

## CONVECTIVE INSTABILITIES AND TRANSPORT PROPERTIES IN HORIZONTAL FLUID LAYERS

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**Abstract** – This paper concerns the analysis of convective instabilities and fully developed transport properties in Bénard convection. The onset of convective instabilities driven by surface-tension variations and buoyancy forces is analyzed theoretically by using the propagation theory we have developed. Based on these stability criteria, the subsequent transport correlations of fully developed buoyancy-driven convection in horizontal fluid layers are suggested. It is found that the present predictions are compared favorably with existing experimental results.

Key words: Bénard Convection, Convective Instabilities, Marangoni Number, Nusselt Number, Rayleigh Number

### INTRODUCTION

It is well known that Bénard convection sets in due to the combined effects of surface tension and buoyancy forces. Rayleigh-Bénard convection is driven by buoyancy forces, while Bénard-Marangoni convection is driven by surface-tension variations. Its instability mechanism was first analyzed in favor of buoyancy forces by Lord Rayleigh [1916], but Pearson [1958] proved that surface-tension variations with temperature along a free surface can incur convective motions. In a fully developed conduction state the Rayleigh-Pearson analysis indeed describes the onset of convection very well, but the stability criteria in unstable, time-dependent nonlinear temperature fields still remain cloudy. This interesting behavior of natural convection often occurs in a variety of important engineering systems such as solvent extraction, gas absorption, distillation and crystal growth.

The stability problem with a time-dependent base temperature profile has been analyzed theoretically by several models: (a) the frozen-time model, (b) the amplification theory, (c) the energy method, (d) the stochastic model, and (e) the propagation theory. Of these, the amplification theory has been quite popular, but it requires the initial conditions and the amplification ratio. The propagation theory we have developed decides deterministically the stability criteria to mark the onset time by using the thermal penetration depth as a length-scaling factor and transforming the linearized perturbation equations similarly. With this theory the predicted values of the critical conditions have been consistent with most of the experimental results [Yoon et al., 1996; Kim et al., 1996; Hwang and Choi, 1996; Kang and Choi, 1997].

In this study the onset of convection in an initially quiescent, horizontal fluid layer cooled from above with a uniform,

constant heat flux is analyzed by using the propagation theory. The effects of coupling between buoyancy forces and surface-tension variations are considered. A new heat transfer correlation on the fully developed buoyancy-driven convection is produced by incorporating the resulting stability criteria into both the boundary-layer instability model of Howard [1964] and Busse [1967], and the theoretical equation of Long [1976] and Cheung [1980]. Also, a heat transfer phenomenon involving the effects of surface-tension variations are discussed with Hinkebein and Berg's [1978] experiments on silicone oil.

### GOVERNING EQUATIONS

The system considered in the present study is shown schematically in Fig. 1. A horizontally infinite fluid layer is placed between the upper free and the lower rigid boundaries of depth  $L$ . The layer with a flat, nondeformable free surface is initially quiescent at a constant temperature  $T_i$ . At the time  $t=0$  cooling starts. The governing equations are given under the Boussinesq approximation by

$$\nabla \cdot \mathbf{U} = 0 \quad (1)$$

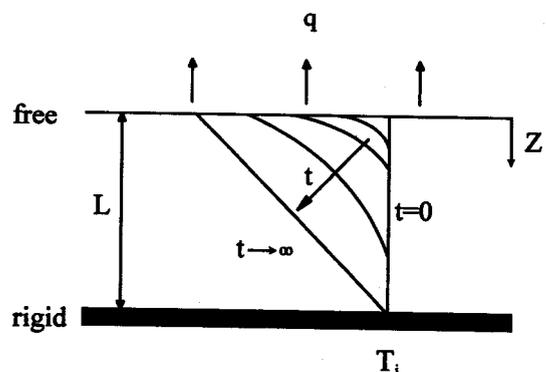


Fig. 1. Schematic diagram of present system.

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$$\rho_i \left[ \frac{\partial}{\partial t} + \mathbf{U} \cdot \nabla \right] \mathbf{U} = -\nabla P + \mu \nabla^2 \mathbf{U} + \rho g \mathbf{k} \quad (2)$$

$$\left[ \frac{\partial}{\partial t} + \mathbf{U} \cdot \nabla \right] T = \alpha \nabla^2 T \quad (3)$$

$$S = S_i + \gamma(T_i - T) \quad (4)$$

$$\rho = \rho_i [1 + \beta(T_i - T)] \quad (5)$$

where  $\mathbf{U}$  denotes the velocity vector,  $\rho$  the density,  $P$  the pressure,  $\mu$  the viscosity,  $\mathbf{k}$  the unit vector in the downward  $Z$ -direction,  $\alpha$  the thermal diffusivity,  $T$  the temperature,  $S$  the surface tension,  $\beta$  the thermal expansion coefficient and  $\gamma$  the negative value of  $dS/dT$ . The subscript  $i$  refers to the initial state.

