Korean J. Chem. Eng., **26**(1), 21-25 (2009)

Onset of solutal convection in liquid phase epitaxy system

Min Chan Kim*,† and Sang Goo Lee**

Department of Chemical Engineering, Cheju National University, Jeju 690-756, Korea
*Ibule Photonics, Incheon 406-840, Korea
(*Received 9 May 2008 • accepted 7 July 2008*)

Abstract−The onset of convective instability in the liquid phase epitaxy system is analyzed with linear stability theory. New stability equations are derived under the propagation theory, and the dimensionless critical time τ_c to mark the onset of the buoyancy-driven convection is obtained numerically. It is here found that the critical Rayleigh number Ra_c is 8000, below which the flow is unconditionally stable. For $Ra>Ra_c$ the dimensionless critical time τ_c to mark the onset of a fastest growing instability is presented as a function of the Rayleigh number and the Schmidt number. Available numerical simulation results and theoretical predictions show that the manifest convection occurs starting from a certain time $\tau_o(>\tau_c)$. It seems that during $\tau_c \le \tau \le \tau_o$ secondary motion is relatively very weak.

Key words: Onset of Convection, LPE System, Propagation Theory, Critical Rayleigh Number

INTRODUCTION

Liquid phase epitaxy (LPE), which consists of depositing thin crystalline layer from a solution onto a substrate, is an important crystal growth process from both practically and fundamentally. In horizontal substrate-solution-substrate sandwich systems under the programmed cyclic temperature condition, dissolution occurs mainly on the lower substrate while the growth on the upper substrate [1]. In this case, the buoyancy-driven convection accelerates the dissolution near the lower substrate and is the driving force of the upward solute transport, *i.e.*, the solutal convection breaks the symmetric concentration field and makes LPE process possible.

Sukegawa et al. [2] experimentally investigated the effects of solutal convection in LPE growth of silicon in an indium solution. Later, Kimura et al. [3,4] made similar experiments to better understand the effects of solutal convection on the dissolution of silicon. For the silicon-indium system, Saitou and Motoyama [5] found an interesting experimental result that there is a critical depth between the two silicon substrates where the silicon growth rate on the upper substrate becomes zero. The critical depth was independent of the temperature profile. This means that there is a critical solutal Rayleigh number from which the silicon growth starts and the thermal effects can be neglected. Therefore, the onset of solutal convection is an important factor in the design of LPE systems.

To understand the solutal convection during the LPE process, Saitou and Motoyama [6] analyzed the onset of solutal convection by using linear stability analysis, and conducted numerical simulation on the effect of solutal convection on the dissolution rate of substrates. Recently, Coskun et al. [7] simulated the dissolution of silicon in an indium solution numerically by employing spectral method and compared their results with previous experimental results.

In the present study, the onset of solutal convection in LPE processes is analyzed theoretically by using propagation theory which has been applied to various systems [8-13]. This model rescaled all the variables and parameters having the length scale by using penetration depth and transformed the disturbance equations similarly. The critical Rayleigh number and the onset time for a given Rayleigh number are suggested and compared with previous studies.

STABILITY ANALYSIS

1. Governing Equations

The system considered here is a sandwich system given in Fig. 1. Two horizontal silicon substrates are set face to face, and indium solvent is inserted between the substrates. The substrates and indium are assumed to be heated up to 1,247 K and the whole system kept isothermal at 1,247 K. For time $t \ge 0$, the horizontal substrates maintain constant wall concentration C_w, which is the saturation concentration of silicon in indium solvent. In this case, buoyancy-driven solutal convection will set in at a certain time and the governing equations of flow and concentration fields are expressed by employing the Boussinesq approximation as

$$\nabla \cdot \mathbf{U} = 0, \tag{1}$$

$$\left\{\frac{\partial}{\partial t} + \mathbf{U}\cdot\nabla\right\}\mathbf{U} = -\frac{1}{\rho_r}\nabla P + \nu\nabla^2\mathbf{U} - g\gamma C\mathbf{k}, \tag{2}$$

$$\left\{\frac{\partial}{\partial t} + \mathbf{U}\cdot\nabla\right\}C = \alpha_s\nabla^2 C, \tag{3}$$

