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Solidification of a Binary Solution (NH_4Cl+H_2O) on an Inclined Cooling Plate: A Parametric Study

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Abstract

A parametric study on the solidification behaviour of a binary solution ($NH_4Cl + H_2O$) on inclined cooling plate is presented numerically. During solidification of the solution, a shear layer is developed in the mush where a part of the dendrites is fragmented under shear flow and transported into the bulk solution. Accordingly, a slurry layer is found over the mush. The present solidification behaviour primarily depends on inclination angle of the plate and composition of the solution. In the present work, a mathematical model is developed where the free surface of solution is represented by the volume-of-fluid (VOF) method and the solidification behaviour is represented by a set of volume averaged single phase mass, momentum, energy , species conservation equations. The Stokes model is used to represent the solid velocity. The governing equations are solved based on the pressure-based finite volume method according to the SIMPLER algorithm using TDMA solver along with the enthalpy update scheme. The final simulation predicts the effect of process parameters on temperature, velocity, solid fraction and the species distributions.

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Keywords: solidification, inclined cooling plate, shear flow, fragmentation of dendrites, transport phenomena, parametric study

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Introduction

Nowadays, Semisolid Metal Forming (SSM) is an immerging casting technology that produces a wide range of defect-free components for automotive, aerospace, defence and structural applications. The technology involves casting of metallic alloys in presence of induced fluid flow/stirring (Spencer *et al.*, 1972; Joly and Mehrabian, 1976;

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Fleming, 1991). The strong fluid flow arrests the growth of dendrites by detaching them from the solid-liquid interface and carries them into the mould to form a semisolid slurry. The suspended dendrites in the slurry become rosette or globular particles after coarsening (Fleming *et al.*, 1991) in the slurry. This slurry offers less resistance to flow even at a high solid fraction and may be pushed easily into a die at high solid fraction. Finally, the technology offers cast products with uniform macrosegregation, a low porosity and increased strength and/or elongation at low forming efforts. However, this SSM is a developing technology which demands a detailed study under different process parameters towards commercialization of the technology. Hence, in the present work, a parametric study on the transport phenomena during solidification of a binary solution (NH_4Cl+H_2O) under shear flow is considered. It is mentioned here that the present transparent aqueous solution is widely used in experiments for direct observation of the transport phenomena during solidification. Since experimentation is very expensive, a numerical investigation is considered in this work preliminary.

Towards understanding of the solidification process in presence of shear flow, a literature review is performed. It is found that the solidification process under shear flow has been reported by many researchers such as Fleming *et al.*, (1991); Spencer *et al.*, (1972); Joly and Mehrabian., (1972) etc. However, only a few works reported on the solidification process of binary systems on an inclined cooling plate although it is a simple semisolid slurry forming technique where the liquid metal with a slight superheating is poured over the inclined cooling plate. During processing, the liquid melt falls quickly into the semisolid range by removing heat and consequently nucleates. The newly developed grains get sheared under the shear flow and carried out with the flow. Using the inclined cooling channel, Buncka *et al.* (2010) produced the aluminium slurry. Das *et al.* (2012) presented micro-structural evolution of A356 alloy during flow on a cooling slope. Kundu and Dutta (2012) performed a scale analysis of the solidification process of an aluminium alloy flowing down a cooling slope. Dhindaw *et al.* (2012) presented a numerical simulation on solidification of Al- alloy in a cooling slope. However, in the present work, the solidification of a transparent binary solution is considered. In this regards, Mehrara *et al.* (2008) performed a preliminary experimental study on solidification of the transparent succinonitrile-acetone on inclined cooling plate. Mohanty *et al.* (2012) reported investigation on the solidification process of an aqueous ammonium chloride solution on inclined cooling plate where the fragmentation of dendrites is ignored. In the present work, therefore, a parametric study is performed to predict the transport phenomena during solidification under shear flow in details.