The basic conduction state of no convective motion is governed by the following equation:

$$\frac{\partial \theta_0}{\partial \tau} = \frac{\partial^2 \theta_0}{\partial z^2} \quad (6)$$

with proper conditions,

$$\theta_0 = 0 \quad \text{at } \tau = 0 \quad (7a)$$

$$\frac{\partial \theta_0}{\partial z} = 1 \quad \text{or} \quad \frac{\partial \theta_0}{\partial z} = \text{Bi} \theta_0 \quad \text{at } z = 0 \quad (7b)$$

$$\theta_0 = 0 \quad \text{at } z = 1 \quad (7c)$$

where  $\tau$ ,  $z$ , and  $\theta_0 (=k(T_i - T)/qL)$  denote the dimensionless time, vertical distance and temperature, respectively;  $k$  denotes the thermal conductivity;  $\text{Bi}$  denotes the Biot number ( $=hL/k$ );  $h$  is the film heat transfer coefficient. Length has been scaled by the layer thickness  $L$  and time by  $L^2/\alpha$ . The solution of Eq. (6) is well known (for example, refer to Kang and Choi's [1997] work).

Over a certain uniform heat flux  $q$ , i.e.,  $\partial\theta_0/\partial z=1$  at  $z=0$ , exceeding the critical value, convection will set in. In this case the important dimensionless parameters are identified as

$$\text{Rayleigh number} \quad \text{Ra} = g\beta q L^4 / (k\alpha\nu) \quad (8a)$$

$$\text{Marangoni number} \quad \text{Ma} = \gamma q L^2 / (\mu k \alpha) \quad (8b)$$

$$\text{Prandtl number} \quad \text{Pr} = \nu/\alpha \quad (8c)$$

$$\text{Nusselt number} \quad \text{Nu} = Q_{\text{actual}}/Q_{\text{conduction}} \quad (8d)$$

where  $g$  denotes the gravity acceleration,  $\nu$  the kinematic viscosity and  $Q$  the heat transfer rate. In a fully developed state the temperature profile is linear and the critical condition to mark the onset of convection is given by

$$\text{(buoyancy forces)} \quad \text{Ra}_c^\infty = 669 \quad (9)$$

$$\text{(thermocapillary forces)} \quad \text{Ma}_c^\infty = 79.6 \quad (10)$$

wherewith  $\text{Nu}=1$ . With rapid cooling, natural convection will set in at a certain time for a given  $\text{Pr}$ ,  $\text{Ma}$  and  $\text{Ra}$ . Therefore, when  $\text{Ra} > 669$  or  $\text{Ma} > 79.6$  the onset time of convection  $\tau_c$  and its subsequent heat transfer characteristics become the important questions.

## PROPAGATION THEORY

Under linear theory the instantaneous quantities of temperature, velocity and pressure fields at the onset time of motion are perturbed by introducing infinitesimally small disturbances. The dimensionless disturbance equations are obtained as usual in terms of the temperature component  $\theta_1$  and the vertical velocity component  $w_1$ :

$$\left( \frac{1}{\text{Pr}} \frac{\partial}{\partial \tau} - \nabla^2 \right) \nabla^2 w_1 = - \frac{\text{Ra}}{\text{Ma} a^2} \nabla^2 \theta_1 \quad (11a)$$

$$\frac{\partial \theta_1}{\partial \tau} + \text{Ma} a^2 \frac{\partial \theta_0}{\partial z} = \nabla^2 \theta_1 \quad (11b)$$

where  $w_1$  has the scale of  $\alpha/L$ ,  $\theta_1$  has the scale of  $\Delta T/(a^2 \text{Ma})$ .  $\Delta T (=qL/k)$  represents the characteristic temperature difference in the present system. The proper boundary conditions, subject to Eqs. (11a) and (11b), are given by

$$w_1 = \left( \frac{\partial \theta_1}{\partial z} \text{ or } \frac{\partial \theta_1}{\partial z} - \text{Bi} \theta_1 \right) = \frac{\partial^2 w_1}{\partial z^2} + \frac{\nabla_1^2 \theta_1}{a^2} = 0 \quad \text{at } z = 0 \quad (12a)$$

$$w_1 = \frac{\partial w_1}{\partial z} = \theta_1 = 0 \quad \text{at } z = 1 \quad (12b)$$

where  $\nabla_1^2 = (\partial^2/\partial x^2 + \partial^2/\partial y^2)$ .