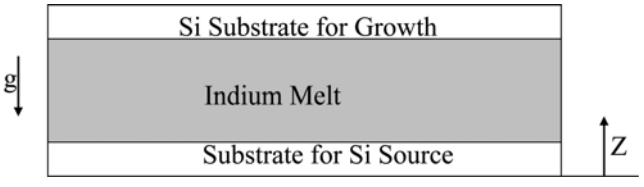

Fig. 1. Schematic diagram of Si-In-Si sandwich system considered here.

†To whom correspondence should be addressed.
E-mail: mckim@cheju.ac.kr
‡This article is dedicated to Professor Chang Kyun Choi for celebrating his retirement from the School of Chemical and Biological Engineering, Seoul National University.

where **U**, t, ρ, P, ν, g, γ, **k**, C and α_s represent the velocity vector, the time, the density, the dynamic pressure, the kinematic viscosity, the gravitational acceleration constant, the solutal expansion coefficient, the vertical unit vector, the concentration and mass diffusivity, respectively. The subscript "r" represents the reference state. In the present study, the thermal effects are neglected because the temperature difference between the substrates and the center of a layer of the solution is negligibly small in the real LPE processes [6].

The important parameters to describe the present system are the Schmidt number (Sc) and the Rayleigh number (Ra) defined by

$$\mathrm{Sc}=\frac{\nu}{\alpha_s} \quad \text{and} \quad \mathrm{Ra}=\frac{g\gamma C_w d^3}{\alpha_s \nu}, \tag{4}$$

where d is the distance between the substrates. In LPE system under the unstable condition, buoyancy-driven motion can be set in near both the upper substrate and the lower one, and breaks the symmetry of the concentration field. Asymmetric concentration field is essential to pump the solute from the lower substrate to the upper one. Therefore, it is important to understand the buoyancy-driven motion to design the LPE process. For a time-dependent system of large Ra, the resulting transient stability problem is complicated and the critical time t_c to mark the onset of convective motion becomes an important question.

For the basic state of pure diffusion during the dissolution period, the dimensionless concentration profile is represented by

$$\frac{\partial c_0}{\partial \tau}=\frac{\partial^2 c_0}{\partial z^2}, \tag{5}$$

with the following initial and boundary conditions,

$$c_0(0,z)=0, \quad c_0(\tau,1)=1 \quad \text{and} \quad \frac{\partial c_0}{\partial z}(\tau,1/2)=0. \tag{6}$$

In the above equations, $\tau=\alpha_s t/d^2$, $z=Z/d$ and $c_0=C_0/C_w$. The subscript '0' denotes the basic state. By using the Laplace transform method, the exact solution of Eqs. (5) and (6) is obtained as

$$c_0=\sum_{n=0}^{\infty}(-1)^n\left\{\mathrm{erfc}\left(\frac{n}{2\sqrt{\tau}}+\frac{\zeta}{2}\right)+\mathrm{erfc}\left(\frac{n+1}{2\sqrt{\tau}}-\frac{\zeta}{2}\right)\right\}, \tag{7}$$

where $\zeta=z/\sqrt{\tau}$ and erfc is the complementary error function. Based on the above solution, the dimensionless dissolution depths of both substrates can be calculated by using the following relation:

$$h(\tau)=\frac{H(\tau)}{d}\int_0^{\tau}\left.\frac{\partial c_0}{\partial z}\right|_{z=0 \text{ or } 1} d\tau, \tag{8}$$

where $H(\tau)$ is the dissolution depth at time τ. Since the length between the substrates is of the order of mm and the dissoution rate is a few μm/hr for a typical LPE system [6], the moving boundary effects can be neglected. For deep-pool systems of small τ, the Leveque-type solutions are given by

