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Onset of marangoni convection in an initially quiescent spherical droplet subjected to the transient heat conduction

Min Chan Kim*,†, Do-Young Yoon, and Eunsu Cho*****

*Department of Chemical Engineering, Jeju National University, Jeju 690-756, Korea
**Department of Chemical Engineering, Kwangwoon University, Seoul 139-701, Korea
***Bead & Micro, Inc., Yongin-si, Gyeonggi-do 449-923, Korea
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Abstract−The onset of Marangoni convection in an initially quiescent spherical droplet subjected to the impulsive change in boundary temperature is analyzed under the linear theory. For this system in which instabilities are driven by interface-tension gradients, a stability analysis on regular cell modes is conducted on the basis of the propagation theory we have developed. The present stability analysis predicts that τ_c decreases with increasing Ma. For the limiting case of $\tau\rightarrow 0$, the present study approaches the planar limit as expected.

Key words: Marangoni Convection, Spherical Droplet, Propagation Theory

INTRODUCTION

The convective stability of fluids bounded by a free surface has been studied extensively in connection with Marangoni convection. Excellent illustrations on this subject are given by Pearson [1], Scriven and Sterling [2], Berg et al. [3] and Smith and Davis [4]. Recently, the thermal instability in a single droplet has attracted considerable interest because of its importance in the conventional process such as extraction and in the process under the microgravity condition [5-8].

When heat/mass is transferred through the interface of an initially quiescent liquid droplet, interfacial-tension gradients can often generate Marangoni convection when the Marangoni number exceeds a certain critical value. The stability problem in a liquid with temperature-dependent surface tension was analyzed first by Pearson [1] in planar geometry and by Pirotte and Lebon [5] in the spherical shell. Even though their analyses are indeed appropriate for describing convection with time-independent temperature profiles, its applicability to nonlinear temperature systems of rapid cooling is clouded due to the inherent complexity of time-varying temperature profiles. For the thin liquid layer, the related instability analysis has been conducted by using the quasi-static approximation [9], propagation theory [10,11], energy method [12] and maximum-transient-Marangoni-number criterion [13,14]. And, for the spherical droplet, the quasi-static approximation [15] and energy method [16,17] have been employed. The quasi-static approximation is based on linear theory and yields the critical time as the parameter, and the resulting critical conditions are independent of the Prandtl number Pr. It is well-known that the energy method suggested lower bounds on the experimental onset times.

We have developed the propagation theory to effectively yield that elusive critical condition to mark thermal instability under the developing temperature fields in planar geometry. This theory uses the thermal penetration depth as a length scaling factor and transforms the linearized perturbation equations similarly under the principle of exchange of stabilities. Its core concept is that the most dangerous disturbances will set in, experiencing instantaneous variations in their values upon their onset. The resulting stability criteria have compared well with experimental data in Rayleigh-Bénard-Marangoni convection [10,11,18,19]. Also, a similar approach is found to be successful in cases of Soret-driven convection [20,21] and onset of Taylor-Görtler vortices [22-24]. Hence, the stability analysis based on this theory is extended to the above problem on the onset of Marangoni convection in evaporating liquid layers and its validity is discussed in comparison with extant experimental and theoretical results.

Here we will concentrate on the instability problem in an initially isothermal, quiescent fluid droplet. Starting from time t=0, the outer free surface is cooled uniformly by evaporation. For this specific system, the interfacial-tension-driven instability criteria will be obtained by using the frozen-time model and the propagation theory. This study may be the extension of Pirotte and Lebon's [5] and Kim et al.'s [25] work.