Nomenclature

c	Concentration of soluté (wt % of NH_4Cl)
f	Mass fraction
P	Pressure (Pa)
T	Temperature (^{o}C)
t	Time (s)
$\overline{U}(u, v)$	Velocity Vector (m/s)
g	Acceleration Vector due to gravity
μ	Viscosity (Pa-s)
β_C	Coefficient of solutal expansion
β_T	Coefficient of thermal expansion
μ_l	Dynamic viscosity of liquid alloy
l	Liquid
n	iteration number
p	P^{th} cell
s	Solid

2. Description of the Physical Problem

The present work considers solidification behaviour of a metal analogous transparent binary solution (NH_4Cl+H_2O) under shear flow. The aqueous ammonium chloride solution is poured on an inclined cooling plate, which flows over the plate. While a two-dimensional geometry of the present problem is considered as shown in

Fig. 1, an adiabatic length is considered for flow stabilization. During processing, the solution is cooled at a constant temperature of -30°C. As heat is removed from the cooling plate, the solidification starts at surface of the cooling plate. Since the solidification of a binary solution occurs over a range of temperatures; hence, a mushy zone containing dendrites appears during the solidification process. It also involves the rejection of solute according to the binary phase diagram as shown in Fig. 2. As the liquid solution flows over the cooling plate due to the gravitational force, a shear layer is developed in the mush. The dendrites are fragmented under the shear flow and transported in the solution. It forms a slurry where the solid particles remain suspended in the liquid. In the present work, therefore, distributions of velocity, temperature, solid fraction and solute (NH_4Cl) during processing are studied under different process parameters. The thermo-physical properties of the NH_4Cl+H_2O solution and the system data used in simulation are given in table-1.

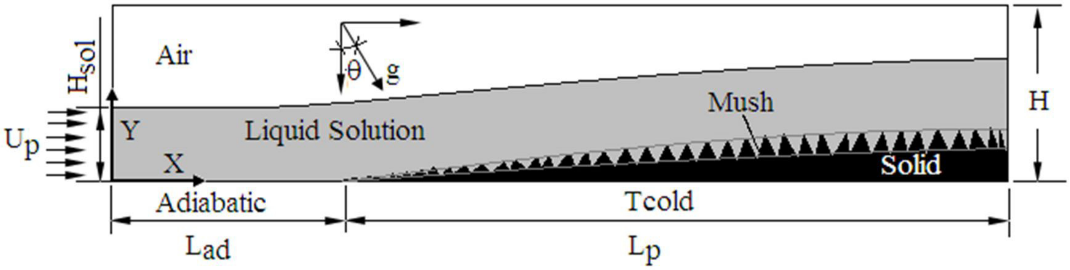

Fig. 1: A schematic of the physical system

. Table 1. The thermo-physical properties of NH_4Cl+H_2O and system data (Barman and Dutta, 2008)

Thermo-physical properties	**Value**
Specific heat (C_p)	3000 J/kg K
Thermal conductivity (k)	0.4 W/m K
Density (ρ)	1000 kg/m^3
Liquid viscosity (μ)	1.0×10^{-3}kg/m s
Species diffusion coefficient (D_l)	4.8×10^{-9} m^2/s
Latent heat of fusion (L_a)	3.0×10^{5} j/kg
Thermal expansion coefficient (β_T)	4.0×10^{-5} K^{-1}
Solutal expansion coefficient (β_C)	0.025
Eutectic temperature (T_e)	-15.4°C
Eutectic concentration (C_e) (wt% NH_4Cl)	19.7
Melting point of pure H_2O (T_m)	0°C
Equilibrium partition coefficient (k_p)	0.3
Permeability coefficient, K_o	5.56×10^{-11} (m^2)
System data	
Solution composition at inlet (C_{in}) (wt% NH_4Cl	8,13 and 18
Solution temperature at inlet (T_{in})	25°C
Bottom surface temperature ($Tcold$)	-30°C
Plate length (L)	300mm
Length of adiabatic section (L_{ad})	50mm
Height of the solution at inlet (H_{sol})	16mm