$$c_0=\begin{cases}\mathrm{erfc}\left(\dfrac{z}{2\sqrt{\tau}}\right) & \text{for } z<1/2\\ \mathrm{erfc}\left(\dfrac{1-z}{2\sqrt{\tau}}\right) & \text{for } z>1/2\end{cases} \quad \text{for } \tau\to 0. \tag{9}$$

and the dimensionless dissolution depth can be $h(\tau)=2\sqrt{\tau/\pi}$. For

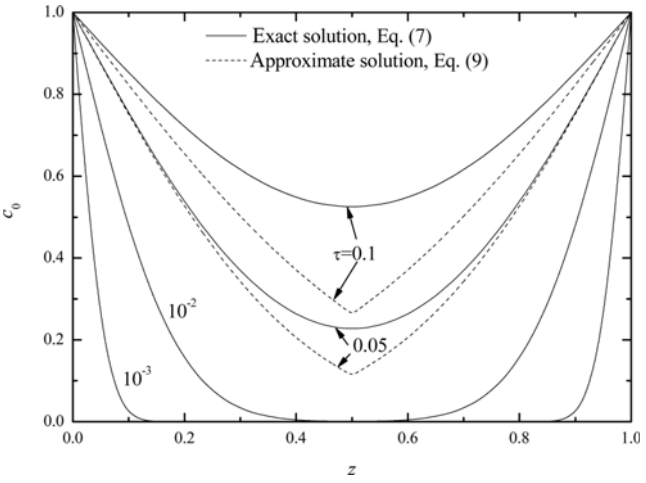

Fig. 2. Base concentration profiles for the various times.

$\tau\le 10^{-2}$ Eq. (9) approximates the exact solution Eq. (7) quite well, as shown in Fig. 2.

2. Stability Equations

Under linear stability theory, infinitesimal disturbances caused by incipient convective motion can be formulated, in dimensionless form, in terms of the concentration component c_1 and the vertical velocity component w_1 by linearizing Eqs. (1)-(3):

$$\left\{\frac{1}{\mathrm{Sc}}\frac{\partial}{\partial \tau}-\overline{\nabla}^2\right\}\overline{\nabla}^2 w_1=-\overline{\nabla}_1^2 c_1, \tag{10}$$

$$\frac{\partial c_1}{\partial \tau}+\mathrm{Ra}w_1\frac{\partial c_0}{\partial z}=\overline{\nabla}^2 c_1, \tag{11}$$

where the Laplacian $\overline{\nabla}^2=(\partial^2/\partial x^2)+(\partial^2/\partial y^2)+(\partial^2/\partial z^2)$ and the horizontal one $\overline{\nabla}_1^2=(\partial^2/\partial x^2)+(\partial^2/\partial y^2)$. Here the velocity component has the scale of α/d and the temperature component has that of $\alpha_s\nu/(g\gamma d^3)$. The proper boundary conditions are given by

$$w_1=\frac{\partial w_1}{\partial z}=c_1=0 \quad \text{at} \quad z=0 \quad \text{and} \quad z=1, \tag{12}$$

wherein the condition of no-slip and constant concentration are applied to the two rigid boundaries. For a given Sc and the Ra, the critical time τ_c should be found by using Eqs. (10)-(12). Detailed procedure to derive Eqs. (10)-(12) is well described in Chandrasekhar's book [14].

Now, convective motion is assumed to exhibit the horizontal periodicity and the normal mode analysis is employed. Then the disturbance quantities can be expressed as

$$[w_1(\tau,x,y,z), c_1(\tau,x,y,z)]=[w_1^*(\tau,z), c_1^*(\tau,z)]\exp[i(a_x x+a_y y)], \tag{13}$$

where "i" is the imaginary number and the horizontal wavenumber "a" has the relation of $a=[a_x^2+a_y^2]^{1/2}$. Propagation theory employed to find the onset time of convective motion, *i.e.*, the critical time τ_c is based on the assumption that in deep-pool systems the infinitesimal concentration disturbances are propagated mainly within the penetration depth at the onset of convective motion $\Delta_c(\propto\sqrt{\alpha_s t})$, and the following scale relations are valid for disturbance quantities from Eqs. (2) and (3):

