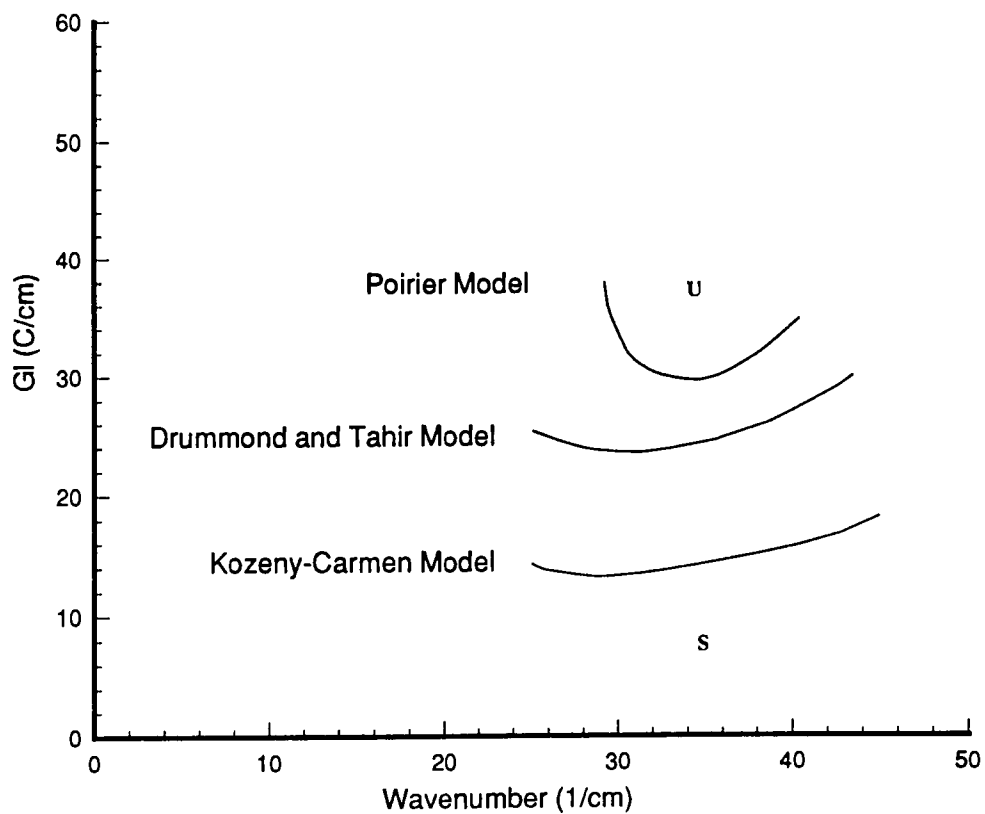


Figure 5.15 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of Permeability Model.

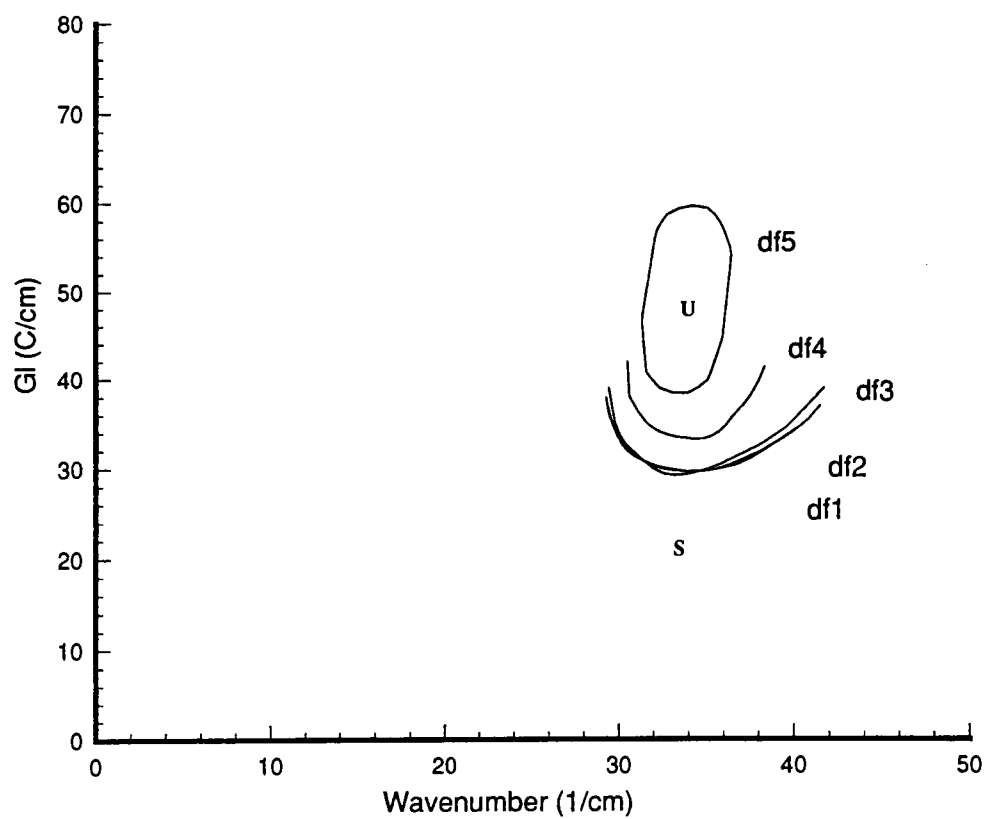


Figure 5.16 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Neutral Stability Curves as a Function of Forchheimer Inertia.

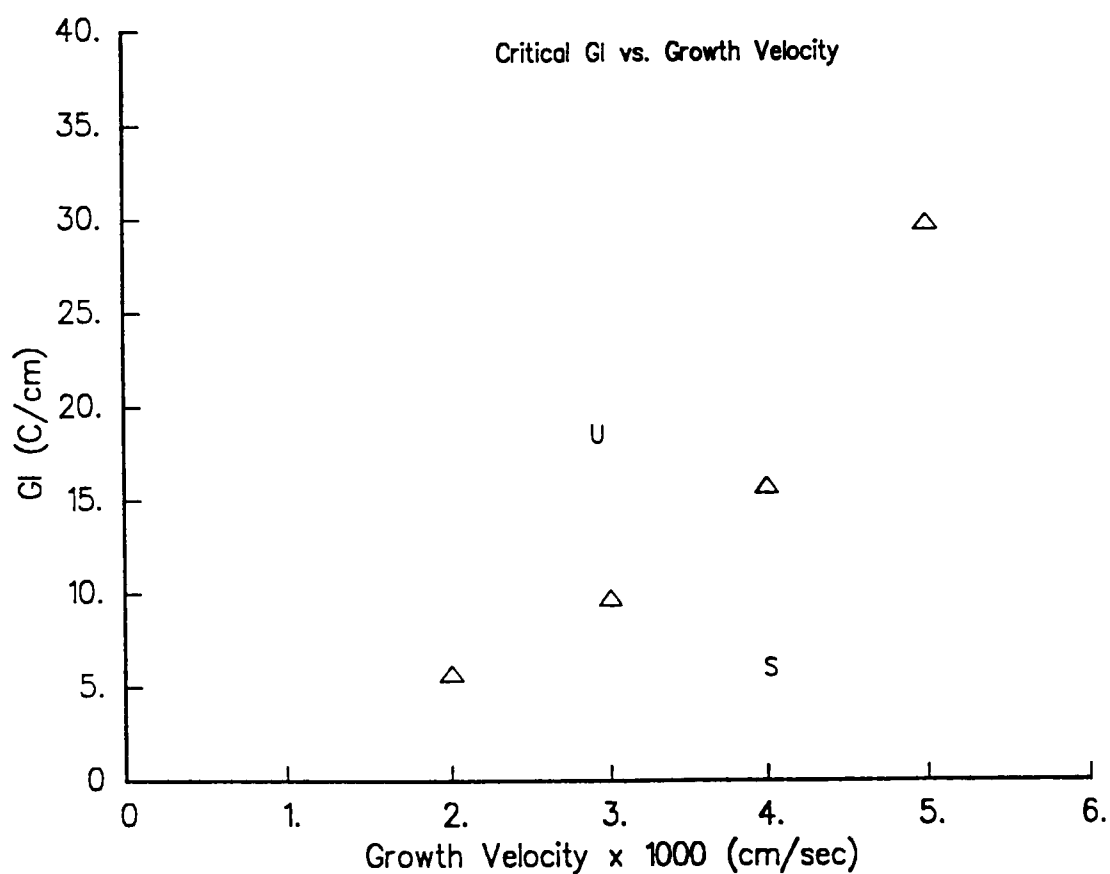


Figure 5.17 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Critical Temperature Gradient as a Function of Growth Rate.

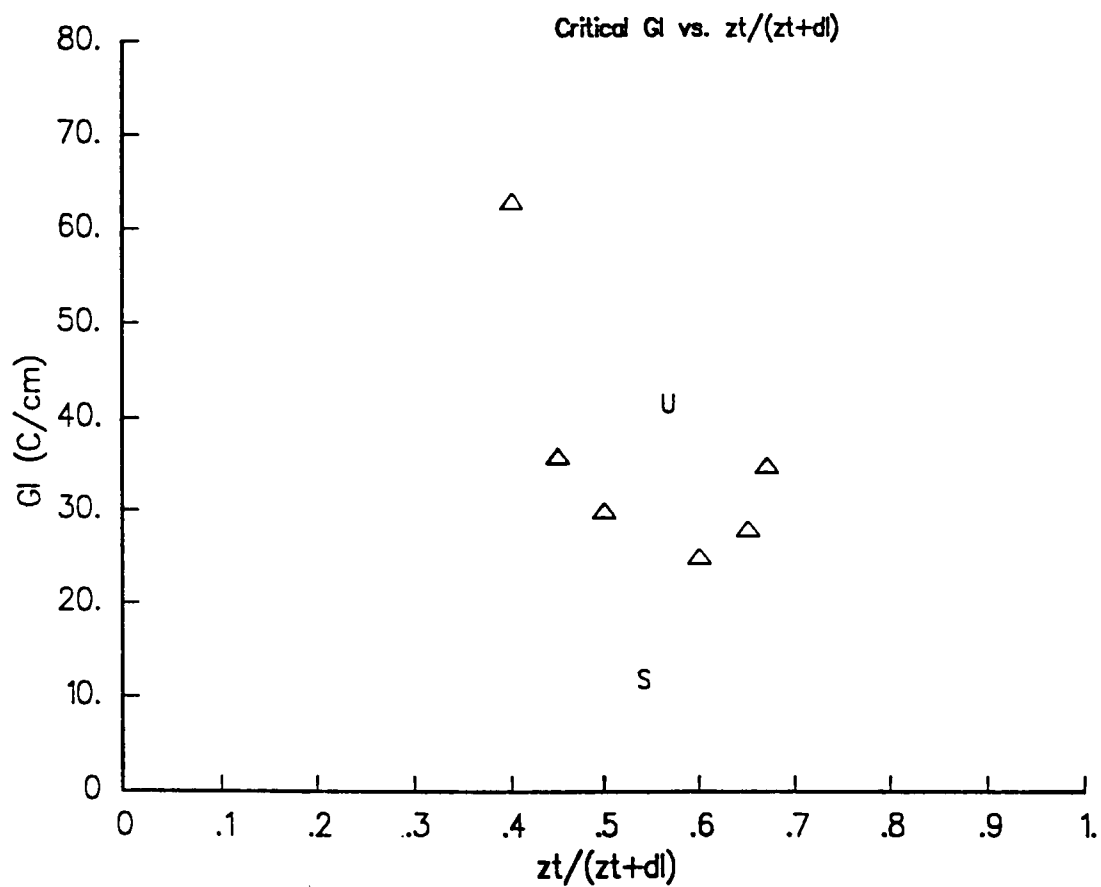


Figure 5.18 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Critical Temperature Gradient as a Function of the Relative Height of the Two-Phase Region.

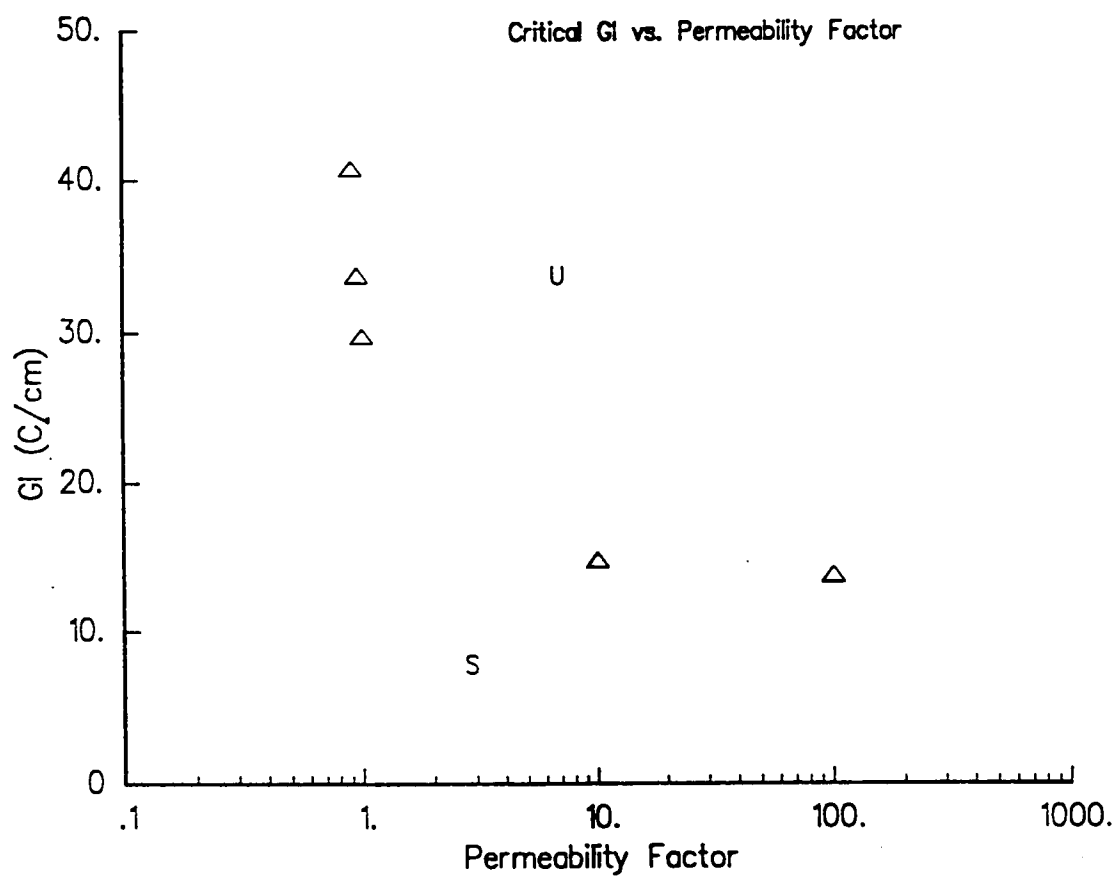


Figure 5.19 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Critical Temperature Gradient as a Function of Permeability.



