

The Effects of Stable Thermal Stratification on the Onset of Double Diffusive Convection

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이중 확산 대류의 발생에서 열 안정화의 효과

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ABSTRACT

The time of the onset of double-diffusive convection in time-dependent, nonlinear concentration fields is investigated theoretically and experimentally. The initially quiescent horizontal fluid layer with a uniform temperature gradient experiences sudden concentration change from below, but its stable thermal stratification is to affect concentration effects which invoke convective motion. The related stability analysis, including Soret effect, is conducted on the basis of the propagation theory. The resulting critical time to mark the onset of regular cells are obtained as a function of the thermal Rayleigh number, the solute Rayleigh number, and the Soret effect coefficient. For a certain value of the Soret effect coefficient, the stable thermal gradient promote double-diffusive convective motion onset of convective instability in an initially quiescent,

Key Words : double diffusion, stability analysis, Soret effect, propagation theory

I. INTRODUCTION

Buoyancy-driven convection in double-diffusive systems has been studied extensively in connection with wide science and engineering situations such as oceanography and crystal growth processing,

solar ponds and natural gas storage tanks¹⁻⁴⁾. But the inherent complexity in practical systems makes it very difficult to predict the stability criteria by which the effect of natural convection is determined in the various systems. This comes from the fact that the solute concentration and temperature profiles are nonlinear and time-dependent.

When an initially motionless, stable thermal-stratified fluid layer is placed between two horizontal plates with its concentration fields change suddenly, natural convection will set in at a certain time, depending on both the thermal Rayleigh

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number and the solute Rayleigh number⁵⁾. Therefore, it becomes an important problem to predict the critical time to mark the onset of convective motion. For this purpose, several theoretical models have been used in deep-pool systems of high Rayleigh numbers: the amplification theory⁶⁾, energy method⁷⁾, stochastic model⁸⁾, and propagation theory⁹⁾. Even though they are all good models, the present double-diffusive convection has been analyzed only by the amplification theory. The amplification theory has been quite popular, but it involves difficulties in deciding the initial conditions and also choosing the growth factor to determine the onset time. Comparing with other methods, the energy method predicts the onset time of buoyancy-driven convection as lower bound. And the stochastic model involves some arbitrariness. But the propagation theory which we have developed, decides deterministically the criteria to mark the onset time by using the concentration or thermal penetration depth as a length-scaling factor and transforming the linearized perturbation equations. Predicted values resulting from the principle of exchange of stabilities have been consistent with most of experimental data in systems of laminar forced convection^{10),11)} and also fluid-saturated porous layers¹²⁾.

Most of analysis on double diffusive phenomena ignored the Soret effect^{13),14)}. In some case, however, Soret effect plays important roles in double diffusion characteristics, even if the temperature gradients are not quite large^{15),16)}. In present study, the stability analysis based on the propagation theory will be extended to the onset of double-diffusive convection incorporating Soret effect, and electrochemical experiments will be conducted to support the theoretical analysis.

II. THEORETICAL ANALYSIS

2.1. Mathematical Formulation

The problem considered here is a horizontal fluid layer confined between two rigid boundaries separated by a distance "d", as shown in Fig. 1. The fluid layer is initially quiescent at a constant concentration C_0 and stably stratified by a uniform temperature gradient. At time $t=0$ the concentration of lower boundary is reduced and kept constant. For large concentration difference systems natural convection will set in at a certain time due to buoyancy forces. Under this condition the density variation of fluid is assumed to follow the usual equation of state⁵⁾.

$$\rho = \rho_0 [1 - \beta(T - T_0) + \gamma(C - C_0)] \quad (1)$$

where ρ , T , C , β and γ represent the fluid density, the temperature, the concentration, thermal expansion coefficient and solutal expansion coefficient, respectively. The subscript "0" denotes the reference state.