THEORETICAL ANALYSIS

1. Governing Equations

The system considered here is a spherical Newtonian fluid droplet with an initial temperature T_i. For time t≥0, the spherical drop of radius R experiences impulsive cooling with heat flux q_w through the interface. The heat flux q_w is assumed to be constant. The schematic diagram of the basic system of pure conduction is shown in Fig. 1. For a high q_w, interfacial tension-driven convection will set in at a certain time and the governing equations of flow and temperature fields are expressed as

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

$$\left(\frac{\partial}{\partial t}+\mathbf{U}\cdot\nabla\right)\mathbf{U}=-\frac{1}{\rho}\nabla P+\nu\nabla^2\mathbf{U}, \tag{2}$$

†To whom correspondence should be addressed.
E-mail: mckim@cheju.ac.kr

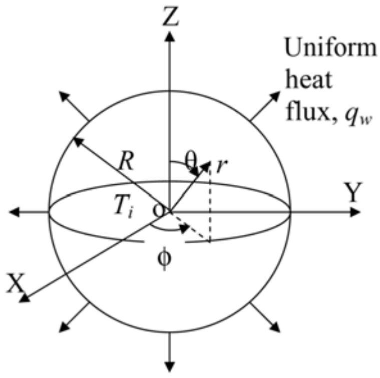

Fig. 1. The schematic diagram of system considered here.

$$\frac{\partial \mathrm{T}}{\partial \mathrm{t}}+(\mathbf{U}\cdot\nabla)\mathrm{T}=\alpha\nabla^2\mathrm{T}, \tag{3}$$

where **U**, P, ρ, ν and α represent the velocity vector, the dynamic pressure, the density the kinematic viscosity and the thermal diffusivity, respectively. The distance has the scale of R, the velocity that of α/R and the pressure that of $\alpha\mu/\mathrm{R}^2$. The surface temperature T_s at the spherical surface r=R decreases with time during the conduction period.

The important parameters to describe the present system are the Prandtl number (Pr) and the Marangoni number (Ma) defined by

$$\mathrm{Pr}=\frac{\nu}{\alpha}\text{ and }\mathrm{Ma}=\frac{\gamma q_w \mathrm{R}^2}{\mathrm{k}\alpha\mu},$$

where μ and k represent the viscosity and the thermal conductivity, respectively. The surface tension S is assumed to decrease with the temperature at the constant rate $\gamma[=-(1/\mathrm{S})(\mathrm{dS/dT})]$.

For the basic state of conduction the dimensionless temperature profile is represented by

$$\frac{\partial\Theta_0}{\partial\tau}=\frac{1}{\bar{\mathrm{r}}^2}\frac{\partial}{\partial\bar{\mathrm{r}}}\left(\bar{\mathrm{r}}^2\frac{\partial\Theta_0}{\partial\bar{\mathrm{r}}}\right), \tag{4}$$

with the following initial and boundary conditions,

$$\Theta_0=0 \quad \text{at } \tau=0, \tag{5a}$$
$$\Theta_0=\text{finite} \quad \text{at } \bar{\mathrm{r}}=0, \tag{5b}$$
$$\frac{\partial\Theta_0}{\partial\bar{\mathrm{r}}}=-1 \quad \text{at } \bar{\mathrm{r}}=1. \tag{5c}$$

In the above equations, $\tau=\alpha t/\mathrm{R}^2$, $\bar{\mathrm{r}}$=r/R and $\Theta_0=\mathrm{k}(\mathrm{T}-\mathrm{T}_i)/\mathrm{q}_w\mathrm{d}$. The subscript "0" denotes the basic state. Carslaw and Jaeger [26] and Crank [27] give the exact solutions to the basic temperature field within the drop as:

$$\Theta_0=-3\tau-\frac{\bar{\mathrm{r}}^2}{2}+\frac{3}{10}+\frac{2}{\bar{\mathrm{r}}}\sum_{n=1}^{\infty}\frac{\sin\alpha_n\bar{\mathrm{r}}}{\alpha_n^2\sin\alpha_n}\exp(-\alpha_n^2\tau), \tag{6a}$$

here α_n is the positive roots of

$$\alpha_n\cot\alpha_n-1=0. \tag{6b}$$

For the limiting case of large τ, the basic temperature field approaches $\Theta_0+3\tau=0.3-\bar{\mathrm{r}}^2/2$.