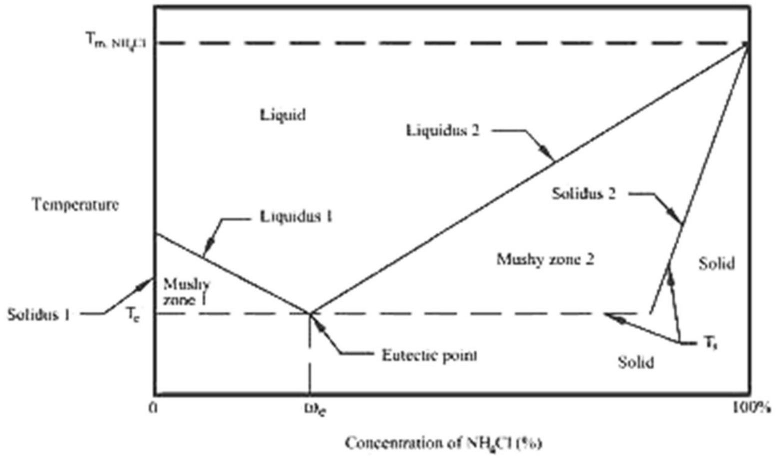

Fig. 2: A typical phase diagram of NH_4Cl+H_2O (ω_e =19.7% and T_e = -15.4°C) [10]

Mathematical Modelling

In the present work, the fixed-grid-single-domain approach is used for mathematical modelling of the process where the solution flows over an inclined cooling plate. In the model, the free surface between liquid solution and the surrounding air is tracked first using volume-of-fluid (VOF) method (Hirt et al., 1981). Then, the fluid flow is represented by the mass and momentum conservation equations where volume-average properties are considered in the entire domain (Chakraborty et al., 2001). The Stokes model is used to calculate the solid velocity in the domain (Vreeman et al., 2000). Then, the solidification process is accounted by coupling the energy and species equations (Kumar et al., 2005) with the flow equations. In the present work, the solidification shrinkage is neglected and the flow is considered incompressible. The corresponding governing equations are given below.

Conservation of Fluid Fraction Function:

$$\frac{\partial \alpha}{\partial t} + \nabla.\left(\overline{U}\alpha\right) = 0 \tag{1}$$

where α is a fluid fraction function. The α is equal to 1 in fluid region and equal to 0 in air.

Conservation of Mass:

$$\frac{\partial}{\partial t}(\rho) + \nabla.(\rho\overline{U}) = 0 \tag{2}$$

Conservation of Momentum:

$$\frac{\partial}{\partial t}(\rho\overline{U}) + \nabla.(\rho\overline{U}\,\overline{U}) = \nabla.(\mu_{eff}\nabla\overline{U}) - \nabla P - \frac{\mu_l}{K}\left(\overline{U} - \overline{U}_s\right) - \nabla.\left[\rho f_l f_s\left(\overline{U}_l - \overline{U}_s\right)\left(\overline{U}_l - \overline{U}_s\right)\right] + \rho\overline{g}\left[\beta_T\left(T - T_{ref}\right) + \beta_C\left(C_l - C_{ref}\right)\right] \tag{3}$$

Conservation of Energy:

$$\frac{\partial}{\partial t}(\rho C_p T) + \nabla.(\rho C_p \overline{U}T) = \nabla.\left(k\nabla T\right) - \frac{\partial}{\partial t}(\rho\Delta H) - \nabla.\left(\rho\overline{U_l}\Delta H\right) \tag{4}$$

Species Conservation:

$$\frac{\partial}{\partial t}(\rho C_l) + \nabla\cdot(\rho\overline{U}C_l) = \nabla\cdot(D^+\nabla C_l) + S_c \tag{5}$$

where $D^+ = \rho f_l D_l$ (5a)

$$S_c = -\frac{\partial}{\partial t}(\rho f_s C_l) - k_p C_l \frac{\partial}{\partial t}(\rho f_s) + \nabla.\left[\rho C_l f_s\left(1 - k_p\right)\overline{U}_s\right] \tag{5b}$$

The above continuum equations are valid over the entire domain, the specific nature of the five regions (air, liquid, solid, mush and slurry) are accounted through the volume average properties and the source terms. Each property (ϕ) in the entire computational domain is calculated as $\phi = \alpha\left(f_l\, \phi_{liquid} + f_s\, \phi_{solid}\right) + (1-\alpha)\phi_{air}$ and the superficial velocity is considered as $\overline{U} = f_l\, \overline{U}_l + f_s\, \overline{U}_s$.