The phenomenological equation relating heat (J_Q) and solutal diffusion flux (J_C) to the temperature and concentration can be found as^{15),16)}

$$J_Q = -k \nabla T - TC \left(\frac{\partial \mu}{\partial C} \right) D \nabla C \quad (2)$$

$$J_C = -\alpha_s [\nabla C - S_T C C \nabla T] \quad (3)$$

where k , μ , D , α_s , S_T and C are thermal conductivity, chemical potential, Dufour

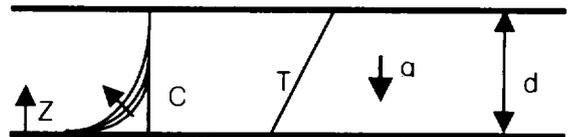


Fig. 1. Schematic diagram of system.

coefficient, mass diffusivity, Soret coefficient and solvent concentration respectively. Since from the Onsager's reciprocal rule $D = \alpha_s S_T$, the Dufour coefficient is many order of magnitude smaller than Soret coefficient in liquid. Therefore the Dufour effect is normally neglected in liquid.

The important parameters to characterize the onset of motion in the present system are the solutal Rayleigh number R_s , the thermal Rayleigh number R_a , the Schmidt number Sc , Lewis number Le and Soret effect coefficient defined by

$$R_s = \frac{g\beta\Delta T d^3}{\alpha\nu}, \quad R_s = \frac{g\gamma\Delta C d^3}{\alpha_s\nu}, \quad Sc = \frac{\nu}{\alpha_s}, \quad Le = \frac{\alpha}{\alpha_s},$$

$$\epsilon = S_T \overline{CC} Le \frac{\gamma}{\beta}$$

where g , α , ν , ΔT , ΔC , and \overline{C} denote the gravitational acceleration, the thermal diffusivity, the kinematic viscosity, the temperature difference, concentration difference, and mean concentration of layer. Under the linear stability theory, the nondimensionalized conservation equations incorporating the Soret effect are constituted as follows¹³⁾⁻¹⁶⁾:

$$\left(\frac{1}{Sc} \frac{\partial}{\partial \tau} - \nabla^2 \right) \nabla^2 w = \nabla_1^2 \phi_1 - Le \nabla_1^2 \theta_1 \quad (4)$$

$$\frac{\partial \phi_1}{\partial \tau} + R_s \frac{\partial \phi_0}{\partial z} w = \nabla_1^2 (\phi_1 + \epsilon \theta_1) \quad (5)$$

$$\frac{\partial \theta_1}{\partial \tau} + Ra \frac{\partial \theta_0}{\partial z} w = Le \nabla_1^2 \theta_1 \quad (6)$$

where ∇^2 is the three-dimensional Laplacian, and ∇_1^2 is the horizontal one with respects to x and y . Here z , τ , w , θ_0 , θ_1 , ϕ_0 and ϕ_1 are the dimensionless vertical distance, time, basic temperature, perturbed temperature, basic concentration and perturbed concentration,

respectively. Each variable has been nondimensionalized by using d , d^2/α , α/d , ΔT , $\nu\alpha/g\beta d^3$, ΔC and $\nu\alpha_s/g\gamma d^3$, respectively. The proper boundary conditions are

$$w_1 = \frac{\partial w_1}{\partial z} = \phi_1 = \theta_1 = 0 \quad \text{for } z=0 \text{ and } z=1 \quad (7)$$

$$\phi_0 = \theta_0 + 1 = 0 \quad \text{at } z=0 \quad (8)$$

$$\phi_0 = \theta_0 + 1 = 1 \quad \text{at } z=1 \quad (9)$$

Eq. (7) satisfies the condition of no fluctuation of perturbed quantities at rigid boundaries.

Through the method of separation of variables, the Graetz type solution for the basic concentration field is easily obtained as

$$\phi_0 = z - \sum_{n=1}^{\infty} \frac{2}{n\pi} \sin(n\pi z) \exp(-n^2\pi^2\tau) \quad (10)$$