Based on the normal mode analysis, the amplitude functions  $w^*$  and  $\theta^*$  are constructed as a function of  $\zeta (=z/\tau^{1/2})$  only. By assuming periodic motion of disturbances in form of regular cells over the horizontal plane the following relationship is obtained:

$$[w_1(\tau, z), \theta_1(\tau, z)] = [\tau w^*(\zeta), \theta^*(\zeta)] \exp[i(a_x x + a_y y)] \quad (13)$$

where  $i$  is the imaginary number and the horizontal wave number  $a$  has the relation of  $a = (a_x^2 + a_y^2)^{1/2}$ . The above relationship means that  $w_1 = O(\delta_\tau^2)$  and  $\delta_\tau = 3.21 \tau^{1/2}$  for  $\tau < 0.1$ .  $\delta_\tau$  denotes the dimensionless thermal penetration depth, with  $\theta_1(\tau, \delta_\tau)/\theta_1(\tau, 0) = 0.01$ .

Now, for deep pool systems of small  $\tau$  the following set of stability equations is derived from Eqs. (11)-(13):

$$[(D^2 - a^2)^2 + \frac{1}{2\text{Pr}}(\zeta D^3 - a^2 \zeta D + 2a^2)] w^* = \frac{\text{Ra}^*}{\text{Ma}^*} \theta^* \quad (14a)$$

$$[D^2 + \frac{1}{2} \zeta D - a^2] \theta^* + \text{Ma}^* a^2 w^* D \theta_0 = 0 \quad (14b)$$

with the boundary conditions,

$$w^* = D^2 w^* - \theta^* = 0 \quad \text{at } \zeta = 0 \quad (15a)$$

$$D \theta^* = 0 \text{ or } D \theta^* = \text{Bi}^* \theta^* \quad \text{at } \zeta = 0 \quad (15b)$$

$$w^* = D w^* = \theta^* = 0 \quad \text{for } \zeta \rightarrow \infty \quad (15c)$$

where  $a^* = \tau^{1/2} a$ ,  $\text{Ma}^* = \tau \text{Ma}$  or  $\tau^{1/2} \text{Ma}$ ,  $\text{Ra}^* = \tau^2 \text{Ra}$  or  $\tau^{3/2} \text{Ra}$  and  $D = d/d\zeta$ . The upper boundary conditions at  $\zeta=0$  mean that constant heat flux is maintained and no surface deformation is possible. It is known that for fluids under usual conditions the crispation number ( $=\mu\alpha/(S_i L)$ ), which represents the degree of deformability of the free surface, is quite small, ranging from  $10^{-6}$  to  $10^{-2}$ . Under special cases of thin layers with

low surface tension or viscoelastic fluids, the value of the crispation number approaches unity and surface deflection becomes significant. The lower boundary conditions indicate that the lower boundary is rigid and its boundary temperature is constant. We assume that the parameters  $a^*$ ,  $Ma^*$  and  $Ra^*$  involving time implicitly are all eigenvalues. In buoyancy-driven convection Goldstein and Volino [1995] summarized this kind of treatment.

The appropriateness of Eqs. (14)-(15) is verified to a certain degree from the following dimensional scale analysis, based on Eq. (3) and the last boundary condition in Eq. (12a):

$$\frac{\partial T_1}{\partial t} \sim W_1 \frac{\partial T_0}{\partial Z} \sim \alpha \nabla^2 T_1 \sim \alpha \frac{T_1}{\delta_t^2}, \quad \mu \frac{\partial^2 W_1}{\partial Z^2} \sim \frac{\gamma a^2}{L^2} T_1 \sim \mu \frac{W_1}{\delta_t^2} \quad (16)$$

at  $t=t_c$

where  $\delta_t$  denotes the thermal penetration depth. For small  $\tau$ ,  $\delta_t \propto \sqrt{\tau}$  and for large  $\tau$ ,  $\delta_t=L$ . According to the above balances  $W_1/T_1$  has the order of magnitude  $\delta_t^2$  and this is represented by Eq. (13). From the two relationships of Eq. (16) the following interesting balance is obtained:

$$\frac{\partial T_0}{\partial Z} \sim \frac{\alpha}{W_1} \frac{T_1}{\delta_t^2} \sim \frac{qL}{\delta_t k} \frac{\alpha \mu k}{\gamma \delta_t qL} \frac{L^2}{a^2 \delta_t^2} \sim \frac{qL}{\delta_t k} (Ma^* a^{*2})^{-1} \quad (17)$$

These balances show that nondimensionalization of  $\partial T_0/\partial Z$  is the backbone of Eq. (15), for this term is the driving force to incur convective motion. Therefore, for large  $Ma$  the magnitude of  $W_1/T_1$  is very small, having the order of magnitude  $O(\delta_t^2)$  at  $t=t_c$ , but the magnitude of  $\partial T_0/\partial t$  becomes much larger in comparison with that of  $T_1$ . For a given  $Pr$  and  $Ma^*$ , the minimum value of  $Ra^*$  and its corresponding value  $a_c^*$  are sought numerically. In other words, the minimum value of  $\tau$ , i.e.,  $\tau_c$  to represent the onset time of convection is found deterministically for a given  $Pr$ ,  $Bi$ ,  $Ma$  and  $Ra$ . The above procedure is the essence of our propagation theory.