Figure 5.20 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Critical Temperature Gradient as a Function of Gravity Level.

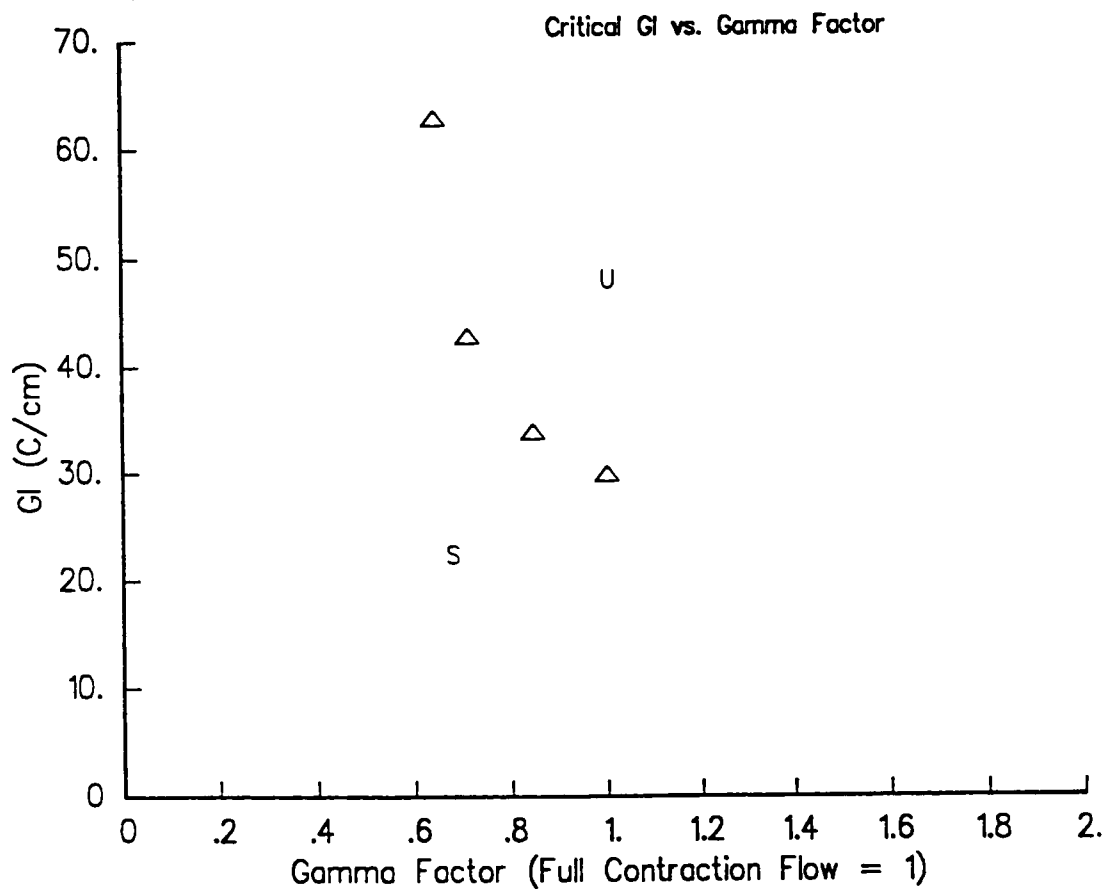


Figure 5.21 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Critical Temperature Gradient as a Function of Contraction Flow.

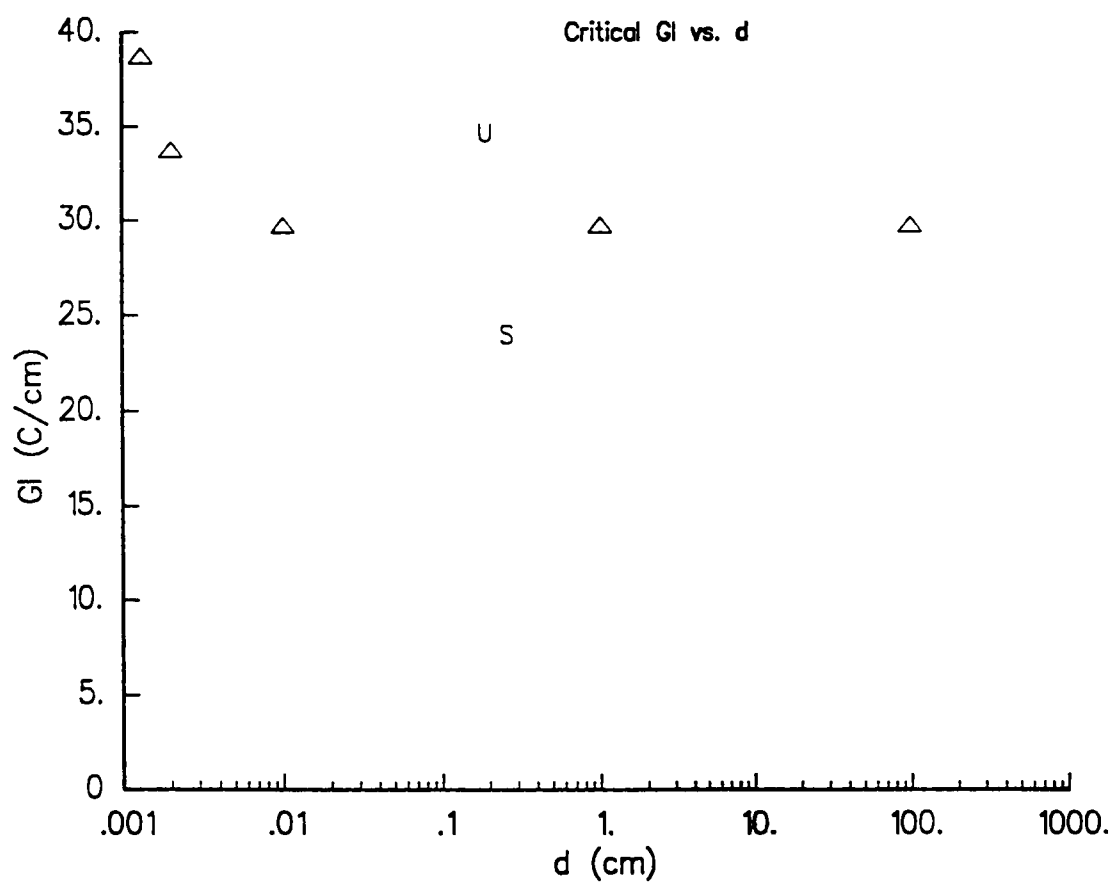


Figure 5.22 $\text{NH}_4\text{Cl-H}_2\text{O}$ General Comparison: Critical Temperature Gradient as a Function of Forchheimer Inertia.

these figures, the letter "U" denotes the region of conditions for an unstable system, and the letter "S" denotes the region of conditions for a stable system.

The effect of solidification velocity is seen in figures 5.10 and 5.17. The system becomes less susceptible to breakdown as the velocity is increased. Figures 5.11 and 5.18 show the influence on the critical combined Rayleigh number of two-phase region height z_t relative to total height, $z_t + d_l$. At small values of z_t , the concentration difference from $z=0$ to $z=z_t + d_l$ (which is primarily due to the concentration difference from $z=0$ to $z=z_t$) is not as great as it is for large values of z_t . Since the concentration difference provides the potential for flow, its value is very important in determining stability. Thus, the region of stable growth decreases as a function of z_t . However, the effects of added momentum dissipation due to the two-phase region become more important as z_t increases. This is evidenced in Figures 5.11 and 5.18 by the increase in stability for $z_t > 0.06$ cm.

Figures 5.12 and 5.19 show the influence of permeability on stability. The constant term in equation 5.67, 4.70×10^{-4} , was multiplied by a constant to investigate how the magnitude of this term affects stability. As the permeability is increased, the momentum dissipation in the two-phase region increases and the size of the region of stable growth also increases.

Figures 5.13 and 5.20 show the influence of acceleration (gravity level) on stability. Since the potential for flow is due to differences in density working in combination with an external acceleration, the acceleration level is expected to have

a significant influence on the size of the stable regions. The predictions show this trend, which is part of the appeal for low gravity materials processing.^{22,8-10}

To investigate the influence of contraction flows on the stability regions, the contraction coefficient, γ , was set to various values. Figures 5.14 and 5.21 show the results. The value of γ has a significant impact on the size of the stable regions. This is a significant result. Most stability models applied to solidifying systems to date have not included this term.

The permeability model chosen has a large effect on the size of the stable regions, as shown in Figure 5.15. The resulting stable region using the Kozeny-Carmen relation is substantially smaller than those resulting from either the Poirier or the Drummond and Tahir models.

The influence of the Forchheimer term was investigated by varying the characteristic length in equation 5.5. This term is only significant for small characteristic lengths, with d values greater than 100 μm showing little influence on stability. The results are shown in Figures 5.16 and 5.22. Notice that the stability curves are closed at low values of d . This represents a transition from a case having a critical temperature gradient at one wavenumber to a case that is either stable for all wavenumbers or has a different critical temperature gradient at a different wavenumber.

5.5.5 SOLIDIFICATION OF $\text{NH}_4\text{Cl-H}_2\text{O}$: EXPERIMENT MODELLING

In order to model the experiments, it was necessary to make several modifications to the LSM. The boundary conditions for the base state case were

modified to account for the fact that the experiments were not truly steady state. As in the previous section, experimentally determined boundary conditions (e.g. temperature gradient, the extent of the solutal diffusion layer and the steady growth velocity) were used in the calculation of the base state case and iterations were carried out on the solutal concentration at the dendrite tips until numerical convergence to within .0005 wt% H₂O was achieved.

The permeability model of Poirier was used since the fraction of liquid for these experiments is very high, and the Drummond and Tahir model is not believed to be as correct for large values of fraction liquid (unlike the Poirier model, the Drummond and Tahir permeability does not approach infinity as the fraction of liquid approaches one). This use is consistent with the conclusions of Poirier.¹⁷

The dendrite arm spacings required for the Poirier model were determined experimentally using a microscope and a solidification apparatus similar to that illustrated in Section 4. The system is described in Appendix C. The microscope system was required due to the small size of the dendrites. The measured temperature gradient and growth velocities in the microscope system were comparable to those in the experiments, so the measured arm spacings are reasonable estimates of those for the experiments. In keeping with the transient nature of the solidification, the measured primary arm spacings varied between 30 and 140 μm , and their orientation sometimes deviated significantly from the heat flow direction. Secondary arm spacings were not measured.

The stability is given in terms of a combined Rayleigh number as defined by the sum of equations 4.8 and 4.9

$$Ra_c = \frac{g B_T \Delta T L^3}{\nu \alpha} + \frac{g B_C \Delta C L^3}{\nu D} \quad 5.70$$

where

$$L = zt + dl$$

$$\Delta C = C_s(0) - C_s(zt + dl) \quad 5.71$$

$$\Delta T = T_s(0) - T_s(zt + dl) = -G_s(zt + dl)$$

Initially, for each set of otherwise identical experimental conditions, two experiments were selected to model and illustrate the influence of zt , one with a low zt and one with a high zt . In the LSM the dendrite arm spacing was set to 50 microns and the characteristic length in the Forchheimer term was set to one tenth the arm spacing, i.e. 5 microns. The arm spacing value is an estimated average value from the microscopic measurements. The value for the Forchheimer term was obtained by assuming that there is an analogy between the sphere diameter on which the Ergun model is based and a characteristic dendrite trunk diameter which is approximately one tenth of the primary arm spacing. Table 5.5 gives the inputs to LSM and the results and Figures 5.23-5.27 show the critical Rayleigh number as a function of zt for LSM and experiments. The effects of zt as calculated by LSM match experimental measurements, but there is an offset in the calculated critical Rayleigh numbers.

In order to include the influence of the secondary arms and the effect of misaligned dendrites, an effective arm spacing of 10 microns appropriate for the

Table 5.5 Experiment Matching Case 1 Results.