For the deep-pool system of small τ , the basic concentration field is represented by

$$\phi_0 = \text{erf} \left(\frac{z}{4\sqrt{\tau}} \right) \quad (11)$$

For the thermally stably-stratified fluid layer, the dimensionless temperature field satisfying eqs. (8) and (9) will be linear as shown in Fig. 1. Therefore the basic density field satisfying the equation of state can be defined as

$$\bar{\rho} = -1 + \phi_0 - \frac{RaLe}{Rs} \theta_0 \quad (12)$$

where $\bar{\rho}$ denotes the nondimensionalized basic density defined as $(\rho - \rho_0)\Delta C\gamma/\rho_0$. The resultant variation of the profile of the basic density is shown in Fig. 2 where maximum magnitude of density locates within the fluid layer. The density profile is quite similar to

that of the Ueda et al.'s system¹⁷⁾

2.2. Propagation Theory

For a given Rs , Sc , Ra , Le and ϵ the time to mark the onset of convective motion is to be found under the principle of exchange of stabilities from eq. (4)–(6), subjected to the boundary condition (7). Even though the initially stratified density field may reduce the magnitude of the generated disturbances, the disturbances are to be generated continuously. Therefore the density distribution for molecular diffusion of heat and ion in aqueous solution is time-dependent. This is a formidable task to obtain quantitative results for the onset time of the double-diffusive convection. In frozen-time model the terms involving $\partial(\cdot)/\partial\tau$ are neglected and therefore the system becomes time-independent. The proper initial conditions at $\tau=0$ are required in stochastic model and amplification theory. Among the models the amplification theory is quite popular but its amplification factor to represent manifest convection should be decided experimentally. However, the propagation theory described below is a rather simple, deterministic approach even

though it involves the transient effect and for $Rs=10^7$, $Le=300$ and $\tau=0.001$.

According to the normal mode analysis, convective motion is assumed to exhibit the horizontal periodicity¹⁸⁾. Then the perturbed quantities are written in terms of dimensionless wave numbers a_x and a_y as

$$[w_i(\tau, x, y, z), \phi_i(\tau, x, y, z), \theta_i(\tau, x, y, z)] = [w_i^*(\tau, z), \phi_i^*(\tau, z), \theta_i^*(\tau, z)] \exp[i(a_x x + a_y y)] \quad (13)$$

where 'i' is the imaginary number. Substitution of the above eq. (13) into eq. (4)–(6) produces the usual amplitude functions in terms of the dimensionless horizontal wave number $a = (a_x^2 + a_y^2)^{1/2}$. The propagation theory employed for finding the critical time t_c is based on the assumption that disturbances at the onset of convection are mainly confined within the solutal penetration depth Δ_c and the following scale analysis in terms of Δ_c ($\propto t^{1/2}$) would be valid for perturbed quantities of eq. (4)–(6), respectively:

$$\frac{\partial C_1}{\partial t} \sim w_1 \frac{\partial C_0}{\partial Z} \sim \alpha_s \nabla^2 C_1 \sim \alpha_s \frac{C_1}{\Delta_c^2} \quad (14)$$

$$g\gamma C_1 \sim v \frac{w_1}{\Delta_c^2}, \quad w_1 \sim \frac{g\gamma \Delta_c^2}{v} C_1 \quad (15)$$

Then, the following peculiar relation is obtained from the above equations:

$$\frac{\partial C_0}{\partial Z} \sim \frac{\alpha_s v}{g\gamma \Delta_c^2} = \frac{\Delta C}{\Delta_c} \left(\frac{g\gamma \Delta T \Delta_c^3}{\alpha_s v} \right)^{-1} = \frac{\Delta C}{\Delta_c} Rs_{\Delta c}^{-1} \quad (16)$$

where $Rs_{\Delta c}$ is the solutal Rayleigh number based on the length Δ_c and concentration difference ΔC . With increasing Rs both the dimensionless critical time τ_c and the corresponding Δ_c will become smaller. Now it

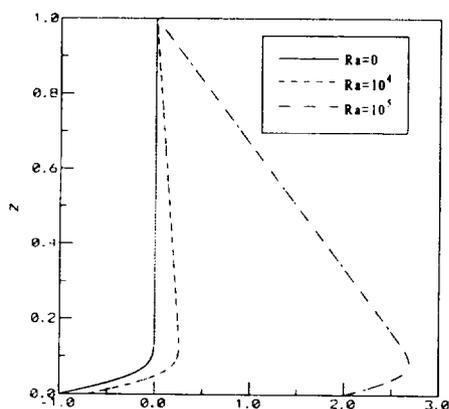


Fig. 2. Base density profiles with respect Ra

is assumed that for small t the characteristic value of $Rs_{sc} (\propto Rst^{3/2})$ will become a constant since $|\partial C_0 / \partial z| \sim \Delta C / \Delta_c$ in eq. (16). This trend was predicted by Foster⁶⁾, Wankat and Homsy⁷⁾, Jhavarly and Homsy⁸⁾ and Choi et al.⁹⁾. In this analysis, we assumed $g\gamma C_i \gg g\beta T_i$, that is, natural convection is governed mainly by concentration difference.