In solving Eqs. (14) and (15) the outer boundary may be assumed to be infinity. Therefore the shooting method is used in the solution of the present problem [Press et al., 1992]. For a specified  $Pr$ ,  $a^*$ , and  $Bi^*$  three initial guesses on  $D^3 w^*$ ,  $\theta^*$  and  $Ma^*$  at  $\zeta=0$  are given. Consequently the problem is converted to an initial value problem. The method used to integrate the disturbance equations is the fifth-order Runge-Kutta scheme. The Newton-Raphson iteration modifies the guessed values at  $\zeta=0$  until the integration results agree with the outer boundary condition within the error tolerance of  $10^{-6}$ .

**ONSET OF CONVECTIVE MOTION**

**1. Heat Conduction Systems**

The results of the present stability criteria obtained for  $Pr=7$  by using the propagation theory are featured in Fig. 2. It is assumed that for a given  $Ma^*$  the minimum value of  $Ra^*$ , i.e., the  $Ra_c^*$ -value for each curve represents the critical conditions to mark the convective motion. From this figure it is known that the  $Ra_c^*$ -value decreases with increasing  $Ma^*$  and this trend is also found in Nield's [1964] work. From these critical values we can obtain the critical time  $\tau_c$  to mark the onset of convective motion.

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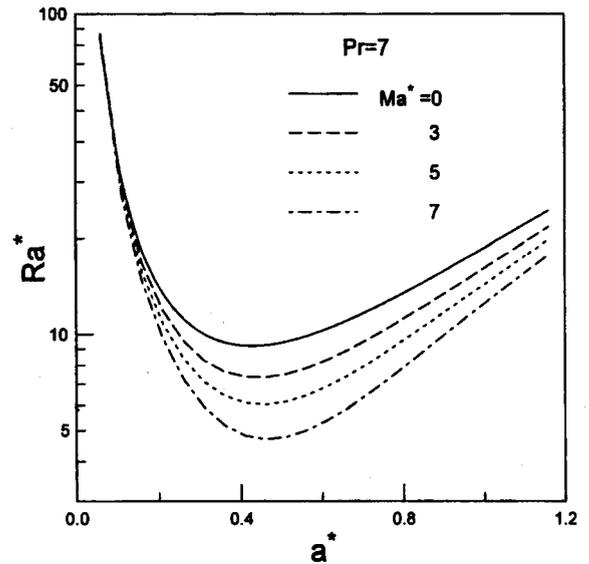


Fig. 2. Marginal stability curves with  $Pr=7$  for various  $Ma^*$ -values.

The critical time obtained numerically for the uniform heat-flux system is correlated within the maximum error of less than 5% by

$$\tau_c = 2.54 \left( 1 + \frac{1.25}{Pr^{0.723}} \right)^{0.696} Ra^{-1/2} \quad \text{for } Ma=0 \quad (18a)$$

$$\tau_c = 11.0 \left( 1 + \frac{0.753}{Pr^{0.681}} \right)^{1.27} Ma^{-1} \quad \text{for } Ra=0 \quad (18b)$$

It is certain that  $\tau_c$  decreases with an increase in  $Ma$  and  $Ra$ . With  $\tau_c=0.01$  the combined effect of  $Ma$  and  $Ra$  is shown in Fig. 3. It is interesting that the  $Ma$  effect becomes smaller with decreasing  $Pr$  since the value of  $Ma/Ra$  decreases. In this figure  $Ma_0$  and  $Ra_0$  are the values obtained with  $Ra=0$  and  $Ma=0$ , respectively, for  $\tau_c=0.01$  from Eq. (18). A similar trend of the  $Ra$  effect is shown in Fig. 4. It is known that for given

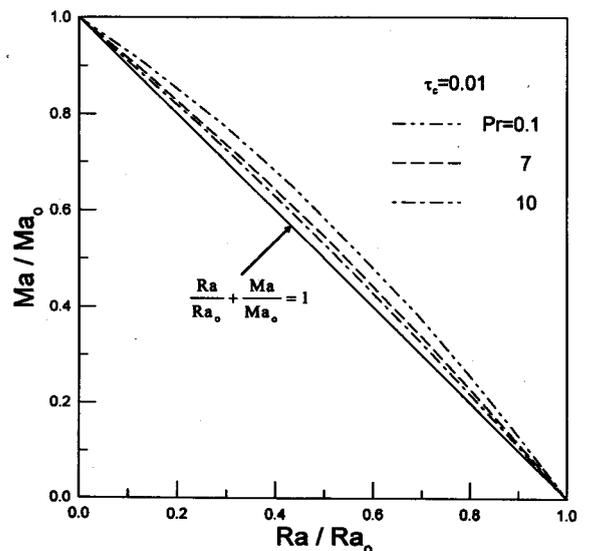


Fig. 3. Combined effect of  $Ma$  and  $Ra$  at  $\tau_c=0.01$ .