GI C/cm	V cm/sec	z _t cm	d _l cm	exp C _{up} wt%	exp Ra _c	LSM a _c 1/cm	LSM C _{up} wt%	LSM Ra _c
12	119e-6	.03646	.06354	.350	5339	23	.0655	1868
12	134e-6	.03542	.06771	.361	5959	23	.0615	1960
14	104e-6	.06354	.04996	.336	9198	25	.0725	4498
14	74e-6	.06250	.04896	.335	8627	25	.0755	4259
10	27e-6	.03414	.08166	.287	6731	19	.0455	2150
11	27e-6	.03293	.07317	.300	5413	21	.0575	1890
10	78e-6	.05104	.06146	.329	7472	22	.0525	2696
10	119e-6	.05000	.06146	.336	7359	23	.0525	2587
10	17e-6	.03260	.08090	.254	5689	20	.0485	2021
10	9e-6	.03214	.08095	.254	5596	20	.0485	1983
11	40e-6	.06840	.05580	.214	8533	23	.0545	4821
11	33e-6	.05263	.05790	.240	5863	23	.0625	2949
8	15e-6	.02840	.06320	.147	1807	24	.0705	1085
10	5e-6	.03158	.06000	.159	2081	25	.0815	1354
10	4e-6	.06280	.05210	.176	5605	25	.0675	3591
10	9e-6	.06170	.05000	.174	5076	25	.0675	3262
16	69e-6	.04790	.05000	.347	5737	27	.0945	2847
16	77e-6	.04479	.04896	.354	5001	28	.1015	2470
18	35e-6	.05625	.04895	.365	8082	27	.0965	4269
18	66e-6	.05625	.04792	.363	7817	27	.0965	4147

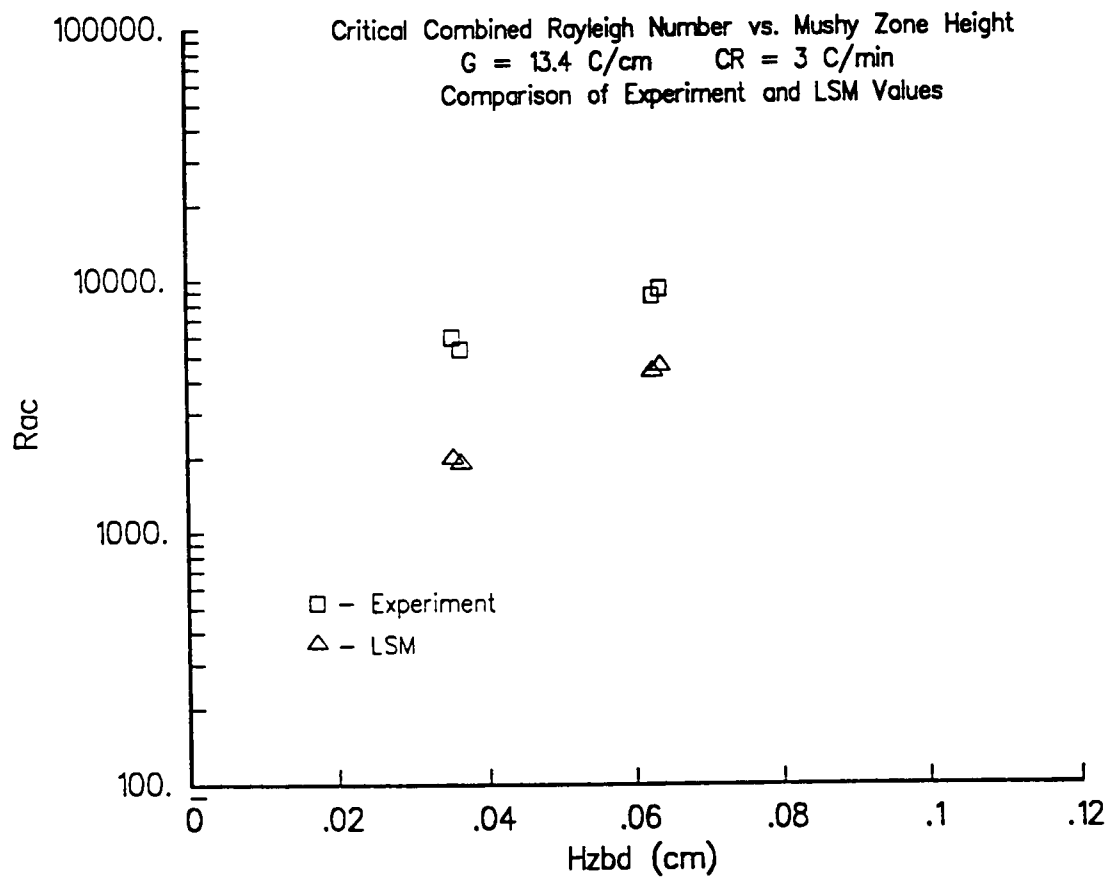


Figure 5.23 Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=50 \text{ }\mu\text{m}$, $d=5 \text{ }\mu\text{m}$ $G=13.4 \text{ }^\circ\text{C/cm}$ $CR=3 \text{ }^\circ\text{C/min}$.

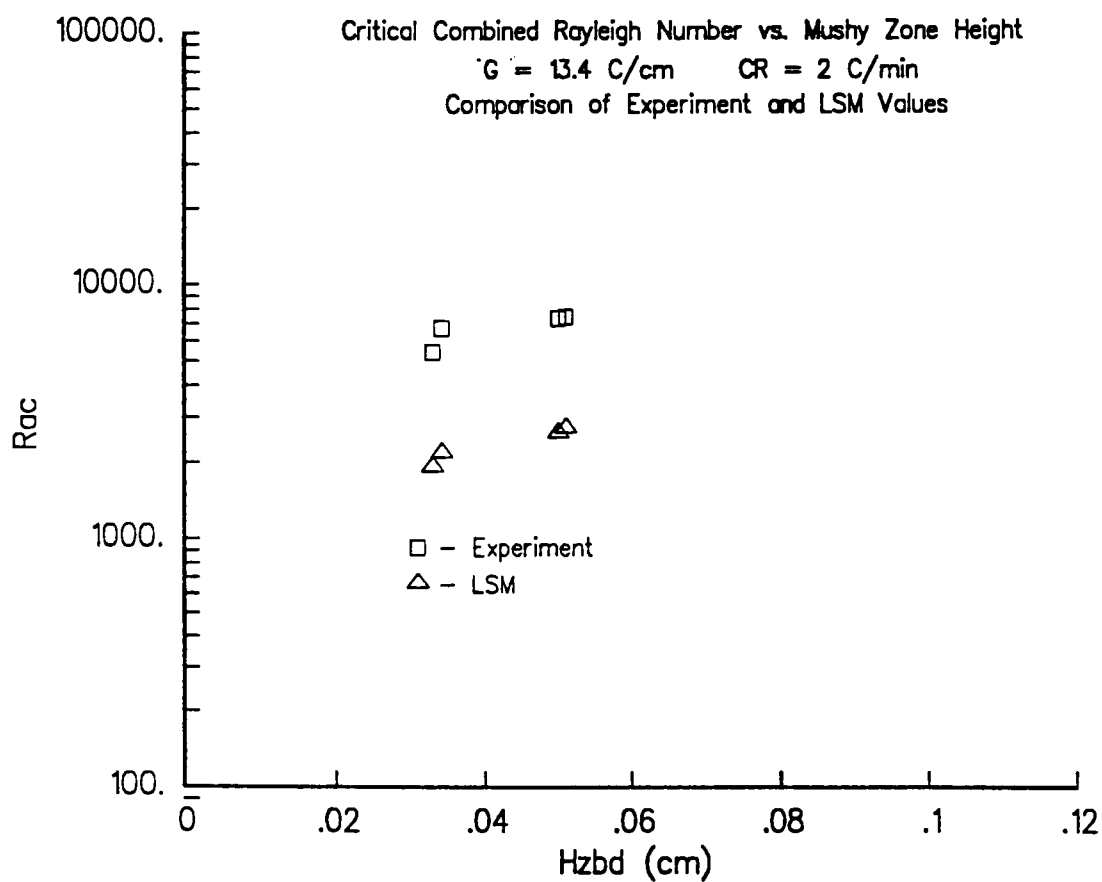


Figure 5.24 Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=50 \text{ } \mu\text{m}$, $d=5 \text{ } \mu\text{m}$ $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=2 \text{ } ^\circ\text{C/min}$.

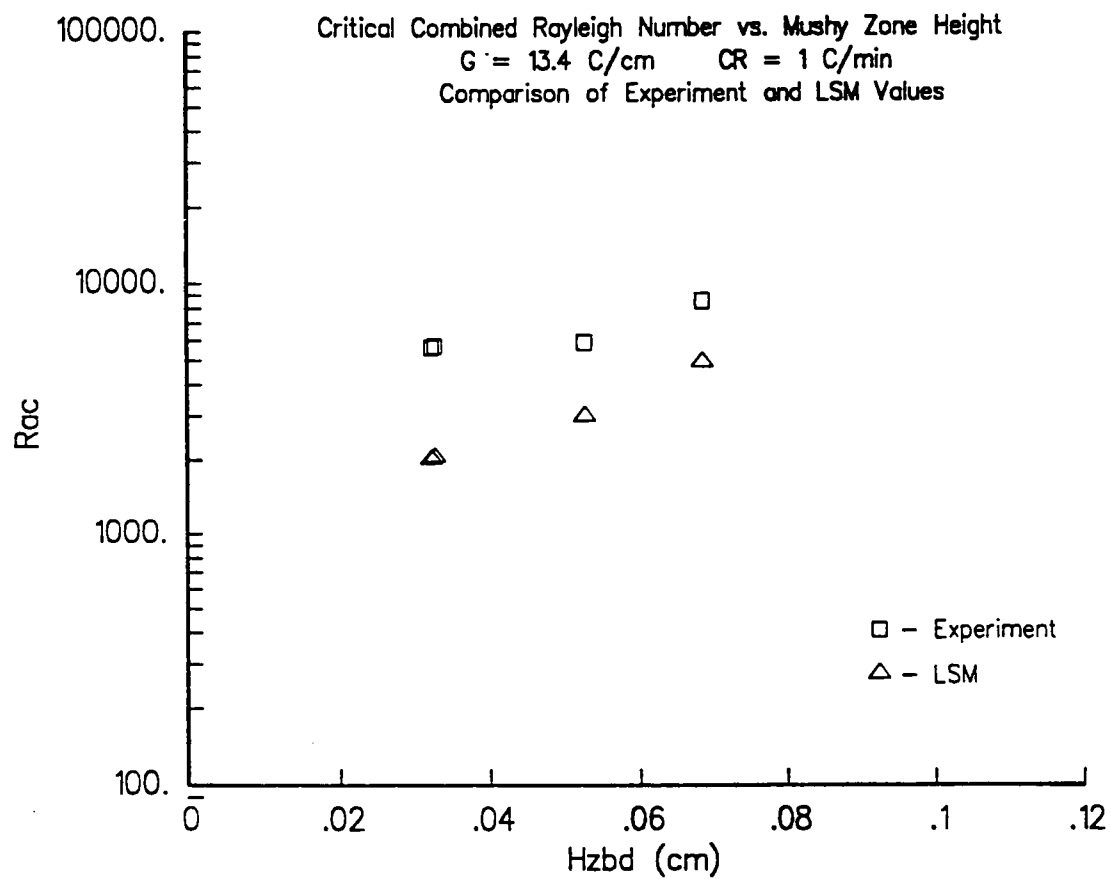


Figure 5.25 Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=50 \text{ } \mu\text{m}$, $d=5 \text{ } \mu\text{m}$ $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=1 \text{ } ^\circ\text{C/min}$.

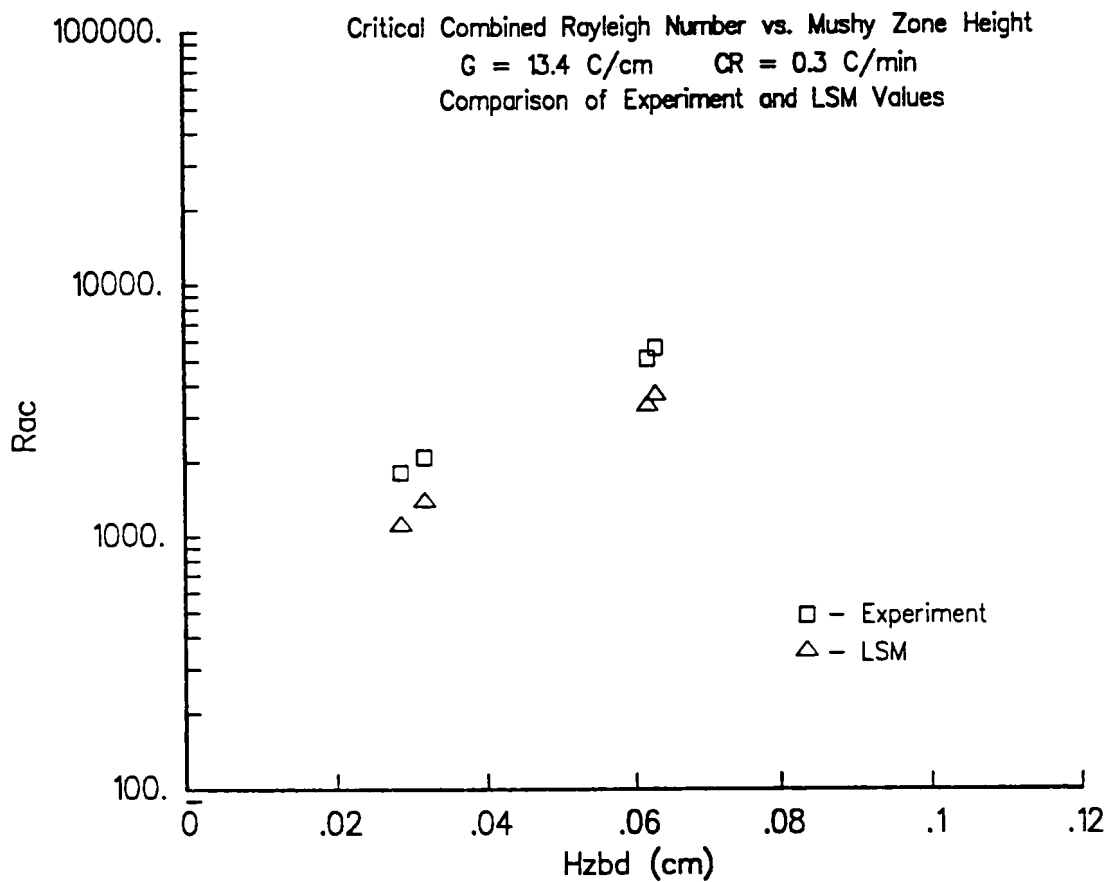


Figure 5.26 Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=50 \text{ } \mu\text{m}$, $d=5 \text{ } \mu\text{m}$ $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=0.3 \text{ } ^\circ\text{C/min}$.