With the above reasoning the dimensionless amplitude functions of disturbances, based on relation (13), are assumed to have the form of

$$[w_1^*(\tau, z), \theta_1^*(\tau, z), \phi_1^*(\tau, z)] = [\tau w^*(\zeta), \theta^*(\zeta), \phi^*(\zeta)] \quad (17)$$

where $\zeta = z/\sqrt{\tau}$. The similarity variable ζ is introduced to take into account of the position and temporal dependencies of disturbances. By using relations (13) and (17) the following new set of dimensionless stability equations are obtained,

$$\begin{aligned} (D^2 - a^{*2})^2 w^* &= \frac{1}{Sc} \left(-\frac{1}{2} \zeta D^3 + \frac{1}{2} a^{*2} \zeta D - a^{*2} \right) w^* \\ &+ a^{*2} (\phi^* - Le \theta^*) \end{aligned} \quad (18)$$

$$\left(D^2 + \frac{1}{2} \zeta D - a^{*2} \right) \phi^* = Rs^* w^* D \phi_0 + \epsilon (D^2 - a^{*2}) \theta^* \quad (19)$$

$$\left(D^2 + \frac{1}{2Le} \zeta D - a^{*2} \right) \theta^* = -\frac{Ra^*}{Le} w^* \quad (20)$$

with boundary conditions:

$$w^* = Dw^* = \phi^* = \theta^* = 0 \text{ for } \zeta = 0 \text{ and } \zeta \rightarrow \infty \quad (21)$$

where $a^* = a\sqrt{\tau}$, $Rs^* = Rst^{3/2}$, $Ra^* = Ra\tau^2$ and $D = d/d\zeta$. These equations involve time-dependent properties implicitly. It is assumed that a^* , Ra^* and Rs^* are all eigenvalues and the principle of exchange of stabilities is kept. This is essence of the propagation theory. For a given Sc , Le , a^* , ϵ and Ra^* the minimum value of Rs^* will be found numerically.

2.3. Solution Method

In order to integrate the stability equations, eq. (18)–(21), trial value of the eigenvalue Rs^* and the boundary conditions $D^3 w^*$, $D\phi^*$ and $D\theta^*$ at $\zeta=0$ are assumed properly for given Sc , Le , a^* , ϵ and Ra^* . Here the value of Sc and Le are fixed at 2100 and 300, respectively, in order to consider double diffusion of heat and copper ion in aqueous sulfuric acid and copper sulfate solution. Since boundary conditions represented by eq. (21) are all homogeneous, the value of Dw^* at $\zeta=0$ can be assigned arbitrarily. This procedure is based on the outward shooting method in which the boundary value problem is transformed into the initial value problem. The trial values, with together the four known conditions at the heated boundary, give all the information to make numerical integration. The integration based on the 4th-order Runge-Kutta method is performed from $\zeta=0$ to fictitious distance to satisfy the infinite boundary conditions. By using the Newton-Raphson iteration the trial value of Ra^* , $D^3 w^*$, $D\phi^*$ and $D\theta^*$ are corrected until the stability equations satisfy the infinite boundary conditions within the maximum relative tolerance of 10^{-8} . Then, by increasing the distance step by step, the above integration is repeated. Finally, the value of Rs^* is decided through the extrapolation.

III. RESULTS AND DISCUSSION

With changing values of a^* and then Rs^* , the neutral stability curves are obtained for $Le=300$ which denotes the ratio of molecular diffusivity for heat to that for ion transfer. And, also the Schmidt number is taken to be 2100 for the present system involving double diffusion of heat and copper ion in aqueous solution. The resulting neutral stability curves are shown in Fig. 3.