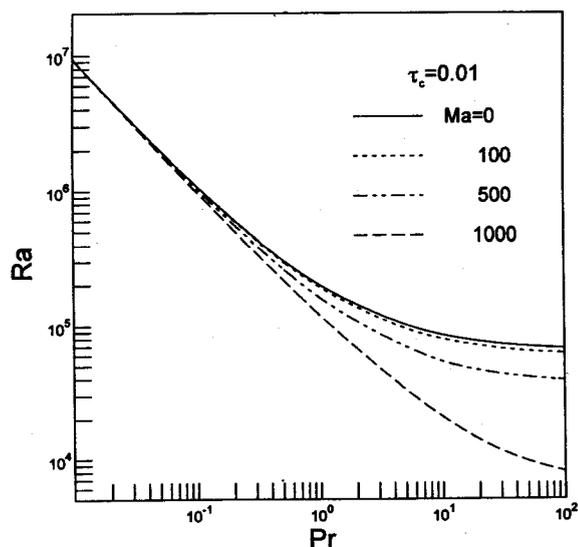


Fig. 4. Effect of Pr on Ra for given Ma at  $\tau_c=0.01$ .

$\tau_c$  the relationship between Ra and Ma constitutes

$$\frac{Ra}{Ra_0} + \frac{Ma}{Ma_0} \cong 1 \quad (19)$$

which was suggested by Nield [1964].

It is very difficult to conduct experiments on convective instabilities in case of Marangoni-Bénard convection, for the layer thickness is on the order of 1 mm. For example, a 1 mm-deep pool of water with an average temperature of 20°C and  $\Delta T=0.1^\circ\text{C}$  would have an Ra-value of 1.41 and the Ma-value of 111. With constant surface temperature maintained, no thermocapillary convection would be expected. Therefore, few refined experimental data exist in comparison with the above predictions. For propanol systems of  $Pr=30$ , Vidal and Acrivos [1968] conducted experiments when  $Ma \gg Ra$ . They obtained the theoretical relation of  $\tau_c=2/Ma$  by employing the frozen-time model. Gumerman and Homsy [1975] obtained lower bounds on  $\tau_c$  by the energy method. All these results are compared in Fig. 11 of Kang and Choi's [1997] paper. From this figure it is known that the present predictions of  $\tau_c$  are closer to the experimental values and our  $4\tau_c$  values are in good agreement with experiments. This may prove Foster's [1969] viewpoint that even though the correct dimensional relations would be obtained in terms of a time-dependent Rayleigh number using the thermal penetration depth as a scale factor, the times predicted for onset would be too short by a factor of about 4. The effect of Bi on the stability criteria can be found in the work of Kang and Choi [1997].

## 2. Mass Diffusion Systems

In gas absorption, natural convection can often occur due to surface-tension variations on the free boundary and also buoyancy forces. Its mechanism of the onset is analogous to that of the aforementioned heat conduction system, if we define  $\tau$ , Bi, Pr, Ma and Ra as

$$\text{dimensionless time} \quad \tau = \frac{tL^2}{D} \quad (20a)$$

$$\text{Biot number} \quad Bi = \frac{Hk_G L}{D} \quad (20b)$$

$$\text{Schmidt number} \quad Pr = \frac{\nu}{D} \quad (20c)$$

$$\text{Marangoni number} \quad Ma = \frac{\gamma_m \Delta CL}{\mu D} \quad (20d)$$

$$\text{Rayleigh number} \quad Ra = \frac{g\beta_m \Delta CL^3}{\nu D} \quad (20e)$$

where H denotes Henry's law constant for solute,  $k_G$  the gas-phase mass transfer coefficient,  $D$  the solute diffusivity,  $\gamma_m$  the negative surface-tension gradient with solute concentration,  $\Delta C$  the concentration difference between initial bulk liquid and bulk gas, and  $\beta_m$  the solutal expansion coefficient. Bi, Ma and Ra represent the Biot number, Marangoni number and Rayleigh number in mass transfer, respectively. The Schmidt number corresponds to the Prandtl number in heat transfer. The Marangoni number and Rayleigh number are the same since  $\Delta C/D$  in Eq. (20) is equivalent to  $qL/\alpha k$ . Therefore, we can use Eqs. (14) and (15) in predicting the critical time to mark the onset of convective motion in simple diffusion systems experiencing solute transfer across the free boundary.