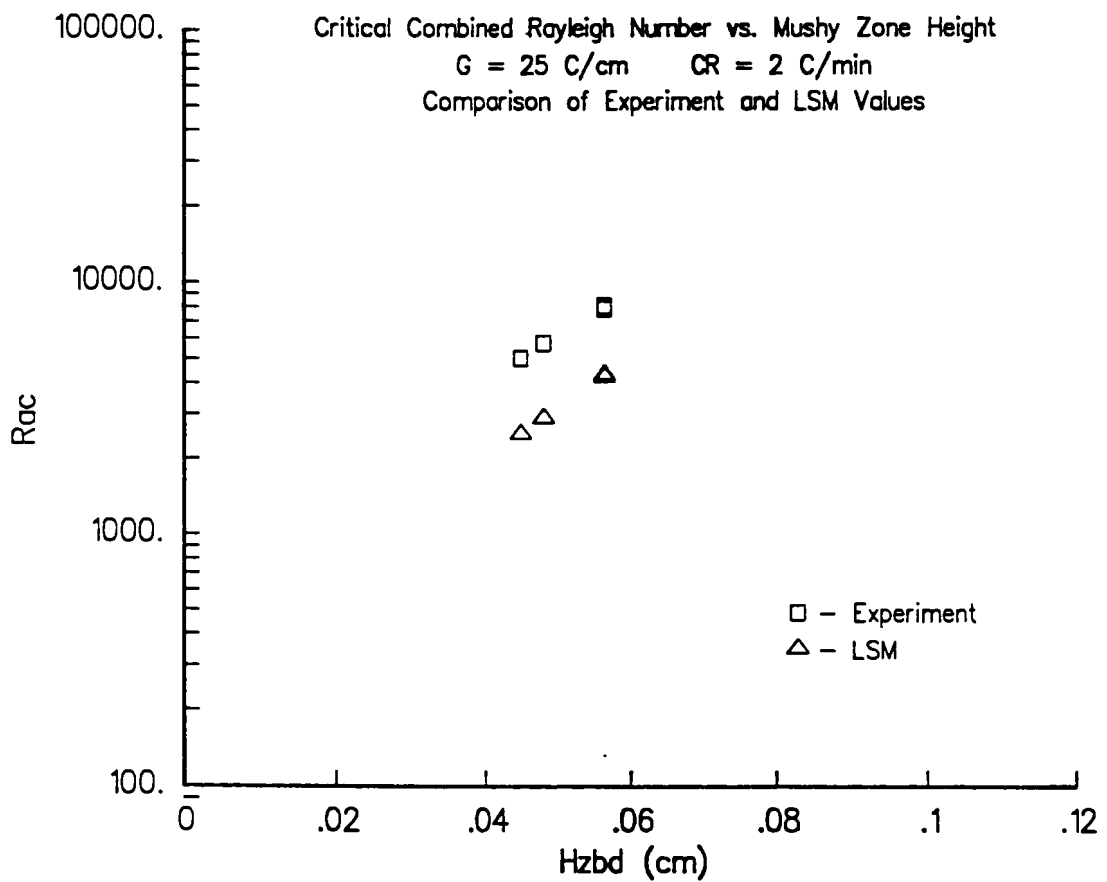


Figure 5.27 Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=50 \mu\text{m}$, $d=5 \mu\text{m}$ $G=25 \text{ }^\circ\text{C/cm}$ $CR=2 \text{ }^\circ\text{C/min}$.

Poirier model was selected (this is the average value for arm spacings required by LSM to match experiment values in the previous section). This value and a value of 1 micron for the Forchheimer term were used for the previous 20 cases and 10 additional cases to fill in the trends in the calculations. Table 5.6 gives the results and Figures 5.28-5.32 show the comparison of LSM calculations and experimental data. The trends are very similar and consistent from case to case. The LSM calculated wavenumbers for these cases also match reasonably well match those of previous investigations of convective breakdown in solidifying $\text{NH}_4\text{Cl-H}_2\text{O}$ systems.⁷⁸ Table 5.7 shows a comparison between previously measured experimental values⁷⁸ and those calculated by LSM. When comparing these wavenumber values, it should be noted that the experimental measurements were taken from photographs of particle streaks at only one plane location within the test cell and that the experiments were subject to finite-domain limitations because of the container walls.

The effect of F on the stability is interesting in that although the general cases show a strong influence of F on breakdown, the experiment matching cases do not show any appreciable influence for F values within the range $.01-10 \times 10^{-4}$ microns. This is due to the considerably slower growth velocities in the experiments (and therefore base case) than in the general cases. Since the magnitude of the inertia term is directly proportional to the base state flow velocity, it can be neglected for the low values associated with these experiments.

The influence of shrinkage flow cannot be neglected, however. A comparison for a selected case shows a critical Rayleigh number of 6379 predicted by LSM

Table 5.6 Experiment Matching Case 2 Results.

GI C/cm	V cm/sec	z cm	dl cm	exp C _{ip} wt%	exp Ra _c	LSM a _c 1/cm	LSM C _{ip} wt%	LSM Ra _c
12	119e-6	.03646	.06354	.350	5339	23	.2045	3606
12	134e-6	.03542	.06771	.361	5959	22	.1805	3599
14	104e-6	.06354	.04996	.336	9198	27	.3335	9282
14	74e-6	.06250	.04896	.335	8627	27	.3535	9074
12	78e-6	.04688	.06142	.319	6690	23	.2215	5254
12	125e-6	.04479	.06351	.327	6733	23	.1915	4708
10	27e-6	.03414	.08166	.287	6731	19	.1145	3509
11	27e-6	.03293	.07317	.300	5413	21	.1535	3331
10	78e-6	.05104	.06146	.329	7472	23	.1985	5303
10	119e-6	.05000	.06146	.336	7359	23	.1985	5121
11	250e-6	.04100	.06200	.269	4784	24	.2005	3930
8	43e-6	.03850	.07300	.236	4897	21	.1365	3391
10	17e-6	.03260	.08090	.254	5689	21	.1525	3161
10	9e-6	.03214	.08095	.254	5596	19	.1165	3228
11	40e-6	.06840	.05580	.214	8533	24	.2475	9474
11	33e-6	.05263	.05790	.240	5863	25	.2525	6154
8	28e-6	.03258	.08542	.242	5880	18	.0955	2994
10	9e-6	.03596	.07644	.274	5978	20	.1315	3570
8	15e-6	.02840	.06320	.147	1807	24	.2155	2461
10	5e-6	.03158	.06000	.159	2081	25	.2595	3045
10	4e-6	.06280	.05210	.176	5605	26	.3045	8068
10	9e-6	.06170	.05000	.174	5076	27	.3235	7696
6	9e-6	.03830	.07340	.168	3627	21	.1295	3013
7	4e-6	.04255	.07025	.171	4025	21	.1505	3720
16	69e-6	.04790	.05000	.347	5737	28	.3885	6283
16	77e-6	.04479	.04896	.354	5001	29	.4225	5753
18	35e-6	.05625	.04895	.365	8082	28	.4215	8999
18	66e-6	.05625	.04792	.363	7817	28	.4315	8878
18	104e-6	.05000	.04792	.352	6100	29	.4145	6908
16	116e-6	.04896	.05000	.356	6065	28	.3755	6379

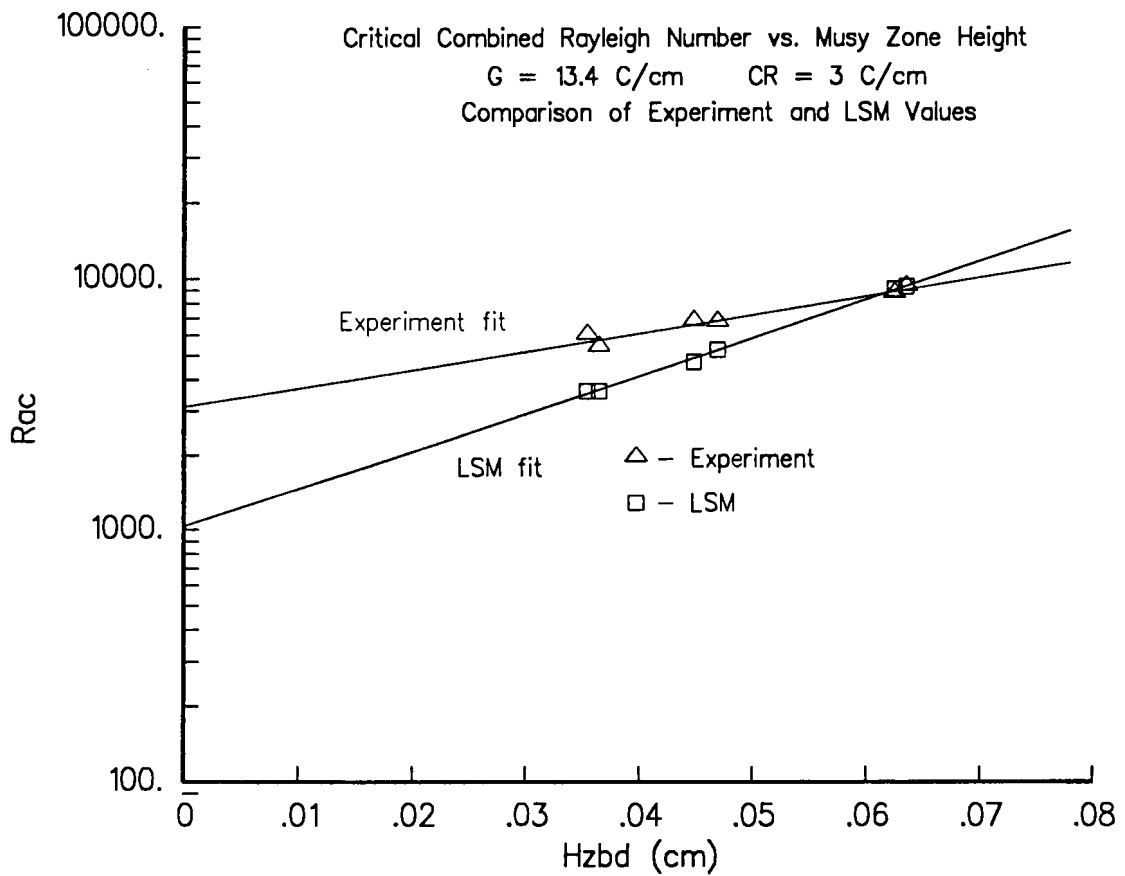


Figure 5.28 Measured and Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=10 \mu\text{m}$, $d=1 \mu\text{m}$ $G=13.4 \text{ }^\circ\text{C/cm}$ $CR=3 \text{ }^\circ\text{C/min}$.

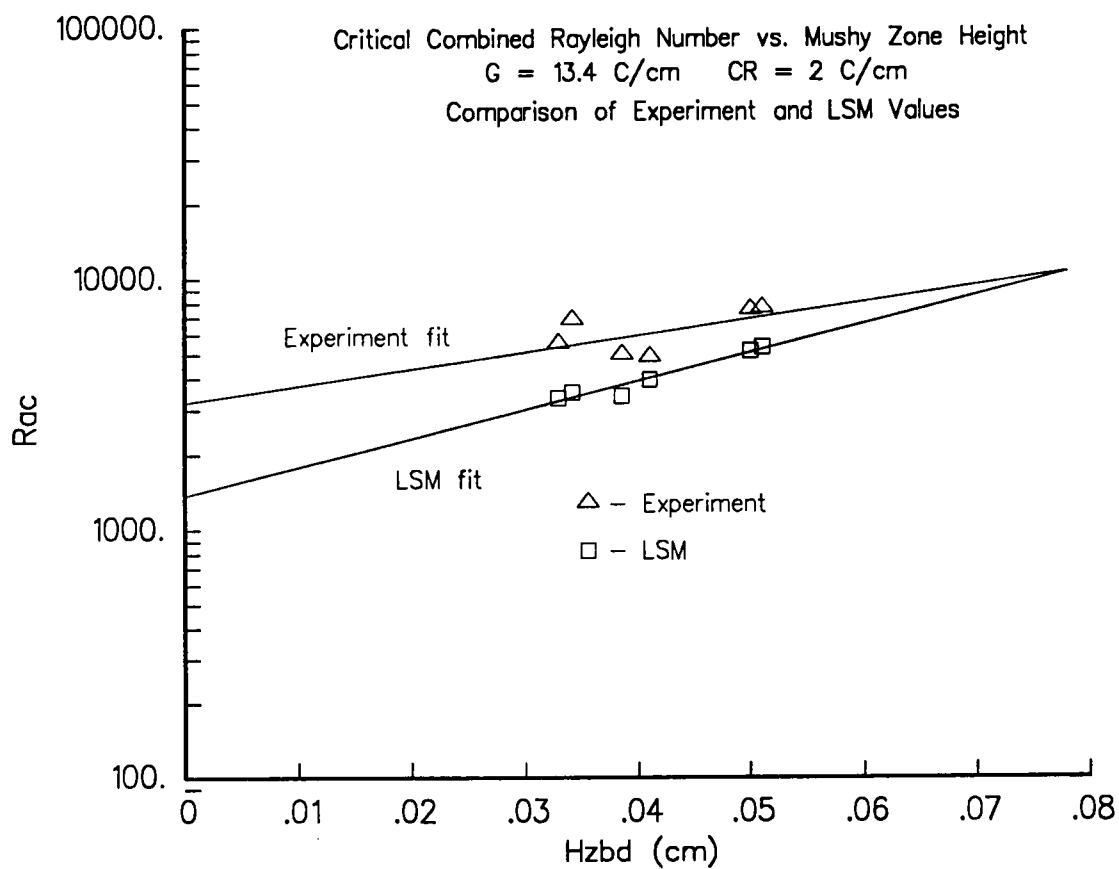


Figure 5.29 Measured and Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=10 \mu\text{m}$, $d=1 \mu\text{m}$ $G=13.4 \text{ }^\circ\text{C/cm}$ $CR=2 \text{ }^\circ\text{C/min}$.