$$\nu\frac{W_1}{\Delta_C^2}\sim g\gamma C_1, \tag{14a}$$

$$W_1\frac{\partial C_0}{\partial Z}\sim\alpha_s\frac{C_1}{\Delta_C^2}, \tag{14b}$$

from the balance between viscous and buoyancy terms in Eq. (2) and also from that among terms in Eq. (3). Now, based on Eq. (14a), the following amplitude relation is obtained in dimensionless form:

$$\frac{w_1^*}{c_1^*}\sim\delta_C^2\sim\tau, \tag{15}$$

where $\delta_C(=\Delta_C/d\propto\sqrt{\tau})$ is the usual dimensionless penetration depth following $c_0=0.01$ at $z=\delta_C$.

For the limiting case of $\tau\rightarrow 0$, based on the relation of Eq. (15), we assume dimensionless amplitude functions of disturbances like

$$[w_1^*(\tau,z),\ c_1^*(\tau,z)]=[\tau^{n+1}w^*(\tau,z),\ \tau^n c^*(\tau,z)]. \tag{16}$$

At this stage the criterion to determine the critical time is necessary. For small times the dissipative forces of viscosity and mass diffusion overweigh the buoyancy force and the diffusional process is stable. However, if the buoyancy force becomes large enough to overcome the dissipative forces, disturbances will grow. There is some critical time at which these are just equal. We define the critical time when

$$r_0=r_1 \quad \text{at} \quad \tau=\tau_c. \tag{17}$$

where the temporal growth rate of the disturbance (r_1) and the base quantity (r_0) are defined as the root-mean-squared quantities of concentration components:

$$r_0=\frac{1}{\langle c_0\rangle}\frac{d\langle c_0\rangle}{d\tau} \quad \text{and} \quad r_1=\frac{1}{\langle c_1\rangle}\frac{d\langle c_1\rangle}{d\tau}, \tag{18}$$

where $\langle\cdot\rangle=\sqrt{\left(\int_A(\cdot)^2dS\right)}/S$ and $dS=(2\pi d/a)dz$. Here denotes the cross-sectional area of one vortex pair. From the base concentraion distribution given by Eq. (9), r_0 is obtained as

$$r_0=\frac{1}{4\tau}. \tag{19}$$

For the case of $\delta_C\rightarrow 0$, the stretched vertical coordinate $\zeta(=Z/\Delta_C=z/\sqrt{\tau})$ is more suitable to describe the concentration field having boundary layer characterisitics. Therefore, we set $c_1(\tau,z)=\tau^n c^*(\tau,\zeta)$. For the case of n=0 and $(\partial c^*/\partial\tau)|_\zeta=0$, i.e. $c_1(\tau,z)=c^*(\zeta)$, the condition of $r_0=r_1$ is fulfiled at $\tau=\tau_c$. Now, we set

$$[w_1,c_1]=[\tau w_1^*(\zeta),c_1^*(\zeta)]\exp[i(a_x x+a_y y)]. \tag{20}$$

This means that the amplitude function of concentration disturbances follows the behavior of c_0 (see Eq. (6)). Even though we assume $(\partial c^*/\partial\tau)|_\zeta=0$, $(\partial c_1/\partial\tau)|_z$ cannot be neglected since $(\partial c_1/\partial\tau)|_z=(\partial c^*/\partial\tau)|_\zeta-(\zeta/2)(\partial c^*/\partial\zeta)|_\tau$. This is a critical difference between the present propagation theory and the frozen-time model where $(\partial c_1/\partial\tau)|_z=0$ is assumed.