With triethylamine Brian and Ross [1972] conducted gas desorption experiments induced by surface-tension variations only, wherein the Schmidt number is  $Pr=1503$  and the Biot number  $Bi=6.74$ . With these values the critical time is predicted, as shown in Fig. 5. It is known that the  $4\tau_c$ -value is much lower than the experimental data, of which the trend is different from that of propanol evaporation by Vidal and Acrivos [1968]. Brian and Ross [1972] and Imaishi et al. [1982] reported that a Gibbs adsorption layer would exist on the free boundary in mass transfer systems such as triethylamine-, methanol- and acetone-water systems and it would make them more stable. This interesting effect is now being investigated theoretically in our laboratory and their above interpreta-

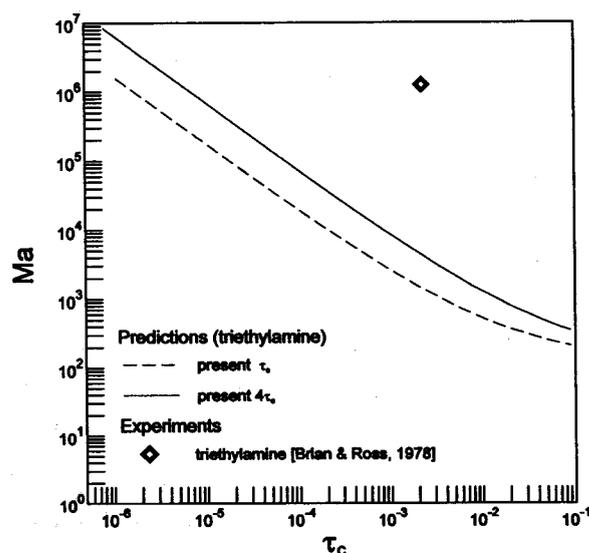


Fig. 5. Comparison of predictions with triethylamine experiment.

tion on the Gibbs adsorption layer has been proven in part. This implies that the mechanism of natural convection in mass transfer is more complicated than that in heat transfer.

**TRANSPORT CORRELATION**

The heat transfer rate increases with an increase in Ra or Ma. The possibility of connecting stability criteria to the fully developed heat transfer in buoyancy-driven thermal convection, the above Rayleigh-Bénard convection, has been investigated theoretically. Howard [1964] and Busse [1967] suggested that the generation of thermals would be the result of thermal instabilities in the conduction layers adjacent to the boundaries. With the fully developed, turbulent thermal convection flow the Nusselt number Nu is given by Busse as

$$Nu = \frac{Q_{actual}}{Q_{conduction}} = \frac{k\Delta T/\delta}{k\Delta T/L} \approx \frac{1}{2} \frac{L}{\delta} \tag{21}$$

where  $\delta$  is called the conduction layer thickness illustrated in Fig. 6.  $\Delta T_\delta$  is the temperature difference over the thickness  $\delta$  and  $\Delta T$  that over the layer thickness L. If we assume the relation of  $\delta = \delta_c$ , in a fully developed state the following correlation is derived from Eq. (6):

$$Nu = Nu_c/4 = 0.208\tau_c^{-1/2} \quad \text{for } Ra \rightarrow \infty \tag{22}$$

where  $\delta_c$  and  $Nu_c$  denote the thermal penetration depth and the Nusselt number at  $\tau = \tau_c$ , respectively. The above relationship is transformed by using Eq. (18a):

$$Nu = A_1 Ra^{1/4} \quad \text{for } Ra \rightarrow \infty \tag{23}$$

where  $A_1 = 0.130(1 + 1.25/Pr^{0.723})^{-0.348}$ .

Long [1976] and Cheung [1980] proposed the following theoretical equation:

$$Nu = \frac{A_1 Ra^{1/4}}{1 - A_2 Ra^{-1/12}} \quad \text{for large Ra} \tag{24}$$

where  $A_1$  and  $A_2$  are empirical constants. The above equation converts to Eq. (22) as  $Ra \rightarrow \infty$ . By employing the shape assumption of Stuart [1964], the following relationship is obtained:

$$1/Nu = 1 - 0.670(Ra - 669)/669 \quad \text{for } Ra \rightarrow 669 \tag{25}$$

Therefore, we can construct a new heat transfer correlation

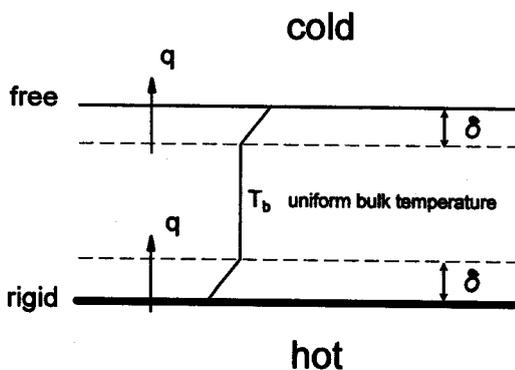


Fig. 6. Illustration of boundary-layer instability model in fully developed buoyancy-driven convection.

by combining Eqs. (21)-(25) as

$$Nu = 1 + \frac{A_1(Ra^{1/4} - 669^{1/4})}{1 - A_2 Ra^{-1/12}} \quad \text{for } Ra \geq 669 \text{ and } Ma=0 \tag{26}$$

where  $A_2 = 1.72 - 3.27A_1$ . The above correlation is shown for various Prandtl numbers in Fig. 7. It is shown that the effect of Pr is almost negligible for  $Pr \geq 10$ .