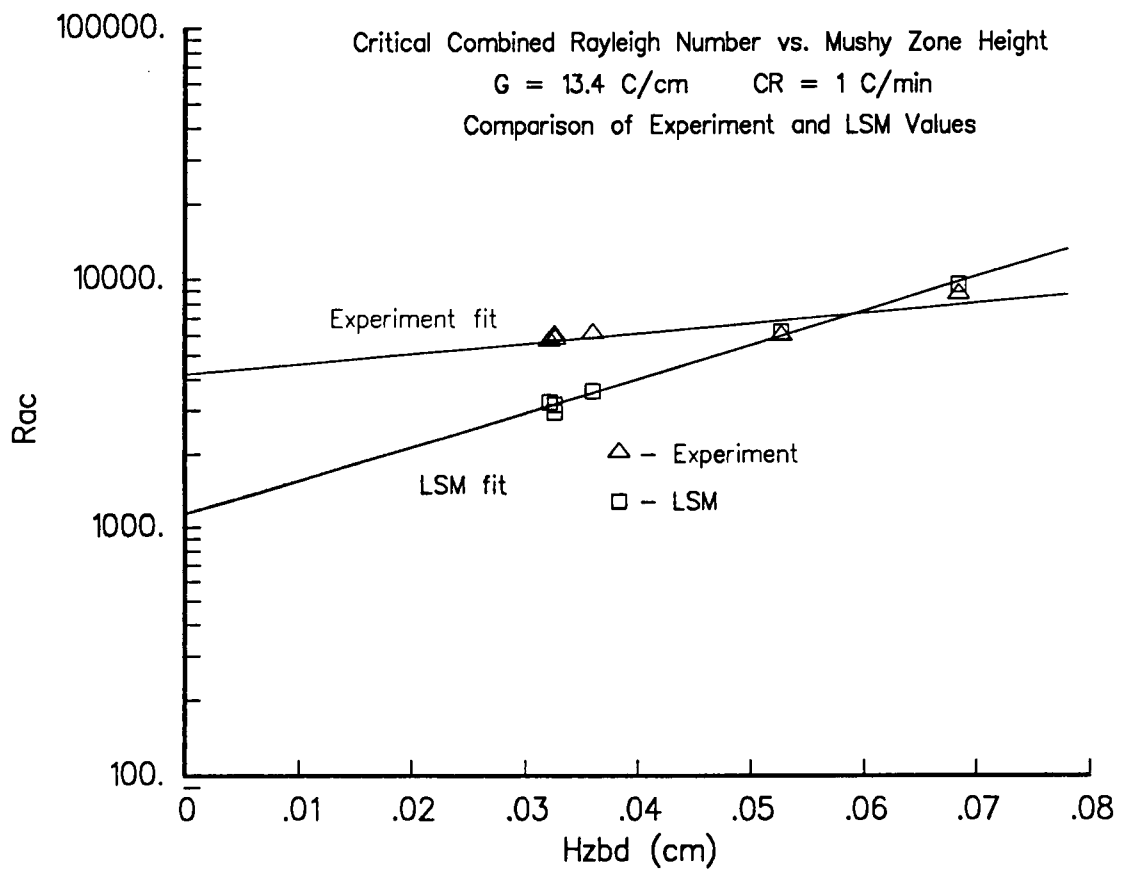


Figure 5.30 Measured and Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=10 \text{ } \mu\text{m}$, $d=1 \text{ } \mu\text{m}$ $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=1 \text{ } ^\circ\text{C/min}$.

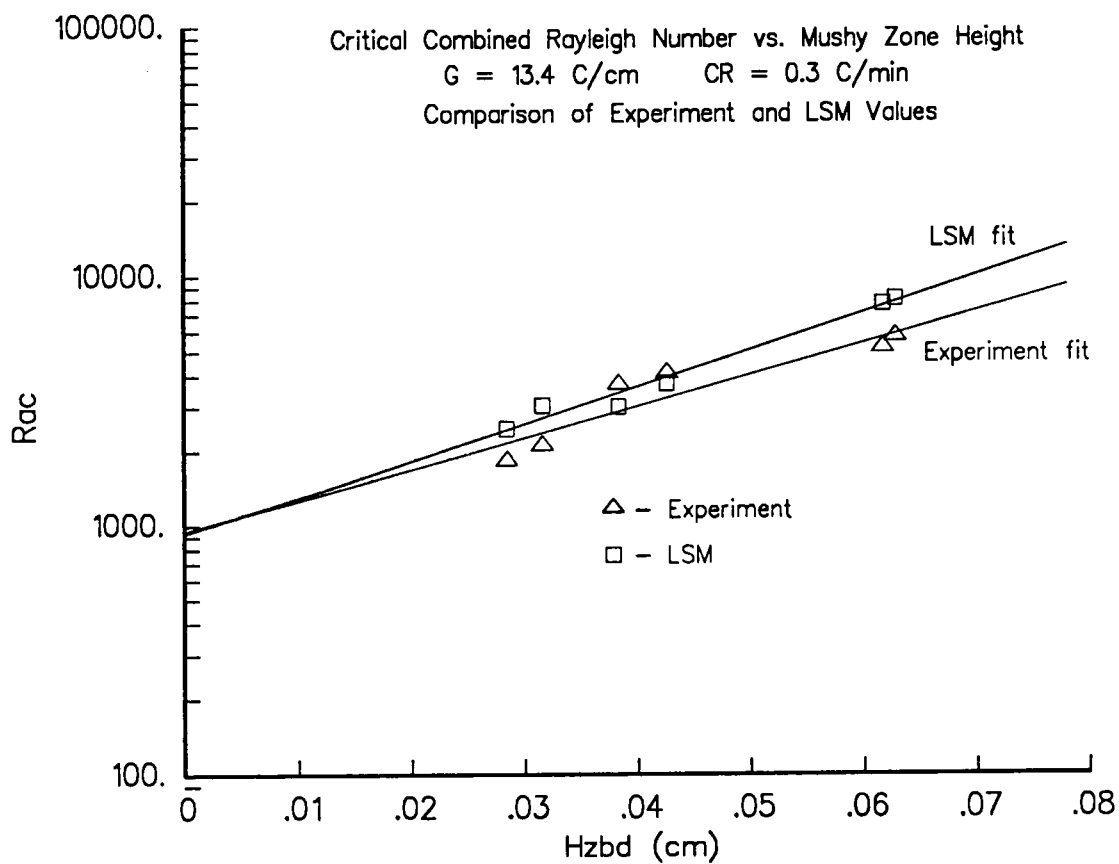


Figure 5.31 Measured and Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=10 \text{ } \mu\text{m}$, $d=1 \text{ } \mu\text{m}$ $G=13.4 \text{ } ^\circ\text{C/cm}$ $CR=0.3 \text{ } ^\circ\text{C/min}$.

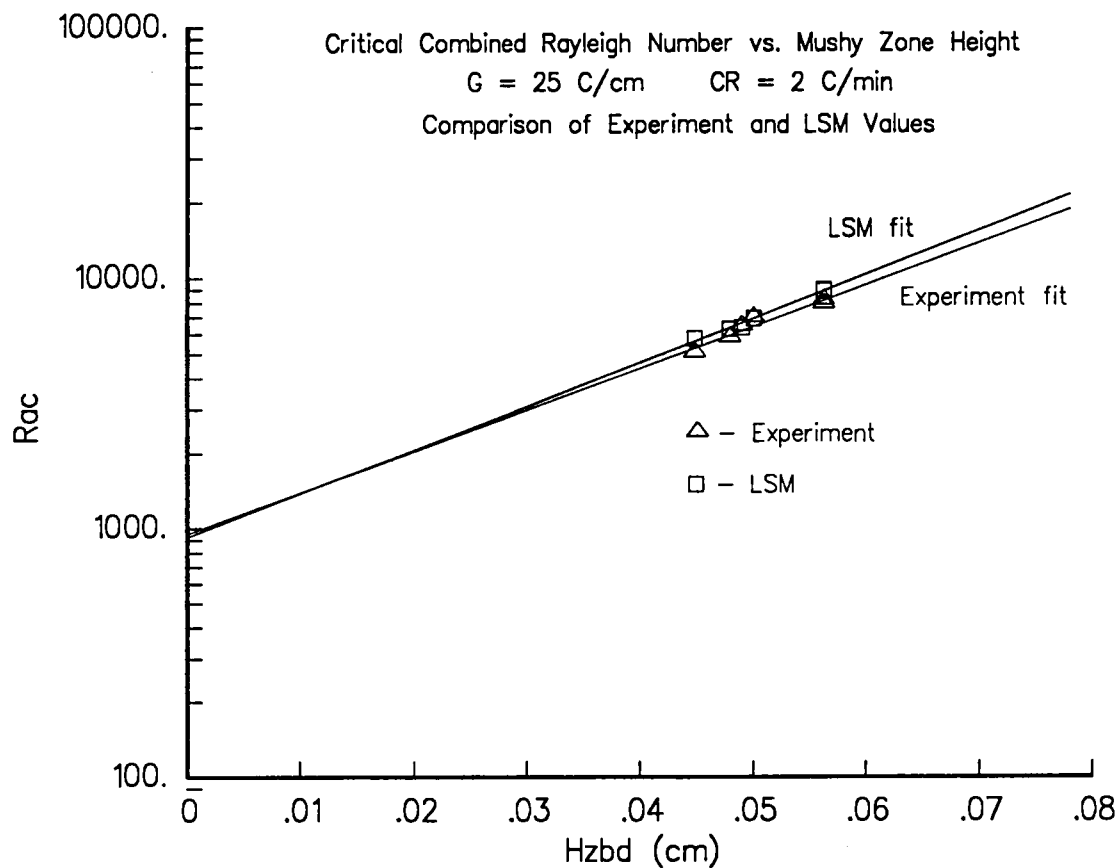


Figure 5.32 Measured and Calculated Critical Rayleigh Numbers vs. Two-Phase Region Height, $d_1=10 \text{ } \mu\text{m}$, $d=1 \text{ } \mu\text{m}$ $G=25 \text{ } ^\circ\text{C/cm}$ $CR=2 \text{ } ^\circ\text{C/min}$.

Table 5.7 LSM Calculated vs. Measured⁷⁸ Convective Cell Size.

Experiment Conditions	LSM λ^* (mm)	Experiment λ^* (mm)
G=13.4 CR=3.0	1.31	1.4
G=13.4 CR=2.0	1.45	1.3
G=13.4 CR=1.0	1.51	1.3
G=13.4 CR=0.3	1.32	1.9
G=25.0 CR=2.0	1.11	1.3

including shrinkage flow and a value of 6805 without shrinkage flow. Hence, the presence of a base flow decreases the stability of the system.

Therefore, with experimental boundary conditions and best values for two-phase region characteristics, LSM has been able to calculate the critical Rayleigh number for VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ (within reasonable bounds), the effect of two-phase region height on convective stability, and the wavelength of the flow structures occurring in the unstable regime.

5.5.6 *SOLIDIFICATION OF $\text{NH}_4\text{Cl-H}_2\text{O}$: EUTECTIC BOUNDARY CONDITIONS*

In the practical application of VDS to casting, the alloy transitions from completely molten to liquid and solid to completely solid as the temperature decreases. To demonstrate the utility of LSM for such applications, the complete solidification of $\text{NH}_4\text{Cl-H}_2\text{O}$ was examined. The base state boundary conditions at $z=0$ are, therefore, the eutectic temperature and concentration. The dendrite solutal tip boundary condition is given by the solute balance as in Section 5.5.3. The allowance for contraction flow is accounted for as in Section 5.5.4. z_l was set to 20 D/V and iterations were performed on g_l as in Section 5.5.3. Table 5.8 gives the results. At accelerations of 1 g_e , growth velocities less than 1×10^{-3} cm/sec are always unstable. Figure 5.33 shows the critical temperature gradient at as a function of growth velocity. At lower gravity levels, slower growth rates become stable. At $2.5 \times 10^{-4} g_e$, all growth rates investigated ($> 1 \times 10^{-5}$ cm/sec) were stable for $G_l < 100$ °C/cm. These results would suggest that low gravity processing and/or very fast growth could be used to yield VDS of NH_4Cl - 72 wt% H_2O without convective flows. However, the

Table 5.8 Solidification of $\text{NH}_4\text{Cl-H}_2\text{O}$ with Eutectic Boundary Conditions Results.