For the case of small τ, by using the Laplace transform method, the above Eqs. (4) and (5) can be solved as

$$\begin{aligned}\Theta_0=-\frac{\sqrt{\tau}}{\bar{\mathrm{r}}}\sum_{n=0}^{\infty}(-1)^n&\left[\frac{2}{\sqrt{\pi}}\exp\left\{-\frac{(1-\bar{\mathrm{r}}+2\mathrm{n})^2}{4\tau}\right\}\right.\\&-\frac{(1-\bar{\mathrm{r}}+2\mathrm{n})}{\sqrt{\tau}}\mathrm{erfc}\left\{\frac{(1-\bar{\mathrm{r}}+2\mathrm{n})}{2\sqrt{\tau}}\right\}-\frac{2}{\sqrt{\pi}}\exp\left\{-\frac{(1+\bar{\mathrm{r}}+2\mathrm{n})^2}{4\tau}\right\}\\&\left.+\frac{(1+\bar{\mathrm{r}}+2\mathrm{n})}{\sqrt{\tau}}\mathrm{erfc}\left\{\frac{(1+\bar{\mathrm{r}}+2\mathrm{n})}{2\sqrt{\tau}}\right\}\right]\end{aligned} \tag{7}$$

For the limiting case of $\tau\to0$, the above exact solution can be approximated as

$$\Theta_0=-\sqrt{\tau}\left[\frac{2}{\sqrt{\pi}}\exp\left\{-\frac{\zeta^2}{4}\right\}-\zeta\,\mathrm{erfc}\left\{\frac{\zeta}{2}\right\}\right], \tag{8}$$

where $\zeta=(1-\bar{\mathrm{r}})/\sqrt{\tau}$. Since erfc(∞)=0, the terms in the series of Eq. (7), except the first two ones, are zero for this limiting case of $\tau\to0$. The above approximation of Eq. (8) is in good agreement with the exact solutions of Eq. (6) in the region of $\tau\le10^{-3}$, as shown in Fig. 2. It is noted that the interface temperature, $\Theta_{0,s}$ is $-\sqrt{4\tau/\pi}$ for small τ.

2. Stability Equations

Under linear stability theory, infinitesimal disturbances caused by incipient convective motion at the dimensionless critical time τ_c can be formulated, in dimensionless form by linearizing Eqs. (1)-(3):

$$\nabla^2\left[\nabla^2-\frac{1}{\mathrm{Pr}}\frac{\partial}{\partial\tau}\right](\bar{\mathrm{r}}\mathrm{u}_r)=0, \tag{9}$$

$$\frac{\partial\Theta_1}{\partial\tau}+\mathrm{Ma}l(l+1)\mathrm{u}_r\frac{\partial\Theta_0}{\partial\mathrm{r}}=\nabla^2\Theta_1. \tag{10}$$

Here the velocity component has the scale of α/R and the temperature component has that of $\alpha\mu/(\gamma\mathrm{R}l(l+1))$, where l is spherical wave number which will be disscused later. The detailed procedure can be found in Pirotte and Lebon's [5] work. The proper boundary conditions are given by [5]

$$\mathrm{w}=\frac{\partial\mathrm{w}}{\partial\bar{\mathrm{r}}}=\Theta_1=0\text{ at }\bar{\mathrm{r}}=0, \tag{11a}$$

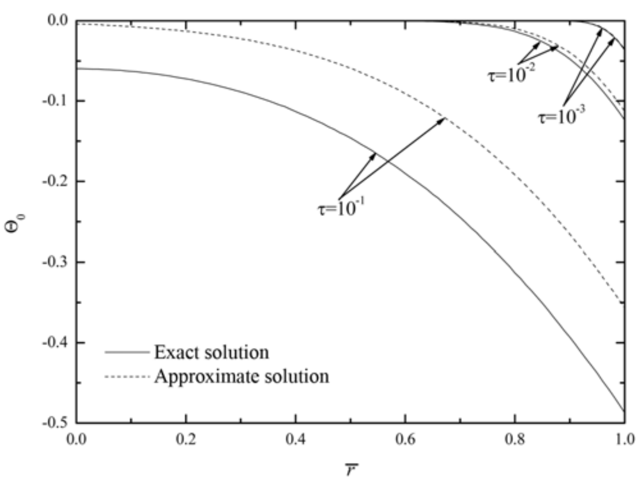

Fig. 2. Basic temperature profiles.