Hinkebein and Berg [1978] conducted experiments with silicone oil and ethylene glycol, as shown in Fig. 8. For a liquid depth larger than 5 mm buoyancy-driven convection is dominant. With these large-Pr fluids Eq. (26) representing heat transfer in buoyancy-driven convection agrees favorably well with the corresponding experimental data points. This kind of correlation is found consistent with experiments and other predictions in a horizontal fluid layer [Choi et al., 1988; Lee and Choi, 1993], volumetrically heated layer [Choi et al., 1992],

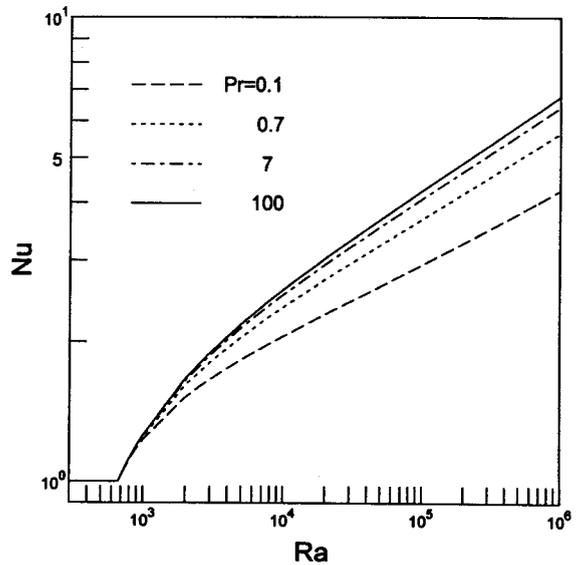


Fig. 7. Variations of Nu with Ra for various Prandtl numbers.

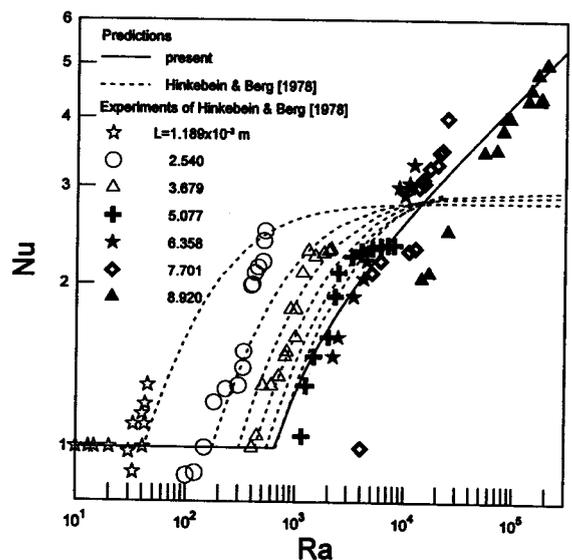


Fig. 8. Comparison of Nu vs. Ra with Hinkebein and Berg's [1978] experiments for Pr=125.

fluid-saturated porous media [Yoon and Choi, 1989], and plane Couette flow [Choi and Kim, 1994]. All these results correspond to the cases of buoyancy-driven convection. The figure also shows that the surface-tension effects become more important with a decrease in Ra. It is seen that  $Ra_c$  decreases with decreasing the liquid depth by following Eq. (19). The theoretical correlation combining both buoyancy forces and surface-tension variations was suggested by Hinkebein and Berg, but it does not agree with experiments for  $Nu > 3$ . For  $Nu > 2$  their correlation shows the relative ratio of Ma to Ra plays a crucial role in deciding Nu. The surface-tension effects will become more important with an increase in the ratio Ma/Ra.

Since the mechanism of heat transfer driven by buoyancy forces is different from that of surface-tension effects, it is not easy to derive the correlation for the latter case. Furthermore, in heat transfer systems decoupling two mechanisms is very difficult. In mass transfer systems Imaishi et al. [1982] conducted experiments on gas desorption using the wetted-wall column. Based on their experimental results, the following correlation is derived:

$$Nu \cong \left( \frac{Ma}{Ma_c} \right)^{2/7} \quad (27)$$

where  $Ma_c$  is the critical Marangoni number corresponding to the onset of convective motion in their experimental systems. Hinkebein and Berg [1978] derived the following theoretical equation:

$$Nu = \frac{2.7936}{1 + 1.7936 \frac{Ma_c^\infty}{Ma}} \quad (28)$$

where  $Ma_c^\infty = 79.6$  for the present system of uniform heat flux. With  $Ma = 79.6$  the above equation produces the same value of  $Nu = 1$ , but it generates an upper limit of  $Nu = 2.7936$  for  $Ma \rightarrow \infty$ . Therefore, Eq. (28) may be used near  $Ma = 79.6$ . Eqs. (27) and (28) indicate that the Nu-correlation in surface-tension-driven convection cannot be produced by following the procedure used in deriving Eq. (22). The mechanism of heat transfer in buoyancy-driven convection seems different from that in surface-tension-driven convection. Therefore, a more refined theory and experiments are still required.