V (cm/sec)	a_c (1/cm)	Gl_c (C/cm)
1.6×10^{-3}	25	39.5
1.4×10^{-3}	22	20.5
1.2×10^{-3}	18	10.5
1.0×10^{-3}	13	3.5
$< 1.0 \times 10^{-3}$	-	unstable for $Gl > 3$

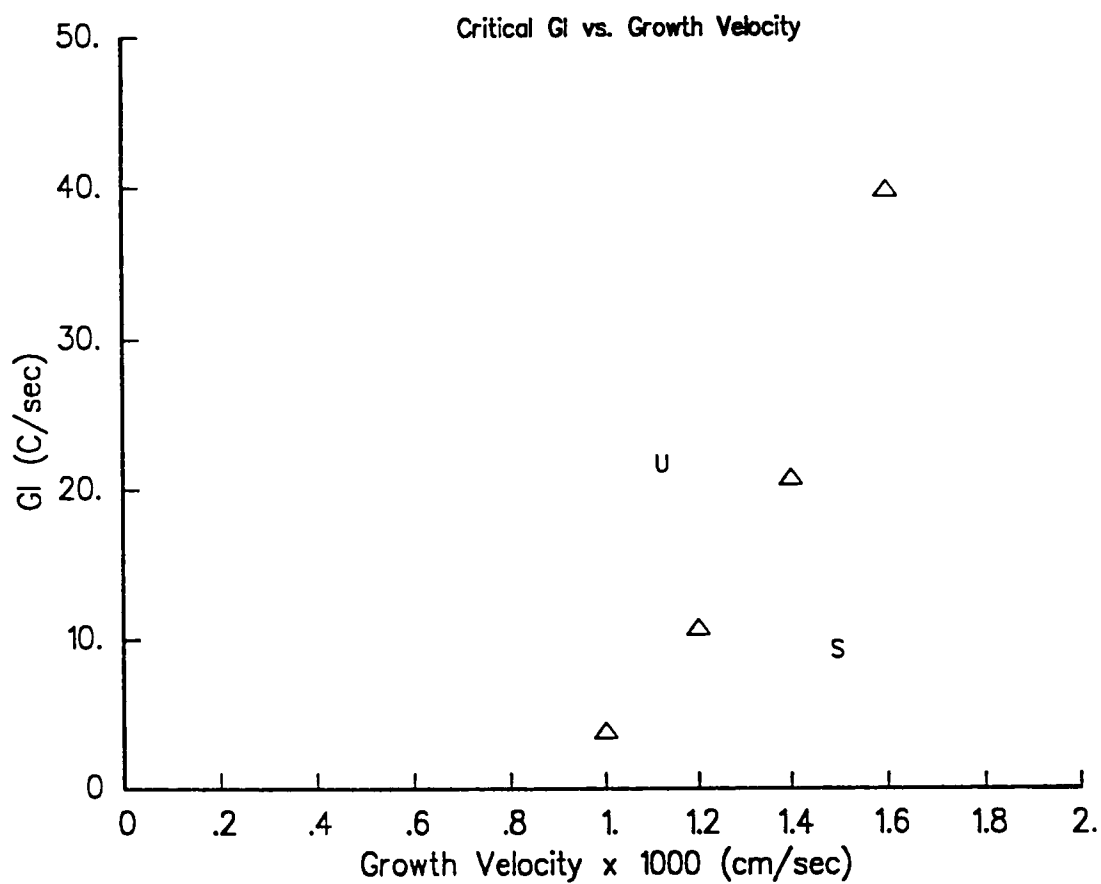


Figure 5.33 $\text{NH}_4\text{Cl-H}_2\text{O}$ Solidification with Eutectic Boundary Conditions: Critical Temperature Gradient as a Function of Growth Velocity.

stability of the initial stages of solidification characterized in section 5.5.5 would also need to be analyzed to determine if the solidification process as a whole was stable. The stabilizing effect of the momentum dissipation and the destabilizing effect of the solute difference, both of which are proportional to $z\tau$, make this necessary.

6. DISCUSSION

6.1 COMPARISON OF EXPERIMENTAL AND NUMERICAL RESULTS

The most straightforward comparison that can be made between the experiments and the numerical analysis is that for the base state solutal and thermal fields. The fact that steady state was not achieved in the experiments necessitated the input of experimental values for the diffusion layer height and temperature gradient in the LSM. The measured growth velocities were also used as input so that the base state solution matched the experimental conditions as closely as possible. Solutal tip concentration was then used as the LSM iteration solution variable.

The experiments revealed an exponential relation between the extent of the two-phase region and the critical combined Rayleigh number. The empirical relation was not the same for all solidification conditions, but the trend was consistent. The experimental data for the $G=13.4$ °C/cm, $CR=3$ and 0.3 °C/min and $G=25$ °C/cm, $CR=2$ °C/min cases showed the least data scatter, and the critical combined Rayleigh numbers calculated by LSM compare favorably for those cases. Previous experimental values of the size of the unstable convective structures for VDS of NH_4Cl - 72 wt% H_2O were also matched by LSM.

If one tries to obtain a reasonable match with experiments, the effects of the two-phase region and the inclusion of shrinkage flow are found to be significant. The

presence of a shrinkage flow of 1×10^{-8} cm/sec results in a 7% difference in the calculated critical Rayleigh number.

The low fluid velocities associated with the growth rates of the experiments resulted in removing any effect from the Forchheimer inertia term. However, the results from Section 5.5.4 showed that for higher base state fluid velocities the inertia term has a marked effect on the stability of the system. This would be applicable to the VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ at higher growth velocities. Such growth velocities were not obtained for the experiments in this study.

Although permeability models for dendritic solidification with the fraction of liquids associated with this study have not yet been developed, the model by Poirier does perform reasonably well when an effective primary dendrite arm spacing of 10 microns is used in the model. Although Ganesan³⁹ has shown the validity of the Drummond and Tahir model for relatively high fractions of liquid, that model does not have the required characteristic of the permeability approaching infinity as the fraction of liquid approaches one. This is necessary when one equation set is used to model both the fluid and two-phase regions. The other permeability model evaluated, the Kozeny-Carmen model, is used frequently in the literature and has seen extensive use in the modelling of dendritic solidification.²⁰⁻²² However, this relation has little justification for use in modelling dendritic systems and gave very different results when compared to the Poirier model.

6.2 USE OF LSM FOR SOLIDIFICATION OF $\text{NH}_4\text{Cl-H}_2\text{O}$ WITH EUTECTIC BOUNDARY CONDITIONS

The solidification cases presented in sections 5.5.4 and 5.5.5 are not representative of the process which would occur during the mass production of a material. In such a situation the $z=0$ boundary condition would be eutectic, i.e. the material for $z<0$ is completely solid. This is the problem modelled by Nandapurkar et al for Pb-Sn.³⁸ In an actual production process, there would also be an initial transient prior to reaching the steady growth regime. This is essentially what is being modelled in sections 5.5.4 and 5.5.5. Application of the LSM for the case of growth with eutectic boundary conditions should be made with the realization that convective flows may be possible in the transient growth regime even if the steady state eutectic regime is stable.

With this caveat, the stability regions for the eutectic boundary conditions show that all growth rates greater than 1×10^{-3} cm/sec are stable for $1 g_e$, and that for a gravity level of $2.5 \times 10^{-4} g_e$, all growth rates considered ($V > 1 \times 10^{-5}$ cm/sec) were stable for $G_I < 100$ °C/cm. This suggests that production in a low gravity environment could be accomplished without convective flows and their detrimental effects.

7. CONCLUSIONS

The convective breakdown of $\text{NH}_4\text{Cl-H}_2\text{O}$ during VDS has been examined experimentally and numerically. The following conclusions are drawn:

1. The resistance to flow due to the two-phase region significantly inhibits convective breakdown, even for large fractions of liquid.
2. Experimental measurements and LSM calculations show that the critical combined Rayleigh number increases with two-phase region size (height).
3. Shrinkage flow can have a significant effect on convective breakdown.
4. The LSM predicts that $\text{NH}_4\text{Cl-H}_2\text{O}$ solidification with eutectic boundary conditions is convectively stable at either high growth rates or low gravity levels.

Although the application of linear stability analysis to VDS has been performed in the past,^{54,55} only recently has the porous nature of a two-phase region been included.³⁸ Also lacking in previous studies were experiments to support the results.

Non-linear analyses have also been applied to directional solidification.^{29,50,87} These usually included some two-phase porous effects and several also included some level of experimental verification. However, these studies typically used boundary conditions that were difficult to duplicate experimentally and/or gravity conditions which prohibit detailed assessment in terms of stability.^{29,50,87}

This study has examined the transition from diffusion-dominated growth to convection-dominated growth using controlled experiments and the LSM, which includes porous media effects and shrinkage flow. LSM was applied to two validation cases and one comparison case with good success. The validation cases have well known theoretical solutions and were matched very well, and the comparison case results also compare favorably.

A set of general test cases was considered to identify the individual influence of various problem parameters. The influence that the growth velocity, two-phase region, gravity level, and shrinkage flow have on the convective stability of the VDS of NH_4Cl - 72 wt% H_2O was examined. The size of the stable region of growth was shown to be directly proportional to growth velocity (i.e. slower growth rates were less stable). The stable region size was also determined to be directly proportional to the amount of momentum dissipation in the two-phase region. This momentum dissipation could be realized through either increased permeability, increased two-phase region extent, or increased inertia losses. The size of the stable region increased with decreasing gravity level. The presence of a shrinkage flow tended to be destabilizing, and the inclusion of a Forchheimer term had a stabilizing effect.

The experiment matching results give a 7% difference between the critical combined Rayleigh numbers calculated with and without a base state shrinkage flow. The effect of the base flow is easily explained if the system is examined in terms of energy. Within the system, there must exist sufficient energy to transition from one state to another. Only the energy due to the gravitational potential is usually

considered since it is the driver for convection in most thermosolutal stability problems. The presence of a base state flow, however, gives the system additional energy, hence the energy needed to transition to the next state is less than when a quiescent base state is considered.

The effective primary dendrite arm spacings used in the LSM to match experiments was 10 microns. This value is outside the range of arm spacings independently measured under similar growth conditions, but is a reasonable value when one considers the dendrites observed were not always aligned with the direction of growth. An alternative approach would be to develop a suitable characteristic length and permeability model for each individual case, but this is generally unrealistic. The value of 10 microns for an effective arm spacing used with the Poirier permeability model adequately characterizes the experiments in this study.

Using experimental boundary conditions, the LSM was able to calculate: 1) the critical Rayleigh number for selected experiments to within an average of 26 % error, 2) the relation between critical combined Rayleigh number and two-phase region height, and 3) the size of the convective cells within 16 % error of previously measured experimental values.

The application of LSM to the VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ with eutectic boundary conditions shows the potential for the use of LSM. The dependence of stability on gravity level and growth rate was shown. Convective breakdown was delayed by either increasing growth rate or decreasing gravity level. This shows the potential for low-gravity processing of materials without the effects of convective flows.

In summary, experimental and numerical analysis of convective breakdown occurring during the VDS of $\text{NH}_4\text{Cl-H}_2\text{O}$ has been undertaken. The stabilizing influence of the two-phase region has been shown by measuring and calculating critical combined Rayleigh numbers at convective breakdown. An existing permeability model and estimates of effective dendrite arm spacings were selected to characterize the two-phase region. The measured and calculated critical combined Rayleigh numbers compare well and the wavelengths of the convective cells match previously measured values. The application of the linear model to constant velocity solidification with eutectic boundary conditions shows that growth is always convectively stable for low gravity levels and high growth rates.