Initial and Boundary Conditions

Initially, the domain contains air at the initial temperature T_{in}. Then, the aqueous solution with composition of C_{in} at initial temperature is poured on the inclined surface. The boundary conditions are as follows:

Left surface:

For $y \le H_{sol}$, $u = U_p$, $v = 0$, $\alpha = 1$, $T = T_{in}$ and $C_l = C_{in}$

For $y > H_{sol}$, $\frac{\partial u}{\partial x} = 0$, $v = 0$, $\alpha = 0$, $T = T_{in}$ and $C_l = 0$

Right surface: $\frac{\partial u}{\partial x} = 0$, $\frac{\partial v}{\partial x} = 0$, $\frac{\partial \alpha}{\partial x} = 0$, $\frac{\partial T}{\partial x} = 0$ and $\frac{\partial C_l}{\partial x} = 0$

Bottom surface: $u = 0$, $v = 0$, $\frac{\partial \alpha}{\partial y} = 0$ and $\frac{\partial C_l}{\partial y} = 0$

For $x \le L_{ad}$, $\frac{\partial T}{\partial y} = 0$

For $x > L_{ad}$, $T = T_{cold}$

Top surface: $u = v = 0$, $\alpha = 0$, $T = T_{in}$ and $\frac{\partial C_l}{\partial y} = 0$

4. Numerical Modelling

The governing equations (1-5) are coupled with the boundary conditions and solved with a pressure-based semi-implicit finite volume method according to the SIMPLER algorithm using TDMA solver (Patankar et al., 1920). To calculate the latent heat content ($\triangle H$) in the energy equation, the well known enthalpy update scheme, proposed by Brent *et al.*, is adopted. Equation (6) shows the equation for enthalpy update scheme. In the method, the latent heat content of each control volume is updated according to the temperature values predicted from the energy equation in previous iteration.

$$[\Delta H_p]_{n+1} = [\Delta H_p]_n + \frac{a_p}{a_p^0}\cdot \lambda \cdot \left\{[h_p]_n - c_p \cdot f^{-1}[\Delta H_p]_n\right\} \tag{6}$$

where a_p, a_p^0 are the coefficients of the discretized energy equation, λ is a relaxation factor and f^{-1} is the inverse of the latent heat function depending on systems (pure or binary system). For binary system, f^{-1} is given (Chakraborty et al., 2001) as

$$f^{-1}(\Delta H) = T_m - (T_m - T_L)\left(\frac{\Delta H}{L_a}\right)^{(k_p - 1)} \tag{7}$$

The liquidus temperature (T_L) is updated based on a linearised phase diagram of the solution (Barman *et al.*, 2008). Subsequently, the liquid fraction and solid fraction in the *p*th cell is calculated as

$$f_l = \frac{\Delta H}{L_a} \quad \text{where } 0 < \Delta H < L_a \tag{8a}$$

$$f_s = 1 - f_l \tag{8b}$$

During simulation, the convergence is declared when $\left|(\phi - \phi_{old}) / \phi_{max}\right| < 10^{-5}$ where ϕ stands for the solved variables at a grid point at the current iteration level, ϕ_{old} represents the corresponding value at the previous iteration level and ϕ_{max} is the maximum value of the variable at the current iteration level in the entire domain. Also, the work conducts a comprehensive grid-independence study. It is found that a 122 × 82 non-uniform grid is suitable for the present simulation and a time step of 0.1s offers a better convergence.

5. Results and Discussion

This work primarily presents a parametric studies on transport phenomena during solidification of a binary aqueous solution on an inclined cooling plate numerically. The numerical code developed on FORTRAN platform is validated by the authors in Mohanty *et al.* (2012). During processing, the liquid solution of 16mm thickness is poured continuously on the plate with a velocity of 0.1m/s. As the aqueous solution flows over the cooling plate, a variation of temperature in the solution layer occurs. Accordingly, the phases appeared in the computational domain are identified first. Fig.3 shows the corresponding air/liquid interface, liquid solution, slurry, mush and solidified layer at steady state condition when the aqueous solution containing 8 wt% of NH_4Cl is cooled on the cooling plate at an inclination angle of 70° with the horizontal plane. Subsequently, the work presents the distribution of velocity, temperature and solute in the solution under two key process parameters: inclination angle and solute composition.