$$\mathrm{w}=\frac{\partial \Theta_1}{\partial \bar{\mathrm{r}}}=0 \text{ and } \frac{1}{\bar{\mathrm{r}}}\frac{\partial^2 \mathrm{w}}{\partial \bar{\mathrm{r}}^2}=\nabla_1^2 \Theta_1 \text{ at } \bar{\mathrm{r}}=1, \tag{11b}$$

where $\mathrm{w}=\bar{\mathrm{r}}\mathrm{u}_r$. The impermeable, q_w=constant($(\partial\Theta_1/\partial\bar{\mathrm{r}})=0$) and the force balance including interfacial tension effect are applied to the outer surface and the finite conditons are applied at the center of the droplet. Here,

$$\nabla_1^2=\frac{1}{\bar{\mathrm{r}}^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{\bar{\mathrm{r}}^2\sin^2\theta}\frac{\partial^2}{\partial\phi^2}=\nabla^2-\frac{1}{\bar{\mathrm{r}}^2}\frac{\partial}{\partial\bar{\mathrm{r}}}\left(\bar{\mathrm{r}}^2\frac{\partial}{\partial\bar{\mathrm{r}}}\right). \tag{12}$$

Now, convective motion is assumed to exhibit periodicity and the following normal mode analysis is employed:

$$[\mathrm{w}, \Theta_1]=[\mathrm{w}(\bar{\mathrm{r}}, \tau), \Theta(\bar{\mathrm{r}}, \tau)]\mathrm{Y}_l^m(\theta, \phi). \tag{13}$$

The spherical harmonics Y_l^m satisfies the following relation:

$$\nabla_1^2\mathrm{Y}_l^m=-\frac{l(l+1)}{\bar{\mathrm{r}}^2}\mathrm{Y}_l^m,\ \ l=0,1,2,\ldots,\ \ \mathrm{m}=-l,-l+1,\cdots l-1,l, \tag{14}$$

where

$$\mathrm{Y}_l^m(\theta, \phi)=\mathrm{P}_l^m(\cos\theta)\exp(\mathrm{i}m\phi), \tag{15}$$

with $\mathrm{P}_l^m(\cos\theta)$ being the associated Legendre polynomials. Therefore, the stability equations are summarized as:

$$\left[\frac{1}{\bar{\mathrm{r}}^2}\frac{\partial}{\partial\bar{\mathrm{r}}}\left(\bar{\mathrm{r}}^2\frac{\partial}{\partial\bar{\mathrm{r}}}\right)-\frac{l(l+1)}{\bar{\mathrm{r}}^2}-\frac{1}{\mathrm{Pr}}\frac{\partial}{\partial\tau}\right]\left[\frac{1}{\bar{\mathrm{r}}^2}\frac{\partial}{\partial\bar{\mathrm{r}}}\left(\bar{\mathrm{r}}^2\frac{\partial}{\partial\bar{\mathrm{r}}}\right)-\frac{l(l+1)}{\bar{\mathrm{r}}^2}\right]\mathrm{w}=0, \tag{16}$$

$$\left[\frac{1}{\bar{\mathrm{r}}^2}\frac{\partial}{\partial\bar{\mathrm{r}}}\left(\bar{\mathrm{r}}^2\frac{\partial}{\partial\bar{\mathrm{r}}}\right)-\frac{l(l+1)}{\bar{\mathrm{r}}^2}-\frac{\partial}{\partial\tau}\right]\Theta=\mathrm{Ma}l(l+1)\frac{\mathrm{w}}{\mathrm{r}}\frac{\partial\Theta_0}{\partial\bar{\mathrm{r}}}. \tag{17}$$

under the following boundary conditions:

$$\mathrm{w}=\frac{\partial \mathrm{w}}{\partial\bar{\mathrm{r}}}=\Theta=0 \text{ at } \bar{\mathrm{r}}=0, \tag{18a}$$

$$\mathrm{w}=\frac{\partial\Theta}{\partial\bar{\mathrm{r}}}=0 \text{ and } \frac{\partial^2\mathrm{w}}{\partial\bar{\mathrm{r}}^2}=\frac{1}{\bar{\mathrm{r}}}\Theta \text{ at } \bar{\mathrm{r}}=1. \tag{18b}$$

