Double diffusive convection during freezing or melting in aqueous solution

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ABSTRACT

The double diffusive convection during the freezing or melting in aqueous solution was numerically studied at the Prandtl number \(Pr=13.4\), the Lewis number \(Le=100\), the buoyancy ratio \(N_0=0,1,5\) or 10 and the Rayleigh number \(Ra_0=10^5\) or \(10^7\). In the case of freezing, a thin boundary layer of concentration was formed near the moving boundary and the solute was accumulated near the bottom of the vessel. In the case of melting, one or two thermal convection roll cells were formed at \(N_0=0\) (\(Ra_0=10^5\) and \(10^7\)), multi-layered roll cells at \(N_0=1\) to 5 (\(Ra_0=10^7\)) and a concentration stratified layer at \(N_0\geq1\) (\(Ra_0=10^5\)) or at \(N_0=10\) (\(Ra_0=10^7\)). The melting of ice in a solution became strong for the lower part of moving boundary owing to the solutal convection near the ice.

KEYWORDS

energy storage, freezing, melting, double diffusive convection

INTRODUCTION

The cold energy storage system based on ice/water phase change is an innovative way of storing nighttime off-peak energy for daytime peak use. The industrial products of the system are commercially available. If the factory waste water instead of water is used for a thermal energy storage medium, the condensation of solutes is simultaneously carried out. In the case of freezing in aqueous solution, the solute concentrated near the moving boundary transfers downward owing to the solutal convection and large amount of solute may be accumulated near the bottom. In the case of melting of ice in solution, the solution is diluted near the moving boundary and the upward buoyancy occurs near the boundary. Consequently, a concentration stratified layer is formed from the bottom or from the top of the system, or a multi-layered convection may be formed in the system by lateral heating or cooling (Huppert and Turner 1978, 1980). Then, a curved boundary between ice and solution is formed and moves with freezing or melting.

The experimental or numerical studies of double diffusive natural convection during freezing or melting in aqueous solution was studied by many investigators (Beckermann and Viskanta 1988; Thompson and Szekely 1988; Nishimura et al. 1993). The mathematical modeling of freezing and melting in aqueous solution developed markedly as described in the reviews (Hu and Argyropoulos 1996; Mackerle 1999). However, few numerical calculations were reported for a multi-layered convection (Thompson and Szekely 1988), because the multi-layered convection occurs at high Rayleigh number. In this paper, the double diffusive natural convection during freezing or melting in aqueous solution as shown in Figure 1 was numerically studied with moving boundary between ice and solution by a finite element method at the Rayleigh number \(Ra_0=10^5\) and \(10^7\).
Aqueous solution

Adiabatic Heating (4 or 10°C) or Adiabatic Cooling (–10°C)

(a) Freezing

Adiabatic

Aq. soln

Ice

Heating (4 or 10°C)

(b) Melting

Figure 1 Schematic illustration of physical system.

NUMERICAL ANALYSIS

The model equations to describe the double diffusive natural convection consist of the stream function, the vorticity, the energy and the concentration equations in dimensionless forms as follows:

\[
\begin{align*}
\zeta &= \frac{\partial V}{\partial X} - \frac{\partial U}{\partial Y} = -\nabla^2 \psi
\end{align*}
\]

(1)

\[
\frac{\partial \zeta}{\partial \tau} + U \frac{\partial \zeta}{\partial X} + V \frac{\partial \zeta}{\partial Y} = PrRa \left( - \frac{\partial \theta}{\partial X} + N \frac{\partial C}{\partial X} \right) + Pr \nabla^2 \zeta
\]

(2)

\[
\frac{\partial \theta}{\partial \tau} + U \frac{\partial \theta}{\partial X} + V \frac{\partial \theta}{\partial Y} = \nabla^2 \theta
\]

(3)

\[
\frac{\partial C}{\partial \tau} + U \frac{\partial C}{\partial X} + V \frac{\partial C}{\partial Y} = \nabla^2 C / Le
\]

(4)

In ice,

\[
\frac{\partial \theta}{\partial \tau} = K_\kappa \nabla^2 \theta
\]

(5)