Now, the self-similar stability equations are obtained in dimensionless form from Eqs. (10) and (11) as

$$\left\{(D^2-a^{*2})^2+\frac{1}{2Sc}(\zeta D^3-a^{*2}\zeta D+2a^{*2})\right\}w^*=a^{*2}c^*, \tag{21}$$

$$\left(D^2+\frac{1}{2}\zeta D-a^{*2}\right)c^*=Ra^*w^*Dc_0, \tag{22}$$

The proper boundary conditions are

$$w^*=Dw^*=c^*=0 \quad \text{at} \quad \zeta=0 \quad \text{and} \quad \zeta\rightarrow\infty. \tag{23}$$

Here $Ra^*(=Ra\tau^{3/2})$ and $a^*(=a\tau^{1/2})$ are eigenvalues and $D=d/d\zeta$. The conventional frozen-time model neglects the terms involving $\partial(\cdot)/\partial\tau$ in Eqs. (8) and (9) in amplitude coordinates τ and z. This results in $(D^2-a^{*2})^2w^*=a^{*2}c^*$ and $(D^2-a^{*2})c^*=Ra^*(Dc_0)w^*$ instead of Eqs. (21) and (22).

3. Solution Procedure

The stability Eqs. (21)-(23) are solved by employing the outward shooting scheme. To integrate these stability equations, the proper values of D^2w^*, D^3w^* and Dc^* at $\zeta=0$ are assumed for a given Sc and a^*. Since the stability equations and their boundary conditions are all homogeneous, the value of $D^2w^*(0)$ can be assigned arbitrarily and the value of the parameter Ra^* is assumed. This procedure can be understood easily by taking into account the characteristics of eigenvalue problems. After all the values at $\zeta=0$ are provided, this eigenvalue problem can proceed numerically.

Integration is performed from $\zeta=0$ to an upper boundary with the fourth order Runge-Kutta-Gill method. If the guessed value of Ra^*, $D^3w^*(0)$ and $Dc^*(0)$ are correct, w^*, Dw^* and c^* will vanish at the upper boundary. To improve the initial guesses the Newton-Raphson iteration is used. In the limiting case of $\tau\rightarrow 0$, the above procedure is conducted for a given fictitious upper boundary corresponding to $\zeta\rightarrow\infty$. When convergence is achieved, the upper boundary for computation is increased by a predetermined value and the above procedure is repeated. Since the concentration disturbances decay exponentially outside the penetration depth, the incremental change of Ra^* also decays fast with increasing a fictitious upper boundary thickness. This behavior enables us to extrapolate the eigenvalue to the infinite depth.

RESULTS AND DISCUSSION

For the limitig case of small τ, the basic concentration field is decoupled and then the concentration profile of the upper layer cannot affect the stability characteristics. For this case the base concentration profile for the lower unstable layer can be approximated as:

$$c_0=\text{erfc}\left(\frac{\zeta}{2}\right) \quad \text{for} \quad \tau>0.01 \quad \text{and} \quad z<1/2, \tag{24}$$

and stability criteria from the propagation theory can be obtaind from Kim et al.'s work [13] as:

$$\tau_c=7.53\left[1+\left\{\frac{0.804}{Sc}\right\}^{3/4}\right]^{8/9}Ra^{-2/3} \quad \text{for} \quad \tau_c<0.01. \tag{25}$$

The critical time τ_c to mark the onset of the fastest growing instability decreases with increasing Ra.

Now, the domain of time is extended to $\tau_c>0.01$ by keeping Eqs. (21) and (22) and using Eq. (7). In condition of Eq. (23) the upper boundary $\zeta\rightarrow\infty$ is replaced with z=1, i.e. $z=1/\sqrt{\tau_c}$ and in Eqs. (16) and (17) Ra^* and a^* are replaced with $Ra\tau_c^{3/2}$ and $a\sqrt{\tau_c}$. Also, in Eq. (7) τ is replaced with τ_c but ζ is maintained. Since τ_c is the fixed parameter, the resulting stability equations are a function of ζ only