For a given Pr, the critical time τ_c should be found by using Eqs. (16)-(18). Here, propagation theory is employed to find the onset time of convective motion, i.e., the critical time t_c is based on the assumption that in deep-pool systems of $\tau\rightarrow0$ the infinitesimal temperature disturbances are propagated mainly within the thermal penetration depth at the onset of convective motion $\Delta_T(\propto\sqrt{\alpha t})$ and the following scale relations are valid for perturbed quantities from Eqs. (10) and (11):

$$\mu\frac{\mathrm{W}}{\Delta_T^2}\sim\frac{l(l+1)\gamma\mathrm{T}_1}{\mathrm{R}}, \tag{19a}$$

$$\frac{\mathrm{W}}{\mathrm{R}}\frac{\partial\mathrm{T}_0}{\partial\mathrm{r}}\sim\alpha\frac{\mathrm{T}_1}{\Delta_T^2}, \tag{19b}$$

from the balance between viscous and interfacial-tension terms in Eq. (10a) and also from that among terms in Eq. (3). Now, based on relation of Eq. (19a), the following amplitude relation is obtained in dimensionless form:

$$\frac{\mathrm{w}}{\Theta}\sim\delta_T^2\sim\tau, \tag{20}$$

$$\mathrm{Ma}^*l^*(l^*+\sqrt{\tau})\frac{\partial\Theta_0}{\partial\bar{\mathrm{r}}}\sim1, \tag{21}$$

where $\delta_T(=\Delta_T/\mathrm{R})\sim\sqrt{\tau}$, $\mathrm{Ma}^*=\mathrm{Ma}\tau$, and $l^*=l\sqrt{\tau}$. The above scaling analysis is described in depth in Kang and Choi [7] and Kim et al. [25]. From above, there are many possible forms of dimensionless amplitude functions of disturbances like

$$[\mathrm{w}(\tau, \bar{\mathrm{r}}), \Theta(\tau, \bar{\mathrm{r}})]=[\tau^{n+1}\mathrm{w}^*(\tau, \bar{\mathrm{r}}), \tau^n\Theta^*(\tau, \bar{\mathrm{r}})], \tag{22}$$

which satisfy the relation of Eq. (19).

Based on the relative condition of Chen et al. [28], Kim et al. [25] suggested that for the case of n=0.5, the relative instability condition be fullfiled at $\tau=\tau_c$. If the related process is still diffusion dominant with Ma^*=constant at $\tau=\tau_c$, it is probable that $\Theta(\tau, \bar{\mathrm{r}})=\tau^{1/2}\Theta^*(\zeta)$. This means that the amplitude function of temperature disturbances follows the behavior of Θ_0 for small τ. Furthermore, the relation of Ma^*=constant is shown even in theoretical results from the frozen-time model [9] and the energy method [12].

By the above scaling reasoning we set $\mathrm{w}_1^*=\tau^{3/2}\mathrm{w}^*(\zeta)$ and $\Theta_1^*=\tau^{1/2}\Theta^*(\zeta)$. For a deep-pool system of $\delta_T\propto\sqrt{\tau}$, the dimensionless time τ is related with the time for the development of penetration depth, so it plays dual roles of time and length. Now, the self-similar stability equations are obtained from Eqs. (16) and (17) as

$$\begin{aligned}&\left(\mathrm{D}^2-\frac{2\sqrt{\tau}}{(1-\zeta\sqrt{\tau})}\mathrm{D}-\frac{l^*(l^*+\sqrt{\tau})}{(1-\zeta\sqrt{\tau})^2}\right)^2\mathrm{w}^*+\frac{1}{2\mathrm{Pr}}\left(\zeta\mathrm{D}^3-\mathrm{D}^2+\frac{4\sqrt{\tau}}{(1-\zeta\sqrt{\tau})}\mathrm{D}\right.\\&\left.-\frac{4\sqrt{\tau}}{(1-\zeta\sqrt{\tau})}\zeta\mathrm{D}^2+3\frac{l^*(l^*+\sqrt{\tau})}{(1-\zeta\sqrt{\tau})^2}-\frac{l^*(l^*+\sqrt{\tau})}{(1-\zeta\sqrt{\tau})^2}\zeta\mathrm{D}\right)\mathrm{w}^*=0,\end{aligned} \tag{23}$$