According to the present theory it is considered that for a given Ra^* the minimum value of Rs^* and a^* , as shown in Fig. 3, characterizes the critical condition of convective motion. By using the relation of $Rs_c^* = Rs\sqrt{\tau_c^3}$ and $Ra_c^* = Ra\tau_c^2$, we can obtain the critical time τ_c for a given Rs and Ra .

For a given Sc , Le and Rs , as Ra increases the onset time is further delayed for $\epsilon \geq 0$. However, this trend is reversed for $\epsilon < -1$, as shown in Fig. 4. This figure shows the effect of the temperature gradient on the critical time. The results are for several values of Ra , the minimum or maximum bound of this figure, i.e. $\tau_{c,0} = 7.53Rs^{-1/3}$ corresponds to the case of the zero temperature gradient of $Ra=0$. In this limiting case the Lewis number and Soret coefficient do not affect the critical time, and the present values of τ_c are about on fourth of Ueda et al.'s results predicted by the amplification theory¹⁷⁾. It is thought that the most dangerous instabilities initiated at the time τ_c will grow to manifest themselves

around the time $4\tau_c$. For deep-pool system, $4\tau_c$ represents manifest convection very well¹⁹⁾. This may support Foster's viewpoint that with correct dimensionless relations obtained the times predicted for the time of convection motion would be too short by a factor of about 4²⁰⁾.

IV. CONCLUSION

In the present study, the onset of double-diffusive convection in an initially stably stratified fluid layer with temperature gradient has been analyzed by using propagation theory. Quantitative results for the onset time are presented for the specific situation of double diffusion of heat and copper ion in aqueous solution. The layer with larger Ra requires the larger buoyancy force to induce convective motion because of more stable stratification for $\epsilon \geq 0$. For a certain range of ϵ , that is $\epsilon < -1$, further increase of Ra promote the convective motion. This feature implies the Soret effect plays an important role in double-diffusive convection.

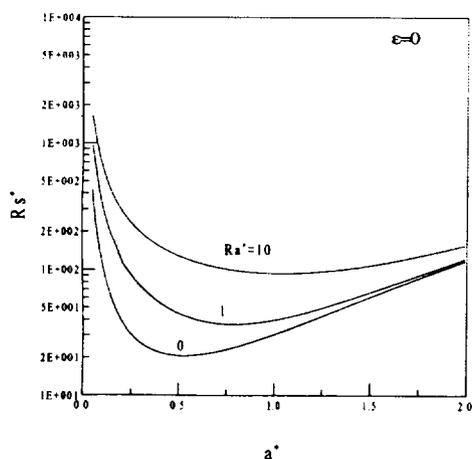


Fig. 3. Neutral stability curves for $Sc=2100$, $Le=300$ and $\epsilon=0$.

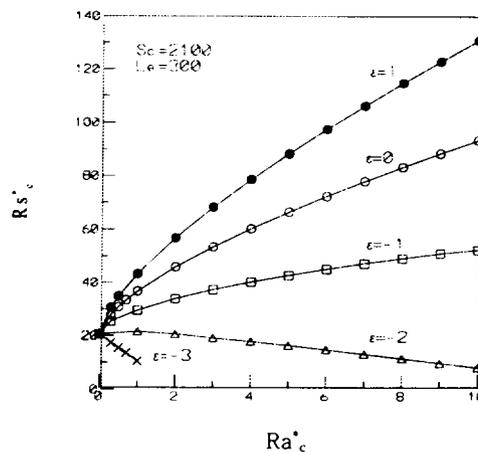


Fig. 4. Critical conditions for various ϵ .

요 약

시간의 함수로 주어지는 비선형 농도장 하에서의 이중확산 대류의 발생을 이론적으로 해석하였다. 초기에 균일하고, 안정된 온도 구배가 정지 상태의 유체층의 하부의 농도를 변화시켜서 유발되는 대류 발생 임계 조건에 미치는 영향을 조사하였다. 안정성 해석은 선형이론을 근거로 한 전파이론을 사용하였고, Soret 효과를 고려하였다. 해석결과 얻어진 대류 발생 임계시점은 온도 Rayleigh 수, 용질 Rayleigh 수, Soret 효과 계수의 함수로 주어진다.

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