Dimensionless parameters \( Pr, Ra, N \) and \( Le, K_\kappa, K_\lambda \) and \( Ste \) are defined as follows:

\[
Pr = \frac{v}{\kappa_L}, \quad Ra = \frac{g \alpha (T_{\text{hot}} - T_{\text{cold}}) h^3}{\kappa_L v}, \quad N = \frac{\beta (C_{\text{max}} - C_{\text{min}})}{\alpha (T_{\text{hot}} - T_{\text{cold}})},
\]

\[
Le = \frac{K_\kappa}{D}, \quad K_\kappa = \frac{K_S}{\kappa_L}, \quad K_\lambda = \frac{\lambda_S}{\lambda_L}, \quad Ste = c_v(T_{\text{hot}} - T_{\text{cold}}) / \gamma
\]
Here, the dimensionless variables are defined as follows:

\[ X = \frac{x}{h}, \quad Y = \frac{y}{h}, \quad U = \frac{uh}{\kappa L}, \quad V = \frac{vh}{\kappa L}, \quad \tau = \frac{t}{\kappa L/h^2}, \quad \zeta = \frac{\omega h^2}{\kappa L}, \quad \theta = \frac{T - T_0}{T_{\text{hot}} - T_{\text{min}}}, \quad C = \frac{c - c_0}{c_{\text{max}} - c_{\text{min}}}. \]

where \( C = \) dimensionless concentration, \( c = \) concentration [kg/m\(^3\)], \( c_{\text{min}} = \) initial minimum concentration [kg/m\(^3\)], \( c_{\text{max}} = \) initial maximum concentration [kg/m\(^3\)], \( c_\rho = \) specific heat [J/kg K], \( \rho = \) density [kg/m\(^3\)], \( D = \) diffusion coefficient [m\(^2\)/s], \( g = \) acceleration due to gravity [m/s\(^2\)], \( H = \) dimensionless height of the system, \( h = \) height of the system [m], \( K_k = \) thermal diffusivity ratio, \( K_L = \) thermal conductivity ratio, \( Le = \) Lewis number, \( N = \) buoyancy ratio, \( Pr = \) Prandtl number, \( Ra = \) Rayleigh number, \( Ste = \) Stefan number, \( T = \) temperature [K], \( T_{\text{cold}} = \) minimum temperature (= -10°C) [K], \( T_{\text{hot}} = \) maximum temperature (= 10°C) [K], \( T_o = \) average temperature (=0°C) [K], \( t = \) time [s], \( U, V = \) dimensionless velocity, \( X, Y = \) dimensionless coordinate, \( \alpha = \) volumetric coefficient of thermal expansion [1/K], \( \beta = \) volumetric coefficient of expansion with concentration [m\(^3\)/kg], \( \gamma = \) latent heat [J/kg], \( \kappa = \) thermal diffusivity [m\(^2\)/s], \( \kappa_L = \) thermal diffusivity of liquid [m\(^2\)/s], \( \kappa_S = \) thermal diffusivity of ice [m\(^2\)/s], \( \lambda_L = \) thermal conductivity of liquid [W/m K], \( \lambda_S = \) thermal conductivity of ice [W/m K], \( \nu = \) kinematic viscosity [m\(^2\)/s], \( \rho = \) density [kg/m\(^3\)], \( \tau = \) dimensionless time, \( \psi = \) dimensionless stream-function (\( U = \partial \psi / \partial Y, \quad V = -\partial \psi / \partial X \)).

The volumetric coefficient of thermal expansion of water \( \alpha \) depends on temperature and the value is zero at about 4°C. The \( \alpha \) is proportional to \( Ra \) and inversely proportional to \( N \) according to the definition of \( Ra \) and \( N \). Then, the temperature dependence of \( Ra \) and \( N \) are expressed as follows:

\[ PrRa = PrRa_0(-1 + d_1 \theta + d_2 \theta^2) \] (6)

\[ N = N_0(-1 + d_1 \theta + d_2 \theta^2)^{-1} \] (7)

We assumed \( T_{\text{hot}} = 10\^\circ\text{C}(\theta = 0.5), \quad T_{\text{cold}} = -10\^\circ\text{C}(\theta = -0.5) \) and \( T_0 = 0\^\circ\text{C}(\theta = 0) \), and then got \( d_1 = 5.293 \) and \( d_2 = 1.268 \). The \( Ra_0 \) and \( N_0 \) are the values at 0°C(\( \theta = 0 \)). The molecular depression of freezing point against the concentration at the boundary were assumed zero. Therefore, the temperature at the boundary is 0°C(\( \theta = 0 \)). The amount of a solute concentrated near the boundary was corrected for the constant total amount in the entire liquid system during freezing or melting and then the error was within ±0.01%. Figure 2 shows the initial and boundary conditions of the system with the moving boundary. All boundaries of the square are solid walls. Equations (1) to (5) were solved by the finite element method under the conditions in Figure 2. The finite element mesh was horizontally expanded or compressed either with freezing or with melting. The boundary between ice and solution was obtained by
where $F = \text{dimensionless width of liquid layer}$, $\{1+(dF/dY)^2\} = \text{correction factor for the area of heat transfer}$. The same number of nodes was assumed for each phase but the nodes was horizontally expanded or compressed for each phase. The nodes were 61x121 (freezing at $Ra_0=10^5$), 61x81 (melting at $Ra_0=10^5$) and 81x161 ($Ra_0=10^7$). The calculations were carried out at $Pr=13.4$, $Le=100$, $Ra_0=10^5$ or $10^7$ and $N_0=0,1$ or 5.