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APPENDICES

**APPENDIX A
EQUIPMENT LISTS**

CONTROL SYSTEM FOR SOLIDIFICATION

See Figure 4.4

Computer

Personal Computer - IBM PC-XT

Interface Card - Keithley

Input/Output Device

Data Acquisition System - Keithley 570

Thermocouple Card - Keithley AIM7

Operational Amplifier/Power Supply Assembly

2 Operational Amplifiers - Burr Brown OPA 512 SM

2 DC Power Supplies - Condor F15-15.0

Cuvette Assembly

Quartz Cuvette with Expansion Tube - Spectrocell - see Figure 4.1

Chromel-Alumel (K) Thermocouple Wire - Omega TT-K-36

2 Copper Plates Slotted for Thermocouples

2 Thermoelectric Devices (TED) - Melcore CP1.4-71-045L

2 Copper Heat Sinks

4 Aluminum Angles

OPTICS SYSTEM FOR INTERFEROMETRY

See Figure 4.5

Laser

Helium-Neon Laser - Aerotech LL25P

2 Adjustable Radius Chucks - Newport AC-3

2 Support Posts - Newport VPH-2

2 Sliding Bases - Newport B-2A

2 Adjustable Mounting Bases - Newport AMB-2

Collimator

Collimating Lens - Spectra-Physics 336

Adjustable Radius Chuck - Newport AC-3

Support Post - Newport VPH-2

Sliding Base - Newport B-2A

Adjustable Mounting Base - Newport AMB-2

Mirror M1

Mirror, 75 mm diameter - Melles Griot 02 MFG 021

Support Post - Newport VPH-3

Sliding Base - Newport B-2A

Adjustable Mounting Base - Newport AMB-2

Beam Splitter B1

Beam Splitter - Ealing 24-5043

Filter Holder - Oriel 12650

Support Post - Newport VPH-3

Sliding Base - Newport B-2A

Adjustable Mounting Base - Newport AMB-2

Mirror M2

Mirror, 100 mm diameter - Melles Griot 02 MFG 023

Rod Mounted Kinematic Mirror Mount - Newport 625

Objective Mount - Newport MM-2A

2 Translation Stages - Newport TSX-1A

Mirror M3

Mirror, 100 mm diameter - Melles Griot 02 MFG 023

Rod Mounted Kinematic Mirror Mount - Newport 625

Cuvette Assembly

Laboratory Jack - Newport 270

Low Profile Translation Stage - Newport 440

Beam Splitter B2

Same as Beam Splitter B1

Lens

Lens, 120 mm focal length, 50 mm diameter - Ealing 42-1727

Adjustable Radius Chuck - Newport AC-2

Support Post - Newport VPH-3

Sliding Base - Newport B-2A

Adjustable Mounting Base - Newport AMB-2

Filter

Neutral Density Filter Set - Newport FS-3

Filter Holder - Newport FH-1

Support Post - Newport VPH-2

Base Clamp - Newport BC-1

Camera

CID Camera - CIDTECH 2205

Support Post - Newport VPH-2

Sliding Base - Newport B-2A

APPENDIX B

DATA

	run	# fringes	Timebdt	Hzi (cm)	Hxbdt(cm)	Hmbdt(cm)	GK(C/cm)	dt (C)	dc (wt%)	RaC
G = 13.4	1009901	3.5	60	0.0604	0.0667	0.1104	10.21	1.12711	0.35675	5821
CR = 2	1009902	5.5	70	0.0469	0.0521	0.1073	10.45	1.12129	0.43972	6589
Tbot = 19	1010901	3.5	60	0.0469	0.0552	0.1042	10.71	1.11518	0.34664	4748
	1010902	5.5	70	0.0458	0.0500	0.1021	10.97	1.12020	0.44004	5679
	1011901	4.5	70	0.0458	0.0510	0.1052	11.25	1.18410	0.40084	5659
	1011902	5.5	80	0.0448	0.0510	0.1125	09.75	1.09734	0.43259	7473
	1012901	5.5	70	0.0417	0.0500	0.1115	10.21	1.13775	0.43812	7359
	1012902	5.5	75	0.0448	0.0521	0.1156	10.45	1.20838	0.44897	8418
	1015901	5.5	90	0.0438	0.0552	0.0990	14.63	1.44788	0.50043	5880
	1016901	5.5	95	0.0469	0.0542	0.0969	16.88	1.63546	0.53089	5851
G = 13.4	828891	1.5	60	0.0320	0.0510	0.0960	08.02	0.77006	0.19497	2088
CR = 2	828892	3.5	80	0.0150	0.0370	0.0940	08.63	0.81093	0.28747	2895
Tbot = 20	829891	3.5	80	0.0160	0.0330	0.0940	07.75	0.72846	0.27245	2745
	829892	3.5	90	0.0150	0.0350	0.0990	07.37	0.73009	0.27285	3211
	830891	4.0	100	0.0160	0.0410	0.1030	11.15	1.14864	0.36122	4784
	831891	3.5	90	0.0170	0.0410	0.1020	09.94	1.01385	0.31774	4087
	831892	4.0	90	0.0160	0.0360	0.0980	09.94	0.97409	0.33287	3799
	901891	4.0	90	0.0170	0.0360	0.1020	07.50	0.76454	0.30193	3887
	901892	3.5	80	0.0190	0.0380	0.1010	07.50	0.75705	0.27836	3478
	905891	3.5	85	0.0140	0.0340	0.0930	06.82	0.63466	0.26014	2539
G = 13.4	922891	3.5	85	0.0344	0.0510	0.1229	06.97	0.85639	0.29590	6663
CR = 2	1006891	3.5	75	0.0229	0.0417	0.1042	05.70	0.59380	0.25635	3517
Tbot = 21	1006892	3.5	70	0.0281	0.0375	0.1021	06.01	0.61381	0.25729	3321
	1009891	3.5	70	0.0323	0.0427	0.1000	07.44	0.74396	0.28022	3399
	1009892	3.0	80	0.0229	0.0333	0.0906	07.44	0.67421	0.24166	2181
	1012891	3.0	80	0.0292	0.0479	0.1167	06.45	0.75308	0.25609	4930
	1012892	4.0	75	0.0156	0.0323	0.0844	06.18	0.52162	0.26921	1964
	1114891	5.0	90	0.0315	0.0382	0.1090	08.66	0.94364	0.37549	5898
	1115891	4.5	90	0.0315	0.0348	0.1079	09.69	1.04509	0.36266	5519
	1115892	3.5	80	0.0281	0.0326	0.0978	05.81	0.56827	0.24889	2822
G = 13.4	1121891	3.5	90	0.0346	0.0385	0.1115	07.75	0.86475	0.29111	4897
CR = 2	1121892	1.5	85	0.0333	0.0359	0.1077	07.28	0.78381	0.18181	2747
Tbot = 22	1122891	4.5	85	0.0280	0.0329	0.1098	09.87	1.08290	0.36543	5858
	1127891	4.0	85	0.0293	0.0329	0.1085	10.71	1.16266	0.35238	5459
	1127892	4.0	90	0.0293	0.0317	0.1012	10.13	1.02567	0.33473	4208
	1128891	4.0	85	0.0280	0.0305	0.1098	09.37	1.02876	0.33155	5313
	1129891	4.0	85	0.0305	0.0329	0.1061	11.03	1.16996	0.35418	5126
	1129892	4.0	90	0.0317	0.0341	0.1159	10.41	1.20657	0.35733	6732
	1130891	4.5	90	0.0305	0.0329	0.1061	10.71	1.13653	0.37387	5413
	1130892	4.0	90	0.0280	0.0341	0.1098	11.03	1.21030	0.35979	5764
G = 13.4	1201891	3.5	120	0.0310	0.0333	0.1131	10.67	1.20657	0.33306	5835
CR = 1	1201892	3.5	130	0.0310	0.0321	0.1131	09.85	1.11375	0.31935	5596
Tb = 22	1204891	3.5	130	0.0298	0.0321	0.1119	10.11	1.13103	0.32201	5466
	1204892	3.5	125	0.0298	0.0310	0.1000	09.60	0.96017	0.30114	3650
	1205891	4.0	135	0.0298	0.0333	0.1143	10.38	1.18631	0.35423	6407
	1206891	4.0	125	0.0281	0.0303	0.1067	11.00	1.17396	0.35169	5183
	1206892	4.0	125	0.0281	0.0303	0.1056	09.93	1.04827	0.33514	4786
	1207891	3.5	125	0.0292	0.0315	0.1056	11.30	1.19387	0.33172	4733
	1207892	3.5	130	0.0303	0.0326	0.1135	09.93	1.12634	0.32136	5689
	1208891	4.0	125	0.0292	0.0315	0.1101	10.43	1.14893	0.34851	5638

	run	# fringes	Timebd	Hzi (cm)	Hzbd(cm)	Hmbd(cm)	GI(C/cm)	dt (C)	dc (wt%)	RaC
G = 13.4	1211891	4.0	125	0.0292	0.0303	0.1067	10.17	1.08591	0.33988	5010
CR = 1	1212891	4.0	120	0.0326	0.0337	0.0831	10.71	0.89039	0.32430	2261
Tbot = 21	1214891	3.5	115	0.0258	0.0292	0.1270	05.28	0.67099	0.25633	6364
	1218891	3.5	120	0.0292	0.0326	0.1180	07.98	0.94135	0.29546	5880
	1219891	4.0	120	0.0348	0.0360	0.1124	09.93	1.11518	0.34775	5978
	1219892	4.0	130	0.0366	0.0378	0.1305	09.14	1.19325	0.35493	9555
	1221891	3.5	120	0.0317	0.0341	0.1293	07.81	1.00971	0.30338	7940
	102901	3.0	115	0.0329	0.0341	0.1122	07.98	0.89499	0.26750	4577
	102902	3.0	105	0.0317	0.0329	0.1146	07.07	0.81093	0.25468	4650
	123901	3.0	115	0.0333	0.0368	0.1057	11.37	1.20185	0.31447	4502
G = 13.4	410901	2.0	95	0.1000	0.1042	0.1537	10.34	1.58940	0.37230	16349
CR = 1	410902	3.0	105	0.0642	0.0684	0.1242	11.14	1.38340	0.36775	8533
Tbot = 19	411901	3.0	85	0.0516	0.0568	0.1095	10.59	1.15979	0.32821	5216
	411902	3.5	95	0.0495	0.0526	0.1105	11.14	1.23099	0.35845	5863
	412901	3.0	85	0.0453	0.0505	0.1032	10.86	1.12020	0.31872	4238
	412902	3.0	85	0.0442	0.0474	0.1000	11.14	1.11375	0.31595	3827
	416901	2.5	75	0.0600	0.0632	0.1074	10.59	1.13749	0.30820	4619
	416902	3.5	100	0.0453	0.0516	0.1053	10.86	1.14306	0.34654	4898
	417901	3.0	90	0.0463	0.0484	0.0989	11.43	1.13103	0.32034	3759
	430901	3.0	100	0.0537	0.0579	0.1105	08.69	0.96017	0.29721	4863
G = 13.4	501901	3.5	100	0.0337	0.0347	0.1000	08.20	0.81955	0.28498	3455
CR = 1	502901	3.5	120	0.0305	0.0316	0.0979	08.04	0.78744	0.27854	3169
Tbot = 20	503901	4.0	120	0.0326	0.0326	0.0979	08.04	0.78744	0.30364	3456
	507901	3.5	85	0.0368	0.0389	0.0905	08.20	0.74191	0.28015	2521
	508901	3.5	100	0.0316	0.0326	0.0979	08.04	0.78744	0.27937	3178
	509901	3.5	110	0.0295	0.0295	0.0905	08.04	0.72817	0.27057	2435
	510901	4.5	110	0.0211	0.0211	0.0832	08.04	0.66890	0.30611	2137
	514901	1.5	90	0.0905	0.0905	0.1274	08.52	1.08479	0.26415	6605
	515901	3.5	105	0.0474	0.0495	0.1074	10.10	1.08458	0.33437	5015
	515902	3.5	115	0.0505	0.0526	0.1095	09.05	0.99066	0.32208	5122
G = 13.4	516901	2.0	220	0.0400	0.0421	0.0979	08.86	0.86780	0.22606	2568
CR = 0.3	516902	1.5	210	0.0389	0.0400	0.0968	07.90	0.76481	0.18520	2035
Tbot = 22	517901	1.5	215	0.0347	0.0368	0.0947	07.76	0.73483	0.17903	1842
	517902	2.0	215	0.0253	0.0284	0.0916	07.76	0.71033	0.19424	1807
	518901	1.5	210	0.0274	0.0305	0.0905	07.24	0.65536	0.16420	1474
	519901	2.0	225	0.0305	0.0316	0.0916	09.87	0.90406	0.22383	2081
	521901	2.0	205	0.0295	0.0305	0.0916	07.24	0.66298	0.18927	1761
	521902	2.0	225	0.0284	0.0305	0.0895	07.90	0.70662	0.19589	1699
	522901	2.5	240	0.0326	0.0368	0.0863	10.10	0.87192	0.25065	1953
	525901	2.5	250	0.0347	0.0400	0.0905	10.10	0.91445	0.25832	2321
G = 13.4	531901	2.5	240	0.0340	0.0394	0.0979	10.74	1.05162	0.27475	3118
Cr = 0.3	531902	2.0	240	0.0404	0.0415	0.0989	10.74	1.06305	0.25396	2976
Tbot = 21	601901	2.0	215	0.0372	0.0394	0.0957	10.48	1.00367	0.24437	2596
	604901	2.5	220	0.0383	0.0415	0.1117	05.97	0.66679	0.21657	3659
	604902	2.5	240	0.0362	0.0383	0.1117	05.97	0.66679	0.21469	3627
	605901	2.5	225	0.0372	0.0394	0.1043	06.23	0.64939	0.21448	2946
	605902	2.5	210	0.0383	0.0394	0.1106	05.89	0.65139	0.21337	3503
	606901	2.5	240	0.0415	0.0426	0.1128	06.82	0.76930	0.23167	4025
	607901	2.5	240	0.0489	0.0500	0.1064	06.14	0.65318	0.22100	3226
	607902	2.5	250	0.0468	0.0489	0.1128	06.61	0.74563	0.23245	4039