Transport phenomena at different inclination angles

In this section, the effect of inclination angle (θ) on the transport phenomena is presented. Fig. 4 (a) shows the variation of liquid velocity along the y-direction at different inclination angels (30°, 50° and 70°) of the plate. It is evident from the Fig. 4(a) that the velocity in the mush increases with increasing angle of inclination, which results in corrsponding increase of the shear rate in the mush which is shown in Fig. 4(b). Increasing shear rate enhances the fragmentation of dendrites in the mush. The fragmented dendrites are lighter than the bulk solution and hence, they are transported into the slurry. These phenomena result in a wider slurry region at high angle of inclination. In Fig. 5 (a), the variation of temperature along the Y-direction is shown where a wider slurry layer is found at 70° angle of inclination. It is also noticed that an identical temperature profile is found in the solidified layer and a uniform temperature of about -1°C is found in the mush for all the cases. During processing, the fragmented dendrites are transported in the relatively hot liquid solution and the slurry layer forms. In the slurry layer, remelting of the suspended particles occurs and results a uniform temperature distribution in the slurry. This phase change process occurs at a temperature of about -1°C.

In Fig. 5(b), a variation of solute (NH_4Cl) fraction in the liquid solution along the Y-direction is presented. It is observed that the solute decreases in the solution with increasing angle of inclination. At high angle of inclination, the liquid velocity increases as found in Fig. 4(a) and it carries more rejected solute from mush to the slurry. The fact reduces the rejected solute in the mush. Subsequently, at low solute fraction, the solution rejection is also less during successive solidification according to the phase diagram (Fig. 2). Hence, the variation of solute in the liquid solution decreases with increasing angle of inclination.

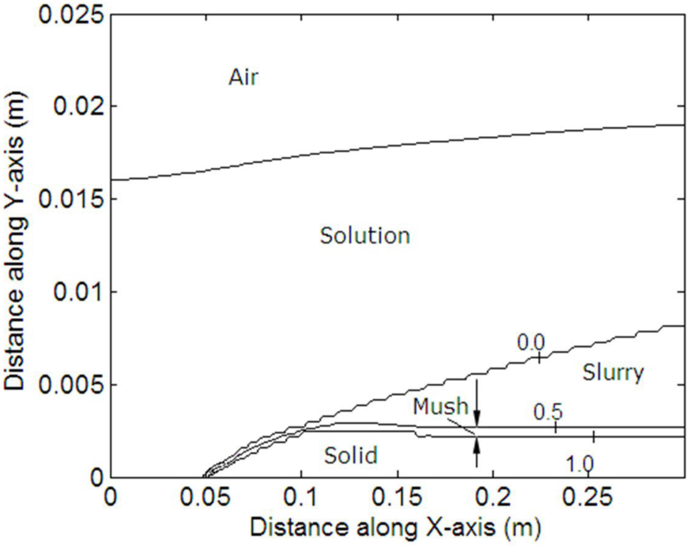

Fig.3: Various phases (air, solution, slurry, mush and solid) in the computational domain