RESULT AND DISCUSSION

**Freezing at $Ra_0=10^5$**

Figure 3 shows the instantaneous contours of stream function ($\psi$), temperature ($\theta$) and concentration ($C$) in the case of freezing at $T_{lw}=4°C$, $Ra_0=10^5$ and $N_0=0, 1$ or 5. At $N_0=0$, an anti-clockwise thermal convection was formed and a solute concentrated at the moving boundary flowed upward owing to the thermal convection. At $N_0=1$, an anti-clockwise thermal convection and a clockwise solutal convection coexisted in the liquid system. A solute concentrated at the moving boundary flowed downward and accumulated above the bottom. At $N_0=5$, the solutal convection became strong and a solute accumulated above the bottom similarly to the case of $N_0=1$.

Figure 4 shows the instantaneous contours in the case of freezing at $T_{lw}=10°C$, $Ra_0=10^5$ and $N_0=0, 1$ or 5. At $N_0=0$, a strong clockwise thermal convection above 4°C and a weak anti-clockwise thermal convection below 4°C (not seen in Figure 4) were formed and a solute concentrated near the moving boundary flowed out with convections as seen at $\tau=0.35$. At $N_0=1$, as the thermal convection above 4°C and the solutal convection have the same direction of rotation, one strong clockwise roll cell was formed and then the solute accumulated above the bottom. At $N_0=5$, the similar tendency to the case of $N_0=1$ was obtained. The freezing at the upper part of moving boundary was slow owing to the clockwise thermal convection above 4°C as seen at $\tau=0.35$ independently of buoyancy ratio.

![Figure 2 Initial and boundary conditions.](image-url)
Figure 3 Instantaneous contours at $Pr=6$, $Le=100$ and $Ra_0=10^5$.
(Freezing at $T_{lw}=4^\circ C$)

Figure 4 Instantaneous contours at $Pr=6$, $Le=100$ and $Ra_0=10^5$.
(Freezing at $T_{lw}=10^\circ C$)
Freezing at $Ra_0=10^7$

Figure 5 shows the instantaneous contours in the case of freezing for the adiabatic left wall of solution system at $Ra_0=10^7$ and $N_0=0, 1$ or 5. At $N_0=0$, convection does not exist and a concentrated solute diffuses from the vertical boundary. Therefore, the boundary layer of concentration kept thin due to the movement of the boundary. At $N_0=1$, the clockwise solutal convection existed and the solute was accumulated above the bottom. The vertical boundary moved to the left owing to the freezing independently of buoyancy ratio. Figure 6 shows the instantaneous contours in the case of freezing at $T_{lw}=4^\circ$C, $Ra_0=10^7$ and $N_0=0, 1$ or 5. In this case, thermal convection is anti-clockwise. At $N_0=0$, thermal convection exist and the solute concentrated near the boundary rotated with thermal convection. At $N_0=1$, the anti-clockwise thermal convection and a clockwise solutal convection (not seen in Figure 6) coexisted and the solute was accumulated above the bottom. At $N_0=5$, the solutal convection became stronger than that at $N_0=1$ and the solute was accumulated above the bottom similarly to the case of $N_0=1$.