	run	# fringes	Timebd	Hzi (cm)	Habd(cm)	Hmbd(cm)	GI(C/cm)	dt (C)	dc (wt%)	RaC
G = 13.4	620901	3.0	260	0.0553	0.0574	0.1149	06.72	0.77157	0.26561	4884
Cr = 0.3	620902	3.0	230	0.0574	0.0617	0.1202	07.28	0.87570	0.28296	5958
Tbot = 20	625901	2.5	240	0.0553	0.0574	0.1170	07.96	0.93139	0.26538	5151
	625902	2.5	210	0.0617	0.0617	0.1149	07.54	0.86632	0.25926	4764
	626901	2.5	225	0.0585	0.0596	0.1117	07.81	0.87288	0.25999	4390
	627901	2.5	235	0.0574	0.0596	0.1128	08.27	0.93204	0.26892	4671
	628901	2.5	190	0.0585	0.0585	0.1074	08.11	0.87132	0.26071	3918
	628902	1.5	245	0.0596	0.0617	0.1128	07.81	0.88120	0.21399	3713
	629901	2.5	225	0.0596	0.0606	0.1085	08.11	0.87994	0.26333	4076
	629902	2.5	240	0.0596	0.0617	0.1170	07.67	0.89812	0.26346	5115
G = 13.4	702901	1.5	180	0.0543	0.0766	0.1170	09.34	1.09337	0.25959	5033
Cr = 0.3	702902	2.0	180	0.0617	0.0649	0.1117	08.43	0.94135	0.25103	4236
Tbot = 19	703901	2.5	240	0.0596	0.0617	0.1117	10.00	1.11648	0.30079	5076
	703902	2.5	260	0.0617	0.0617	0.1213	08.43	1.02203	0.28120	6075
	705901	0.5	120	0.1117	0.1117	0.1457	09.77	1.42363	0.28323	10595
	705902	2.5	210	0.0585	0.0628	0.1128	08.60	0.96932	0.27754	4821
	706901	2.5	230	0.0596	0.0606	0.1106	08.95	0.99066	0.28014	4595
	706902	2.5	300	0.0617	0.0628	0.1149	10.00	1.14838	0.30523	5605
	709901	1.5	170	0.0787	0.0840	0.1213	09.34	1.13312	0.27069	5842
	709902	2.0	170	0.0596	0.0617	0.1053	08.43	0.88756	0.24266	3433
G = 13.4	717901	5.0	70	0.0281	0.0375	0.0990	11.55	1.14306	0.40678	4780
Cr = 3	718901	5.5	70	0.0260	0.0354	0.1031	12.54	1.29329	0.44808	5958
Tbot = 22	718902	5.5	70	0.0281	0.0365	0.1000	12.19	1.21927	0.44026	5339
	719901	6.0	75	0.0302	0.0354	0.1052	12.19	1.28277	0.47000	6638
	720901	6.0	75	0.0365	0.0396	0.1188	08.13	0.96525	0.42542	8647
	720902	6.0	75	0.0313	0.0396	0.1167	07.44	0.86795	0.41240	7951
	723901	5.5	70	0.0292	0.0385	0.0990	12.54	1.24104	0.44641	5246
	723902	5.5	75	0.0292	0.0396	0.1031	13.30	1.37168	0.46454	6177
	724901	6.0	75	0.0323	0.0406	0.1188	07.98	0.94770	0.42380	8614
	724902	6.0	75	0.0271	0.0385	0.1219	07.84	0.95527	0.42242	9282
G = 13.4	726901	5.5	80	0.0344	0.0448	0.1063	11.86	1.26046	0.45321	6593
CR = 3	726902	5.0	80	0.0333	0.0427	0.1052	11.55	1.21526	0.42039	5936
Tbot = 21	727901	5.0	75	0.0333	0.0406	0.1052	11.86	1.24810	0.42275	5969
	727902	5.0	75	0.0333	0.0427	0.1083	11.86	1.28517	0.42913	6614
	730901	5.0	70	0.0354	0.0427	0.1063	12.54	1.33249	0.43701	6354
	730902	5.0	80	0.0333	0.0458	0.1010	11.86	1.19867	0.42361	5299
	731901	5.0	80	0.0344	0.0448	0.1063	12.19	1.29547	0.43413	6313
	731902	5.0	75	0.0375	0.0469	0.1167	11.25	1.31306	0.43416	8358
	801901	5.0	80	0.0406	0.0469	0.1083	11.86	1.28517	0.43402	6690
	802901	5.0	75	0.0354	0.0448	0.1083	12.19	1.32087	0.43682	6733
G = 13.4	802902	5.0	75	0.0490	0.0552	0.1146	12.91	1.47926	0.47010	8572
CR = 3	806901	4.5	75	0.0531	0.0594	0.1104	16.26	1.79503	0.50432	8223
Tbot = 20	806902	5.0	80	0.0500	0.0563	0.1063	15.68	1.66561	0.50660	7363
	807901	5.0	75	0.0490	0.0542	0.1104	15.68	1.73092	0.51030	8323
	808901	5.5	75	0.0469	0.0552	0.1115	15.14	1.68700	0.52857	8870
	808902	5.5	75	0.0490	0.0583	0.1135	15.14	1.71854	0.53659	9519
	809901	5.5	75	0.0521	0.0563	0.1104	16.26	1.79503	0.54782	8936
	809902	4.5	70	0.0490	0.0573	0.1104	15.68	1.73092	0.49088	8004
	810901	5.5	70	0.0510	0.0563	0.1073	15.68	1.68194	0.53259	7972
	810902	4.5	90	0.0490	0.0583	0.1135	15.14	1.71854	0.48806	8653

	run	# fringes	Timebd	Hzi (cm)	Habd(cm)	Hmbd(cm)	GI(C/cm)	dt (C)	dc (wt%)	RaC
G = 13.4 CR = 3 Tbot = 19	813901	1.5	60	0.0792	0.0917	0.1250	14.63	1.82890	0.39950	9435
	813902	5.5	65	0.0573	0.0625	0.1083	13.30	1.44095	0.50203	7740
	814901	5.5	75	0.0542	0.0625	0.1104	13.72	1.51456	0.51241	8363
	815901	5.5	70	0.0573	0.0625	0.1115	13.72	1.52885	0.51392	8627
	815902	5.5	80	0.0542	0.0635	0.1229	12.19	1.49868	0.50256	11315
	816901	5.5	70	0.0563	0.0635	0.1135	13.72	1.55742	0.51837	9198
	816902	4.5	90	0.0542	0.0646	0.1167	10.97	1.28023	0.42432	8168
	817901	5.5	70	0.0531	0.0604	0.1125	13.30	1.49637	0.50517	8720
	817902	5.5	75	0.0531	0.0625	0.1198	12.54	1.50231	0.50384	10500
	820901	3.5	50	0.0844	0.0875	0.1271	12.19	1.54949	0.43978	10933
G = 25 CR = 2 Tbot = 22	820902	4.5	85	0.0375	0.0385	0.0885	14.63	1.29547	0.41162	3463
	821901	4.5	110	0.0448	0.0510	0.1052	15.68	1.64928	0.47253	6666
	907901	4.5	80	0.0313	0.0365	0.0875	14.63	1.28023	0.40699	3305
	907902	5.5	85	0.0313	0.0375	0.0896	14.63	1.31071	0.46026	4012
	910901	5.5	90	0.0417	0.0479	0.0979	15.68	1.53497	0.50408	5737
	910902	4.5	85	0.0406	0.0469	0.0958	16.26	1.55795	0.45907	4895
	911901	4.5	90	0.0406	0.0469	0.0979	14.16	1.38643	0.43114	4905
	911902	4.5	85	0.0375	0.0458	0.0958	15.14	1.45051	0.44091	4702
	912901	5.5	95	0.0375	0.0448	0.0938	16.26	1.52408	0.50065	5001
	913901	4.5	105	0.0396	0.0438	0.0990	14.63	1.44788	0.43533	5112
G = 25 CR = 2 Tbot = 21	913902	5.5	85	0.0458	0.0552	0.1021	18.29	1.86700	0.56488	7281
	914901	4.5	80	0.0521	0.0552	0.1000	17.56	1.75575	0.50055	6063
	917901	4.5	100	0.0333	0.0448	0.0948	16.26	1.54102	0.45392	4684
	917902	4.5	95	0.0427	0.0500	0.1000	15.68	1.56763	0.46225	5600
	918901	4.5	85	0.0365	0.0458	0.0927	15.68	1.45332	0.44366	4284
	920901	3.5	75	0.0375	0.0417	0.0833	17.56	1.46312	0.39746	2785
	920902	3.5	65	0.0385	0.0406	0.0875	15.68	1.37168	0.37839	3070
	921901	3.5	85	0.0344	0.0385	0.0875	14.16	1.23893	0.35529	2883
	921902	5.5	80	0.0344	0.0375	0.0875	15.14	1.32438	0.46358	3766
	924901	5.5	80	0.0344	0.0375	0.0865	15.68	1.35535	0.46887	3674
G = 25 CR = 2 Tbot = 20	925901	4.5	80	0.0385	0.0438	0.0896	16.88	1.51236	0.45191	3936
	925902	5.5	90	0.0396	0.0458	0.0979	17.56	1.71917	0.52892	6017
	926901	4.5	90	0.0406	0.0469	0.0969	17.56	1.70088	0.48026	5289
	926902	5.5	90	0.0385	0.0490	0.0990	16.26	1.60876	0.51633	6065
	927901	4.5	90	0.0406	0.0490	0.0938	18.29	1.71460	0.48888	4879
	927902	5.5	90	0.0406	0.0500	0.0979	17.56	1.71917	0.53616	6100
	928901	5.5	90	0.0406	0.0510	0.1000	18.29	1.82890	0.55330	6704
	928902	5.5	90	0.0458	0.0500	0.0979	16.88	1.65305	0.52580	5983
	1001901	4.5	85	0.0417	0.0479	0.0927	17.56	1.62772	0.47431	4579
	1001902	4.5	85	0.0417	0.0490	0.0958	15.68	1.50231	0.45370	4838
G = 25 CR = 2 Tbot = 19	1002901	4.5	70	0.0490	0.0531	0.0948	15.68	1.48598	0.45842	4732
	1002902	4.5	80	0.0510	0.0583	0.1021	18.29	1.86700	0.52200	6726
	1003901	4.5	80	0.0594	0.0573	0.1031	18.29	1.88605	0.52214	6935
	1003902	5.5	95	0.0531	0.0583	0.1042	18.29	1.90511	0.57458	7869
	1004901	5.5	90	0.0490	0.0552	0.1042	19.08	1.98794	0.58206	7970
	1004902	5.5	90	0.0531	0.0563	0.1063	19.95	2.11986	0.60285	8759
	1005901	5.5	95	0.0500	0.0563	0.1042	18.29	1.90511	0.57081	7817
	1005902	5.5	90	0.0531	0.0563	0.1052	18.29	1.92416	0.57283	8082
	1008901	5.5	80	0.0490	0.0542	0.1000	19.08	1.90842	0.57165	6926
	1008902	5.5	85	0.0510	0.0552	0.1031	19.95	2.05751	0.59418	7894

APPENDIX C
MEASUREMENT OF DENDRITE ARM SPACINGS

The primary dendrite arm spacing is used as a characteristic length in the permeability model suggested by Poirier. A microscope/solidification assembly was used to measure the primary dendrite arm spacings under conditions similar to those of the experiments of this study. In contrast to the earlier experiments, the optical depth of the cuvette was 2 mm, and heating and/or cooling was provided through the upper and lower side walls. Figure AC-1 illustrates the assembly. The controlling hardware and software was identical for all experiments. Sidewall thermocouples inserted into the fluid were used to measure the temperature gradient in the cuvette.

Cooling rates and temperature gradients were matched to a set of conditions from the earlier experiments and video was taken to measure the size of the dendrites. Figure AC-2 shows dendrites at the growth front. Figure AC-3 shows dendrites further within the two-phase region (nearer to the cold end). Notice the significant number of secondary arms and the non-uniformity in overall orientation. Measurements were taken using a digital image processor as discussed in Section 4. The measured primary arm spacings were within the range of 30-140 μm .

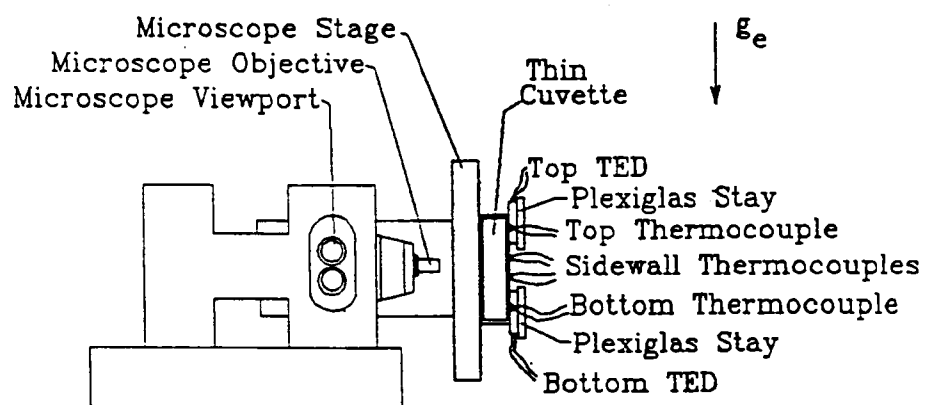
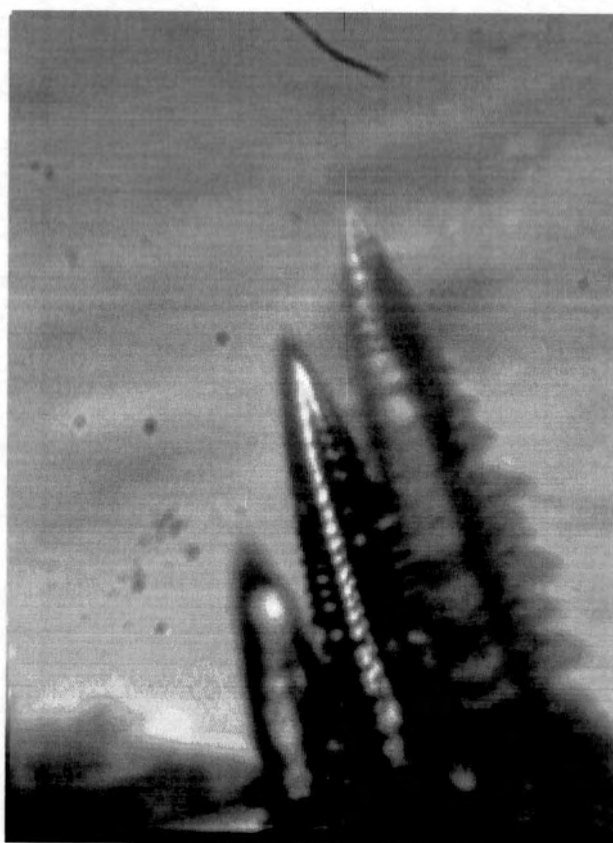


Figure AC.1 Microscope/Solidification Assembly Used to Measure Dendrite Arm Spacings.



120 μm

Figure AC.2 Dendrites at Solid-Liquid Interface.



120 μm

Figure AC.3 Dendrites Within Two-Phase Region.

VITA

John Andrew Hopkins Jr. was born to John and Brenda Hopkins on September 4, 1966 in Morristown, TN. He attended school there and graduated from Morristown Hamblen High School West in 1984. He enrolled at the University of Tennessee at Knoxville in the Fall of 1984. During his studies he worked as a NASA co-op engineer in the Propulsion Division at Marshall Space Flight Center. John and Theresa (Terri) Ann Mandzak were married in December of 1987. He received a Bachelor of Science Degree in Mechanical Engineering from the University of Tennessee in June 1988. During the summer of 1988 he worked as a staff engineer for the University of Tennessee Space Institute's Center for Laser Applications. He enrolled as a full time graduate research assistant working under the NASA sponsored Casting and Solidification Technology (CAST) program in the fall of 1988 and received a Master of Science degree with a major in Mechanical Engineering in August 1989. On June 26, 1991, John and Terri had their first child, Melinda Kay Hopkins. He continued his studies at the Center for Laser Applications in support of the CAST IML-1 space shuttle experiment and received his Doctor of Philosophy degree with a major in Mechanical Engineering in May 1992.