MASS TRANSFER ACROSS AN INTERFACE BETWEEN LAYERS IN A DOUBLE DIFFUSIVE NATURAL CONVECTION

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When a two-layer system which consists of a solvent (upper layer) and a solution (lower layer) is heated from a vertical side wall and cooled from the opposing side wall, convection starts in each layer. A Galerkin finite-element method was employed for the numerical analyses of this two-layer convection. Computations were carried out for the Prandtl number $Pr = 6$, the aspect ratio $A = 4$, the Rayleigh number $Ra = 10^4$, the buoyancy ratio $N = 2$ to 10 and the Lewis number $Le = 10$ to 100. The concentration in the upper layer increased linearly with time through the transfer of the solute across the interface between the layers. In conclusion, the flux of a solute across the interface was independent of the concentration difference between the layers, but dependent on the diffusion coefficient.

Introduction

When a solution having a concentration gradient along the gravitational direction is heated from a vertical wall and cooled from the opposing vertical wall, multi-layered convection separated by almost-horizontal sharp interfaces is observed (Chen *et al.*, 1971). The simplest model for this phenomenon is a two-layer convection (Bergman and Ungun, 1988; Kamakura and Ozoe, 1991) which consists of a solvent (upper layer) and a solution (lower layer) as shown in Fig. 1. When the two-layer system is heated from one side and cooled from the opposite side, convection starts in each layer. Generally a sharp horizontal interface is formed between two flows in opposite directions. The flow in the layer is mainly caused by the temperature difference. Two-layer convection is hydrodynamically stable due to the concentration difference between the two layers. This difference gradually becomes small with the elapse of time because mass transfer is caused by the diffusion of a solute across the interface. The mass transfer across the interface causes concentration distribution in the outer region in each layer by convection (Kamakura and Ozoe, 1995). The solute in the outer region in the upper half layer transfers to the core by both diffusion and convection, and consequently the concentration at the core becomes uniform (Kamakura and Ozoe, 1995). Therefore, the mass flux of a solute across the interface can be approximately measured from the increment of concentration at a point in the core. The transfer of a solute across the interface is important for the studies

of both the stability of multi-layered convection and the structure of the interface.

Periodic oscillation of concentration at the core in the layer was found at $Ra = 10^5$ and 10^6 in the numerical simulation of two-layer convection (Kamakura and Ozoe, 1994). However, at $Ra = 10^4$ the concentration at the outer region was slightly distributed but that at the core was uniform, and the concentration at the core increased linearly with time. Therefore the present calculation was carried out for $Ra = 10^4$. The mechanism of mass transfer of a solute across the interface is expected to hold, even at high Rayleigh numbers, in spite of the periodic oscillations of concentration, because the oscillation occurs in a convection layer, but not at the interface. Computations were carried out for $Pr = 6$, $A = 4$, $Ra = 10^4$, $N = 2, 3, 5$ or 10 and $Le = 10, 20, 30, 50$ or 100. These correspond approximately to the previous experimental papers, except Rayleigh number (Kamakura and Ozoe, 1991). For either large concentration difference (large buoyancy ratio) or small Rayleigh number, the intermediate concentration stratified layer was formed between two convection layers (Kamakura and Ozoe, 1995). For either small concentration difference or large Rayleigh number, a single sharp interface was formed. As this work was carried out at $Ra = 10^4$, the interface has an intermediate layer with concentration gradient.