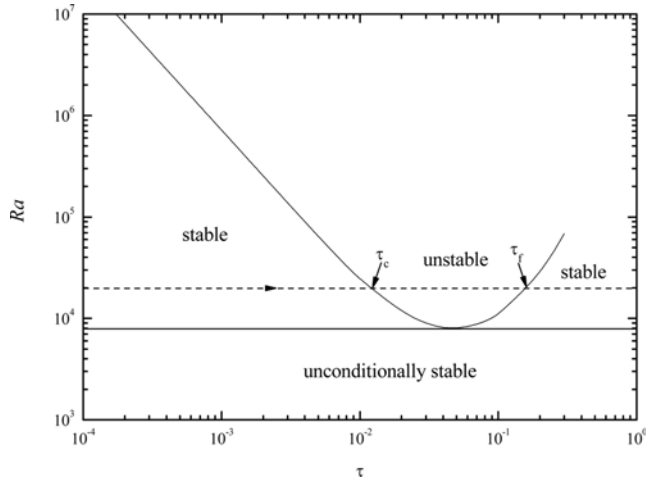

Fig. 3. Stability result for Si-In-Si sandwich system (Sc=15.7).

and the physics of Eqs. (15) and (20) is still alive. For a given Sc and τ_c the minimum Ra-value and its corresponding wavenumber a_c are obtained.

The stability condition obtained from propagation theory is summarized in Fig. 3. In this figure the disturbance trajectory for $Ra=2\times10^4$ is shown as a dashed line in the $Ra-\tau$ plane. The time inteval between $\tau_c\le\tau\le\tau_f$ represents the period during which the system is unstable. Due to the asymptotically unconditionally stable characteristics of the present system, a fastest growing single-mode instability sets in at τ_c and this instability decays out after τ_f. The regime of $Ra<Ra_c$ defines the unconditionally stable state; no growing instability is allowed in this range. Therefore, in order to use solutal convection as solute pumping mechanism, the LPE system should be operated under the condition of $Ra<Ra_c$. For the present system, due to the unconditionally stable characteristic of concentration profile for the steady state, the critical Rayleigh number is not well known value of 1708 but about 8000.

Saitou and Motoyama [6] analyzed the present problem and suggested the critical condition as

$$Ra\left|\frac{\partial c_0}{\partial z}\right|\approx 2\times10^3 \quad \text{for} \quad \sqrt{\tau}<0.25 \tag{26}$$

They approximated

$$\left|\frac{\partial c_0}{\partial z}\right|\sim\frac{1}{\sqrt{\pi\tau}}\exp\left(-\frac{z^2}{4\tau}\right) \quad \text{at} \quad z=\sqrt{2\tau}, \tag{27}$$

by assuming that $|\partial c/\partial z|$ has the maximum value at $z=\sqrt{2\tau}$. However, their analysis has no physical meaning and is mathematically wrong, since $(z\sqrt{\pi}/2)|\partial c_0/\partial z|$ rather than $|\partial c_0/\partial z|$ has a maximum value at $z=\sqrt{2\tau}$ [15]. Similar approach was applied to the onset of buoyancy-driven convection. Later, Tan and Thorpe [16] suggested a simple instability analysis assuming that at the onset of convection the following relation is maintained, based on Eq. (9):

$$\text{Maximum of}\left\{Raz^4\left(\frac{\partial c_0}{\partial z}\right)\right\}=1708, \tag{28}$$

which is satisfied by $(\partial c_0/\partial z)_\tau=(1/\sqrt{\pi\tau})\exp(-z^2/4\tau)$ at $z_{max}=2\sqrt{2\tau}$ from Eq. (24). This results in $\tau_o=49.7Ra^{-2/3}$ independently of Sc. Therefore, Eq. (28) seems to correspond to the lower bound of critical Rayleigh number, wherein the temperature profile assumed to be linear within $z=z_{max}$. It is well known that for deep-pool systems the boundary conditions on the upper boundary have no effects on the critical conditions. However, the critical condition from this model is closely related with the boundary conditions on the upper boundary. In spite of the above drawbacks such as the dependency of Sc and the boundary conditions on the critical conditions, common physics is involved in the above results: Ra^*~constant as shown in Eq. (25). From the above, it may be stressed that the propagation theory is a simple and physically realistic method.