Melting at $Ra_0=10^5$

Figure 7 shows the instantaneous contours in the case of melting at $T_{lw}=4^\circ$C, $Ra_0=10^5$ and $N_0=0, 1$ or 5. At $N_0=0$, an anti-clockwise thermal convection exists and the concentration distribution became homocentric. At $N_0=1$, anti-clockwise thermal and solutal convections existed and the solute formed a concentration stratified layer with time. At $N_0=5$, the solutal convection became strong and formed a stable concentration stratified layer.
Figure 7 Instantaneous contours at $Pr=6$, $Le=100$ and $Ra_0=10^5$. (Melting at $T_{lw}=4\,^\circ C$)

Figure 8 Instantaneous contours at $Pr=6$, $Le=100$ and $Ra_0=10^5$. (Melting at $T_{lw}=10\,^\circ C$)

Figure 8 shows the instantaneous contours in the case of melting at $T_{lw}=10\,^\circ C$ and $Ra_0=10^5$. At $N_0=0$, a strong clockwise thermal convection above $4\,^\circ C$ exists near the heating wall and a weak anti-clockwise thermal convection below $4\,^\circ C$ exists near the lower part of the moving boundary. The strong convection above $4\,^\circ C$ made a homocentric concentration distribution. At $N_0=1$, a clockwise thermal convection near the heating wall and a solutal convection near the moving boundary coexisted first but the solute formed a concentration stratified layer with time. Consequently, two convections became weak with time. At $N_0=5$, the solutal convection became strong and a stable concentration stratified layer was formed. The melting of ice in solution at $T_{lw}=10\,^\circ C$ was strong for the lower part of moving boundary owing to the anti-clockwise solutal convection near the ice, but the melting of ice in water was strong near the top surface due to the clockwise thermal convection above $4\,^\circ C$.

**Melting at $Ra_0=10^7$**

Figure 9 shows the instantaneous contours in the case of melting at $T_{lw}=10\,^\circ C$ and $Ra_0=10^7$. At $N_0=0$, a clockwise thermal convection above $4\,^\circ C$ exists near the heating wall and especially below the top. Moreover, an anti-clockwise thermal convection below $4\,^\circ C$ exists near the lower part of the moving boundary. The strong clockwise convection melted the upper part of moving boundary. The two convections made homocentric concentration distributions. At $N_0=1$ and 2, clockwise thermal convection above $4\,^\circ C$ exists near the heating wall, and anti-clockwise convection due to both solutal buoyancy and thermal buoyancy below $4\,^\circ C$ exists near the moving boundary. Consequently, multi-layered convection was formed and a thermal and a solutal convections coexisted in a layer. At $N_0=5$, the thermal and solutal convections moved downward in a concentration stratified layer. At $N_0=10$, a concentration stratified layer was formed in the entire system. At $N_0=1$ to 5, strong convections accelerated the melting of ice and then a curved moving boundary was formed. The melting of ice in solution was strong for the lower part of moving boundary similarly to the case of $Ra_0=10^5$. 
Figure 9 Instantaneous contours at $Pr=6$, $Le=100$ and $Ra_0=10^7$. (Melting at $T_{tw}=10^0\text{C}$)
Shape of the boundary between phases

Figure 10 shows the shape of the boundary between phases for the melting at $T_{lw}=10^\circ C$ and $Ra_0=10^5$. At $N_0=0$, a strong clockwise thermal convection above $4^\circ C$ near the left hand wall melted the upper part of moving boundary. An weak anti-clockwise thermal convection below $4^\circ C$ melted the lower part of the moving boundary. At $N_0=1$ and 5, an anti-clockwise convection due to both solutal buoyancy and thermal buoyancy below $4^\circ C$ melted the lower part of the moving boundary. Figure 11 shows the shape of the boundary between phases for the melting at $T_{lw}=10^\circ C$ and $Ra_0=10^7$. At $N_0=0$, a strong clockwise thermal convection above $4^\circ C$ near the left hand wall melted the upper part of moving boundary similarly to the case at $Ra_0=10^5$. An anti-clockwise thermal convection below $4^\circ C$ melted the lower part of the moving boundary. At $N_0=1$ to 5, an anti-clockwise thermal convection below $4^\circ C$ melted especially a part of the moving boundary near the bottom. Unlike the case of $Ra_0=10^5$, the curved boundary was formed by multi-layered convection.

**CONCLUSIONS**

The double diffusive convection during the freezing or melting in aqueous solution was numerically studied at $Pr=13.4$, $Le=100$, $Ra_0=10^5$ or $10^7$ and various buoyancy ratios. In the case of freezing, a thin boundary layer of concentration was formed near the moving boundary and the solute was accumulated...
by the convection due to the solutal buoyancy near the bottom. In the case of melting, one or two thermal convection roll cells were formed at $N_0=0$, a multi-layered cells at $N_0=1$ to 5 and $Ra_0=10^7$ and a concentration stratified layer at high buoyancy ratio. The melting of ice in solution was strong for the lower part of moving boundary owing to the solutal convection near the ice, but the melting of ice in water was strong near the top surface due to the thermal convection above 4 °C.

REFERENCES