1. Numerical Analysis

1.1 Mathematical model

The basic equations to describe the double-diffusive

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Fig. 1 Schematic illustration of two-layer convection

Fig. 2 Boundary conditions

natural convection consist of the stream-function equation, the vorticity equation, the energy equation and the concentration equation as follows in dimensionless forms (Kamakura and Ozoe, 1993):

$$\zeta = \frac{\partial V}{\partial X} - \frac{\partial U}{\partial Y} = -\nabla^2 \psi \quad (1)$$

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

$$\frac{\partial \theta}{\partial \tau} + U \frac{\partial \theta}{\partial X} + V \frac{\partial \theta}{\partial Y} = \nabla^2 \theta \quad (3)$$

$$\frac{\partial C}{\partial \tau} + U \frac{\partial C}{\partial X} + V \frac{\partial C}{\partial Y} = \nabla^2 C / Le \quad (4)$$

Dimensionless parameters Pr , Ra , Le , N and A are defined as follows:

Fig. 3 Finite element mesh with 1691 nodes and 3168 elements

$$\left. \begin{aligned} Pr &= \frac{\nu}{k}, Ra = \frac{g a \Delta T_{\max} H^3}{\kappa \nu}, Le = \frac{\kappa}{D} \\ N &= \frac{\beta \Delta C_{\max}}{a \Delta T_{\max}}, A = \frac{2H}{B} \end{aligned} \right\} \quad (5)$$

Here, the dimensionless variables are defined as follows.

$$\left. \begin{aligned} X &= x/H, Y = y/H, U = uH/\kappa \\ V &= vH/\kappa, \tau = t\kappa/H^2, \zeta = \omega H^2/\kappa \\ \theta &= (T - T_0)/\Delta T_{\max}, C = (c - c_0)/\Delta C_{\max} \end{aligned} \right\} \quad (6)$$

1.2 Boundary conditions

The following boundary conditions in dimensionless form are illustrated in Fig. 2.

$$\text{at } Y=0: \quad \frac{\partial U}{\partial Y}=0, V=0, \quad \frac{\partial \theta}{\partial Y}=0, \quad \frac{\partial C}{\partial Y}=0$$

$$\text{at } Y=2: \quad U=V=0, \quad \frac{\partial \theta}{\partial Y}=0, \quad \frac{\partial C}{\partial Y}=0$$

$$\text{at } X=0: \quad U=V=0, \quad \theta=0.5, \quad \frac{\partial C}{\partial X}=0$$

$$\text{at } X=2/A: \quad U=V=0, \quad \theta=-0.5, \quad \frac{\partial C}{\partial X}=0$$

The stream function was assumed to be zero on the boundary. The vorticity on the wall was computed as follows:

$$\zeta = -\frac{\partial^2 \Psi}{\partial n^2} = -\frac{3\Psi_1}{\Delta n^2} - \frac{\zeta_1}{2} \quad (7)$$

Here subscript 1 denotes one grid space away from the wall. The initial concentration in the system is +0.5 in the lower layer and -0.5 in the upper layer.

1.3 Computation

The equations presented above were solved by a Galerkin finite-element method. The rectangular cross

Fig. 4 Vertical profiles of dimensionless concentration at $Pr = 6$, $A = 4$, $Ra = 10^4$, $Le = 50$, $\tau = 1.6$ and $X = 0.25$ (vertical central plane)

section of the two-layer system was divided into a number of triangles, as shown in Fig. 3. The mesh is symmetrical. The calculation algorithm of the finite-element method is the same as that used in our previous paper (Kamakura and Ozoe, 1993). This mesh is finer (19×89 nodes, $Ra = 10^4$) than that (19×69 nodes, $Ra = 10^5$) used in a previous paper (Kamakura and Ozoe, 1995). The time step $\Delta\tau$ for the numerical simulation was 0.000004.

2. Results

2.1 Vertical profile of concentration

The natural convection in a two-layer system was studied from the mass transfer perspective. When convection exists in each layer, the concentration in each layer becomes almost uniform and the interface between two convection layers keeps a steep concentration gradient. Mass transfer is caused by the diffusion of a solute across the interface, and the solute is transferred by both diffusion and convection from the outer region toward the core region, for example, in an upper layer. The numerical simulation was carried out for two-layer convection. Figure 4 shows the vertical profiles of dimensionless concentration at a center ($X = 0.25$) of the enclosure. In the case of $N = 3$ the concentration distribution in each layer is not flat, especially near the bottom or free top surface, reflecting the combined effect of convection and diffusion of a solute. However, the concentration at the core region was flat. In the case of $N = 10$ the concentration distribution became flatter than that of $N = 3$. The concentration distribution