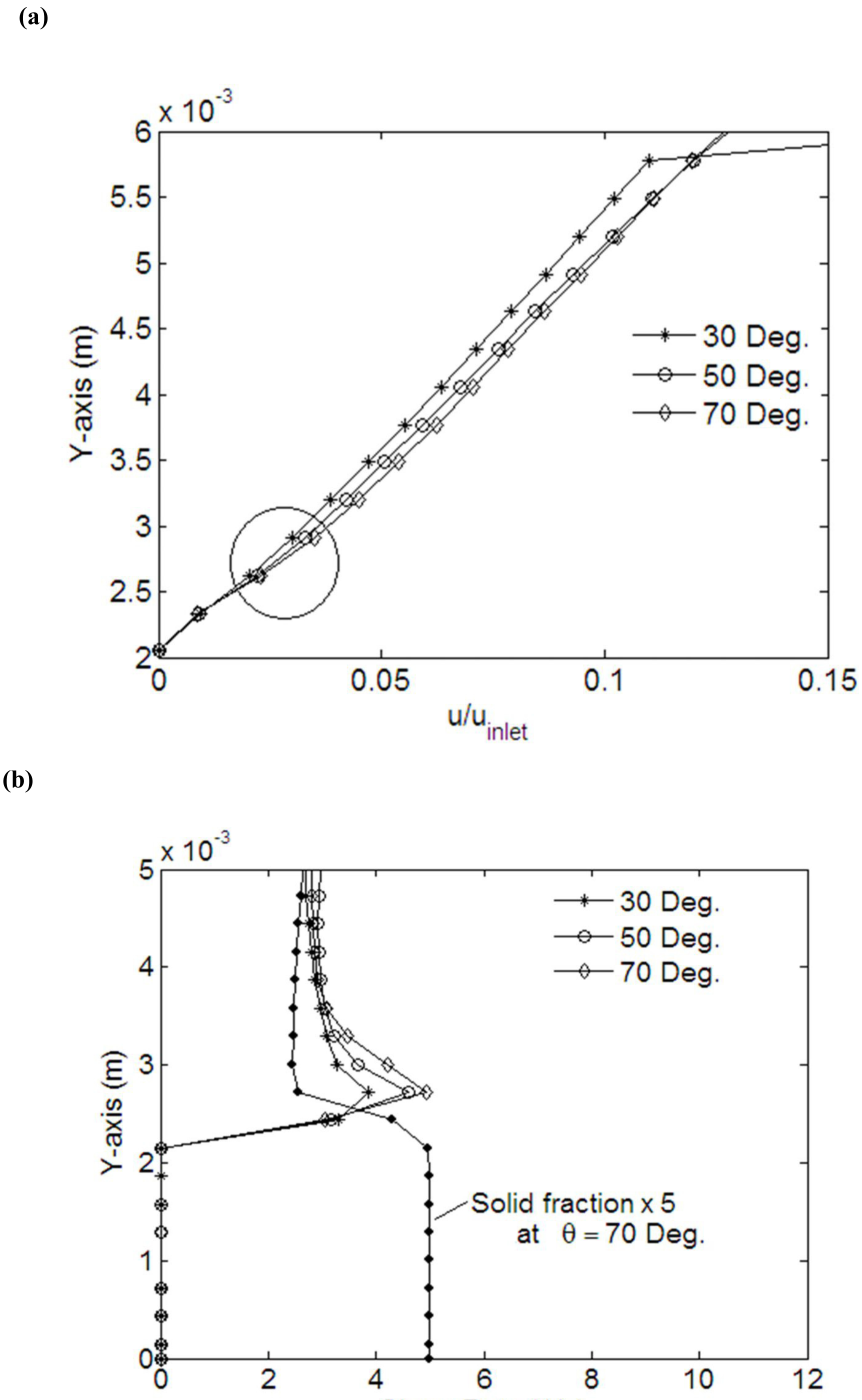

Fig.4: Variation of (a) liquid velocity and (b) shear rate along the layer height (in the Y-direction) at x = 0.25m