Foster [17] commented that with correct dimensional relations $\tau_o\cong4\tau_c$. This means that the fastest growing mode of instabilities, which sets in at $t=t_c$, will grow with time until manifest convection is near the whole bottom boundary detected at $t\cong4t_c$. This relation is kept for the various transient diffusive systems [10-13]. It seems evident that convective motion is very weak during $t_c\le t\le t_o$ since the related heat transport is well represented by the conduction state. For a silicon (solute)-indium (solvent) LPE system of Sc=15.7, by using computational fluid dynamic method, Saito and Motoyama [6] and Coskun et al. [7] determine numerically the onset time of solutal convection as the time when the mean dissolution rate or the mean dissolution depth of the lower substrate deviate from those of upper substrate. Their results are compared with the above theoretical results in Fig. 4. As shown in this figure, the theoretical results underestimate the onset time of buoyancy-driven convection. It seems that the instability motion set in at $t=t_c$ is very weak and, therefore, it should grow to enhance the dissolution rate. However, the trend of Ra^*~constant is kept in deep-pool region of small τ. From the above, it may be concluded that the dissolution period should be larger than $4\tau_c$ for the growth of the epitaxy layer by using the LPE process. To validate the theoretical analysis, the predictions of τ_c should be compared with experimental observations. Unfortunately, no experimental value is available now, so systematic experiments are necessary and opened.

CONCLUSION

The onset of a buoyancy-driven convection in LPE system has been analyzed by propagation theory. The present stability criterion

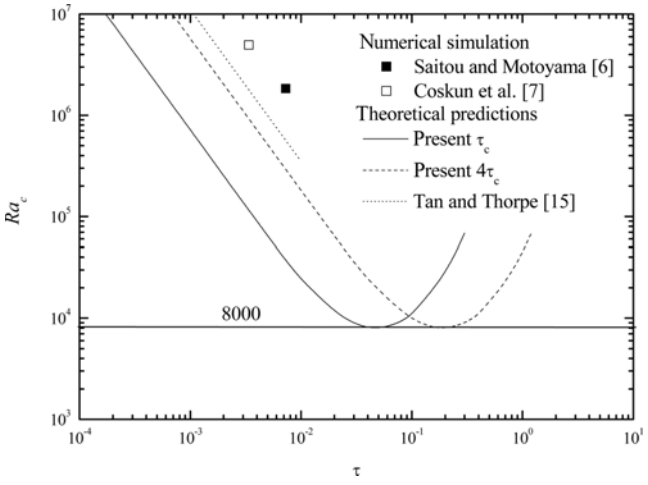

Fig. 4. Comparison of the stability results with the previous results.

bound reasonably well extant numerical simulation results. It seems that for deep-pool systems manifest convection is detected at τ_o, and for $\tau \leq \tau_o$ velocity disturbances are too weak to be observable experimentally. The present critical Rayleigh number (Ra_c=8000) is quite different from that of the conventional Bénard-Rayleigh problem (Ra_c=1708). The present results may give useful information on the design and operation of the LPE systems.

NOMENCLATURE

a : dimensionless wavenumber
C : concentration
c : dimensionless concentration disturbance, $g\gamma d^3 C_1/(\alpha_s \nu)$
c_0 : dimensionless basic concentration, C_0/C_w
d : distance between the substrates
g : gravitational acceleration
P : pressure
Ra : Rayleigh number, $g\gamma C_w d^3/(\alpha_s \nu)$
Sc : Schmidt number, ν/α_s
t : time
(U, V, W) : velocities in Cartesian coordinates
(u, v, w) : dimensionless velocity disturbances in Cartesian coordinates
(X, Y, Z) : Cartesian coordinates
(x, y, z) : dimensionless Cartesian coordinates

Greek Letters

α_s : diffusivity
Δ_C : penetration depth
δ_C : dimensionless penetration depth, Δ_C/d
γ : solutal expansion coefficient
ν : kinematic viscosity
τ : dimensionless time, $\alpha_s t/d^2$
ζ : similarity variable, $z/\sqrt{\tau}$

Subscripts

c : critical conditions
0 : basic quantities
1 : perturbed quantities

Superscript

* : transformed quantities

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