Fig. 5 Typical instantaneous contours of stream function (upper) and concentration (lower) at $Pr = 6$, $A = 4$, $Ra = 10^4$, $N = 5$ and $Le = 30$. Contour lines of stream function were drawn by dividing the instantaneous maximum difference into 10 equal sections and those of concentration by dividing the initial maximum difference into 20 equal sections (This is also the case in the contours of Fig. 6)

linearly changed in the intermediate layer. The concentration gradient between two roll cells was steep for $N = 10$, and very steep for $N = 3$.

2.2 Computed contour maps

Figure 5 shows instantaneous contours of stream function and concentration. The contours of stream function typically consist of two roll cells. The concentration distributions are roughly linear in the intermediate layer, and similar to the case of Fig. 4. The mass transfer of a solute is carried out by diffusion across the intermediate layer. Figure 6 shows instantaneous contours of concentration for two extreme cases studied in the present study. Upper contours show the steepest distribution of concentration at the intermediate layer and lower contours show mostly slight distribution of concentration. The intermediate-layer thickness or the gradient will control the upward flux of salt as described in the succeeding sections.

2.3 Variation of concentration gradient with time

The mass flux is considered by Fick's law of diffusion to be proportional to the gradient (dC/dY) of concentration at the interface. As the concentration distribution of the intermediate layer was roughly linear, the gradient, dC/dY , can be determined easily. Figure 7 shows transient concentration gradient at the intermediate layer ($X = 0.25$). The concentration gradient at the interface gradually approaches to the limiting value with time and the limiting value increases with the Lewis number. If the gradient is constant with time, the rate of mass trans-

Fig. 6 Instantaneous contours of concentration at $Pr = 6$, $A = 4$ and $Ra = 10^4$. Upper: $N = 2$ and $Le = 100$, lower: $N = 10$ and $Le = 10$

Fig. 7 Variation of concentration gradient with time at $Pr = 6$, $A = 4$, $Ra = 10^4$ and $N = 10$

fer across the intermediate layer may be expected to be constant and independent of concentration difference.

2.4 Concentration variation with time

Figure 8 shows the concentration variation with time at $(X, Y) = (0.25, 0.35)$ in an upper layer. The concentration always increased linearly with time. It appears that the

Fig. 8 Variation of dimensionless concentration in upper layer with dimensionless time at $Pr = 6$, $A = 4$, $Ra = 10^4$ and $N = 10$ and $(X, Y) = (0.25, 0.35)$

Fig. 9 Variation of dimensional concentration in upper layer with dimensional time at $Pr = 6$, $A = 4$, $Ra = 10^4$. $\circ$: $N = 2$, $\bullet$: $N = 3$, $\square$: $N = 5$, $\blacksquare$: $N = 10$

mass flux of the solute across the intermediate layer is constant, and does not depend on the concentration difference. The dimensional concentration can be given by the next equation.

$$c = \frac{\alpha \Delta T_{\max}}{\beta} N (C + 0.5) \quad (8)$$

Here, it was assumed that $c_{\min}$ is zero because the initial upper layer is solvent. **Figure 9** shows a plot of the dimensional concentration (Eq. (8)) versus the dimensional time $(\tau H^2 / \kappa)$. We can see that the plots have equivalent slope for each Lewis number, even for different buoyancy ratios. The mass flux is consequently determined by the Lewis

Fig. 10 Relation between $dC/d\tau$ and Le at $Pr = 6$, $A = 4$ and $Ra = 10^4$

number, not by the buoyancy ratio. These characteristics have been observed experimentally in the previous papers (Kamakura, 1979; Kamakura and Ozoe, 1991). Agreement of these characteristics between the calculated and experimental results supports the present numerical computation.