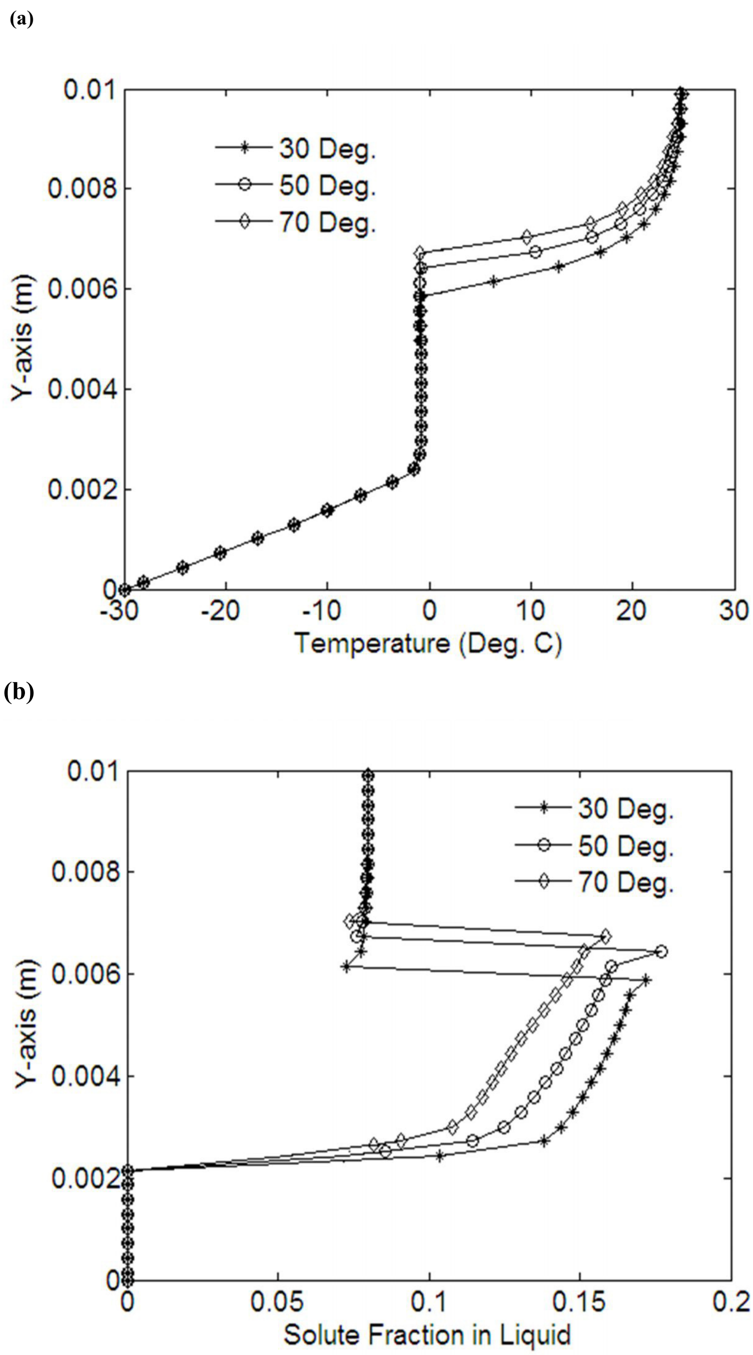

Fig.5: Variation of (a) temperature and (b) solute fraction along the layer height (in the Y-direction) at x = 0.25m

Transport phenomena at different initial compositions

The initial composition of the solution mainly affects the distribution of solid in the mush. Accordingly, in Fig. 6, the effect of initial composition on the fraction of solid during solidification of the binary solution is presented. The simulation is performed considering three different initial compositions (8, 13 and 18% NH_4Cl). It is apparent from the binary phase diagram (Fig. 2) that the mush zone decreases with increasing solute concentration in the solution. Accordingly, the height of the mush/slurry layer decreases as seen in the Fig.6.

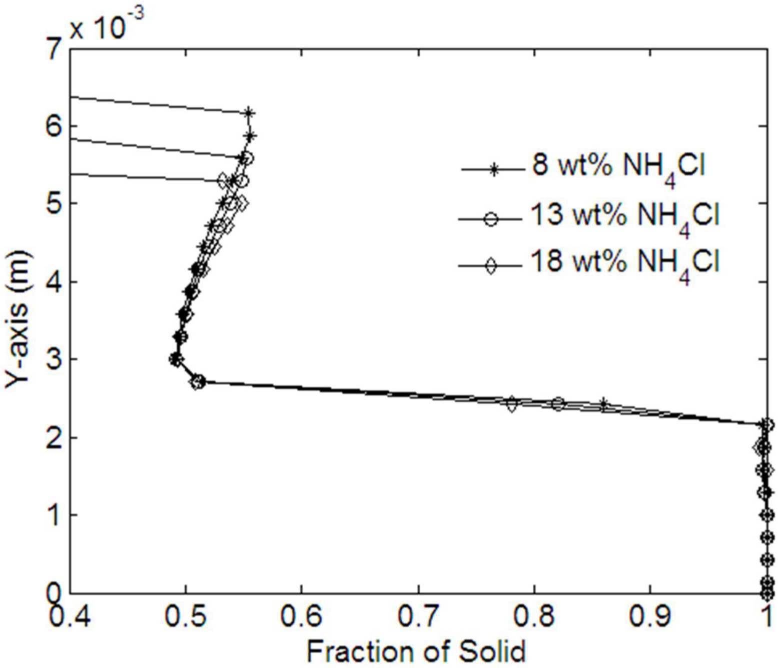

Fig. 6: Variation of solid fraction along the layer height (in the Y-direction) at x = 0.25m

5. **Conclusion**

This work presents a parametric studies on the transport phenomena during solidification of a binary solution (NH_4Cl+H_2O) on an inclined cooling plate. Two process parameters considered here are angle of inclination and the initial composition. The simulation revealed that the fragmentation of dendrites increases and rejected solute decreases in the slurry with increasing angle of inclination. With increasing initial composition, the size of the mush region decreases.

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