## CONCLUSION

For the deep pool system of  $\tau < 0.1$  with its free upper surface under uniform heat flux, the critical time to represent the onset of combined surface-tension and buoyancy-driven convection has been analyzed by using the propagation theory. For the limiting case of  $Ra \ll Ma$  the predictions are compared well with the existing experimental data. It seems evident that the thermal penetration depth is the proper length-scaling factor, and manifest convection is observed experimentally near  $\tau = 4\tau_c$ . Based on the present critical time,  $\tau_c$ , a heat transfer correlation in fully developed buoyancy-driven convection is proposed. This represents the experimental data points reasonably well. But in the case of surface-tension-driven convection the theoretical Nu-correlation is not produced easily, and therefore it needs more refined work in the future.

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## NOMENCLATURE

- a : dimensionless wave number,  $(a_x^2 + a_y^2)^{1/2}$  [-]
- $a^*$  : modified dimensionless number,  $\tau^{1/2} a$  [-]
- $A_1, A_2$  : constants in Eq. (26) [-]
- Bi : Biot number,  $Hk_c L/D$  [-]
- $\Delta C$  : concentration difference [mol/m<sup>3</sup>]
- $\underline{D}$  : differential operator with respect to  $\zeta$  [-]
- $\bar{D}$  : solute diffusivity [m<sup>2</sup>/s]
- g : gravitational acceleration [m/s<sup>2</sup>]
- H : Henry's law constant for solute [J/mol · K]
- h : film heat transfer coefficient [J/s · K]
- i : imaginary number [-]
- k** : unit vector in downward Z-direction [-]
- k : thermal conductivity [J/s · m · K]
- $k_G$  : gas-phase mass transfer coefficient [m/s]
- L : fluid layer thickness [m]
- Ma : Marangoni number,  $\gamma q L^2 / (\mu k \alpha)$  or  $\gamma_m \Delta C L / \mu \bar{D}$  [-]
- $Ma^*$  : modified Marangoni number,  $\tau Ma$  or  $\tau^{1/2} Ma$  [-]
- Nu : Nusselt number,  $Q_{actual} / Q_{conduction}$  [-]
- P : pressure [N/m<sup>2</sup>]
- Pr : Prandtl number,  $\nu / \alpha$  or Schmidt number,  $\bar{\nu} / \bar{D}$  [-]
- Q : heat transfer rate [W]
- q : heat flux [J/m<sup>2</sup>]
- Ra : Rayleigh number,  $g \beta q L^4 / (k \alpha \nu)$  or  $g \beta_m \Delta C L^3 / \nu \bar{D}$  [-]
- $Ra^*$  : modified Rayleigh number,  $\tau^2 Ra$  or  $\tau^{3/2} Ra$  [-]
- S : surface tension [N/m]
- T : temperature [K]
- $T_b$  : bulk temperature [K]
- t : time [s]
- $\Delta T$  : temperature difference [K]
- U : velocity vector [m/s]
- W : vertical velocity [m/s]
- w : dimensionless vertical velocity,  $LW / \alpha$  [-]
- x, y, z : dimensionless Cartesian coordinates based on fluid depth [-]
- Z : vertical position in Cartesian coordinates [m]

## Greek Letters

- $\alpha$  : thermal diffusivity [m<sup>2</sup>/s]
- $\beta$  : thermal expansion coefficient [K<sup>-1</sup>]
- $\beta_m$  : solutal expansion coefficient [%<sup>-1</sup>]
- $\gamma$  : negative surface-tension gradient with temperature [N/m · K]
- $\gamma_m$  : negative surface-tension gradient with solute concentration [Nm<sup>2</sup>/mol]
- $\delta$  : conduction layer thickness [m]
- $\delta_T$  : dimensionless thermal penetration depth [-]
- $\delta_i$  : thermal penetration depth [m]
- $\zeta$  : similarity variable,  $z / \tau^{1/2}$
- $\theta$  : dimensionless temperature [-]
- $\mu$  : viscosity [kg/m · s]
- $\nu$  : kinematic viscosity [m<sup>2</sup>/s]
- $\rho$  : density [kg/m<sup>3</sup>]

$\tau$  : dimensionless time,  $at/L^2$  or  $\overline{Dt}/L^2$  [-]

### Subscripts

c : critical state  
i : initial state  
o : basic state  
1 : perturbed state

### Superscript

\* : amplitude function for perturbation quantities

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