2.5 Dependence of flux on diffusion coefficient

As the concentration at $(X, Y) = (0.25, 0.35)$ in an upper layer increases linearly with time, the rate $dC/d\tau$ which is proportional to the flux of a solute can be calculated from the slope of the plot. Figure 10 shows the logarithmic plot of $dC/d\tau$ versus Lewis number. From the slope and the intercept at $\log(Le) = 0$ the following relations were obtained:

$$dC/d\tau = (2.78/N + 0.28) Le^a \quad (9)$$

$$\text{At } N = 2, \quad a = -0.65.$$

$$\text{At } N = 3, \quad a = -0.64.$$

$$\text{At } N = 5, \quad a = -0.61.$$

$$\text{At } N = 10, \quad a = -0.70.$$

As the Lewis number varies inversely with the diffusion coefficient, the flux of a solute across the interface is proportional to the diffusion coefficient to the power 0.61 ~ 0.70. The power 0.70 at $N = 10$ is larger than the other. This may be due to the large intermediate concentration stratified layer for high buoyancy ratio. In a previous paper (Kamakura, 1979), the flux obtained in the experiment was proportional to the diffusion coefficient to the power 0.5. This difference may be due to the large difference between Rayleigh numbers.

Conclusions

Mass transfer across the interface in a two-layer system was studied by numerical simulation. The concen-

tration in the upper layer increased linearly with time by the transfer of the solute across the interface. The flux of the solute across the interface was independent of the concentration difference between layers, but dependent on the diffusion coefficient. The concentration gradient became constant with time, and then the thickness of the intermediate layer between convection layers became small.

Nomenclature

A	= aspect ratio = $2H/B$	[-]
B	= horizontal width of solution layer	[m]
C	= dimensionless concentration = $(c - c_0)/\Delta c_{\max}$	[-]
c	= concentration	[kg/m ³]
$c_{\max}$	= initial maximum concentration	[kg/m ³]
	(initial concentration of a lower layer)	[kg/m ³]
$c_{\min}$	= initial minimum concentration	[kg/m ³]
	(initial concentration of an upper layer)	[kg/m ³]
c_0	= $(c_{\max} + c_{\min})/2$	[kg/m ³]
D	= diffusion coefficient	[m ² /s]
g	= acceleration due to gravity	[m/s ²]
H	= 1/2-height of solution layer in apparatus	[m]
Le	= Lewis number = κ/D	[-]
N	= buoyancy ratio = $\beta\Delta c_{\max}/(\alpha\Delta T_{\max})$	[-]
n	= dimensionless coordinate normal to boundary	[-]
Pr	= Prandtl number = ν/κ	[-]
Ra	= Rayleigh number = $g\alpha\Delta T_{\max}H^3/(\kappa\nu)$	[-]
T	= temperature	[K]
T_{cold}	= temperature on cold wall	[K]
T_{hot}	= temperature on hot wall	[K]
T_0	= $(T_{\text{hot}} + T_{\text{cold}})/2$	[K]
U	= dimensionless velocity in X-coordinate	[-]
V	= dimensionless velocity in Y-coordinate	[-]
X	= dimensionless coordinate in horizontal direction	[-]
Y	= dimensionless coordinate in vertical direction	[-]
α	= volumetric coefficient of thermal expansion	[1/K]
β	= volumetric coefficient of expansion with concentration	[m ³ /kg]
$\Delta c_{\max}$	= $c_{\max} - c_{\min}$	[kg/m ³]
Δn	= dimensionless distance between wall and next node	[-]
$\Delta T_{\max}$	= $T_{\text{hot}} - T_{\text{cold}}$	[K]
$\Delta\tau$	= dimensionless time step	[-]
ζ	= dimensionless vorticity	[-]
θ	= dimensionless temperature = $(T - T_0)/\Delta T_{\max}$	[-]
κ	= thermal diffusivity	[m ² /s]
ν	= kinematic viscosity	[m ² /s]
τ	= dimensionless time	[-]
Ψ	= dimensionless stream function	[-]
	($U = \partial\Psi/\partial Y$, $V = -\partial\Psi/\partial X$)	[-]

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