$$\begin{aligned}&\left(\mathrm{D}^2-\frac{2\sqrt{\tau}}{(1-\zeta\sqrt{\tau})}\mathrm{D}-\frac{l^*(l^*+\sqrt{\tau})}{(1-\zeta\sqrt{\tau})^2}+\frac{\zeta}{2}-\frac{1}{2}\right)\Theta^*\\&=\mathrm{Ma}^*l^*(l^*+\sqrt{\tau})\frac{\mathrm{w}^*}{(1-\zeta\sqrt{\tau})}\mathrm{D}\Theta_0,\end{aligned} \tag{24}$$

where $\mathrm{D}=d/d\zeta$. The proper boundary conditions are

$$\mathrm{w}^*=\mathrm{D}\Theta^*=\mathrm{D}^2\mathrm{w}^*-\Theta^*=0 \text{ at } \zeta=0, \tag{25a}$$

$$\mathrm{w}^*=\mathrm{D}\mathrm{w}^*=\Theta^*=0 \text{ as } \zeta\rightarrow\infty. \tag{25b}$$

For a given Pr, Ma^* and l^* are treated as eigenvalues, and the minimum value of Ma^* should be found in the plot of Ma^* vs l^* under the principle of exchange of stabilities. In other words, the minimum value of τ, τ_c, and its corresponding wavenumber l_c are obtained for a given Pr and Ma. Since time is frozen by letting $\partial(\cdot)/\partial\tau\equiv0$ within the frame of amplitude coordinates τ and ζ instead of τ and $\bar{\mathrm{r}}$ (see Eqs. (9) and (10)), propagation theory may be called the relaxed frozen-time model by implicitly treating τ_c as the parameter, but it considers the time dependency. For the limiting case of $\tau\rightarrow0$, the above stability equations of Eqs. (22) and (23) are reduced as:

$$\left[(\mathrm{D}^2-\mathrm{a}^{*2})^2+\frac{1}{2\mathrm{Pr}}(\zeta\mathrm{D}^3-\mathrm{D}^2+3\mathrm{a}^{*2}-\mathrm{a}^{*2}\zeta\mathrm{D})\right]\mathrm{w}^*=0, \tag{26}$$

$$\left(\mathrm{D}^2-\mathrm{a}^{*2}+\frac{\zeta}{2}-\frac{1}{2}\right)\Theta^*=\mathrm{Ma}^*\mathrm{a}^{*2}\mathrm{D}\Theta_0\mathrm{w}^*, \tag{27}$$

which are idendtical with those for the planar geometry (see Eqs. (34) and (35) of Kim et al.'s [25]). Here $\mathrm{a}^{*2}=l^*(l^*+\sqrt{\tau})$. However, this stability equation is slightly different from Kang and Choi's [10] work, where n=0 is used in Eq. (21) without any justification.

The conventional frozen-time model neglects the terms involving $\partial(\cdot)/\partial\tau$ in Eqs. (15) and (16) in amplitude coordinates τ and $\bar{\mathrm{r}}$. This results in

$$\left(\mathrm{D}^2-\frac{2\sqrt{\tau}}{(1-\zeta\sqrt{\tau})}\mathrm{D}-\frac{l^*(l^*+\sqrt{\tau})}{(1-\zeta\sqrt{\tau})^2}\right)^2\mathrm{w}^*=0, \tag{28}$$

$$\left(\mathrm{D}^2-\frac{2\sqrt{\tau}}{(1-\zeta\sqrt{\tau})}\mathrm{D}-\frac{l^*(l^*+\sqrt{\tau})}{(1-\zeta\sqrt{\tau})^2}\right)\Theta^* = \mathrm{Ma}^* l^*(l^*+\sqrt{\tau})\frac{\mathrm{w}^*}{(1-\zeta\sqrt{\tau})}\mathrm{D}\Theta_0, \tag{29}$$

instead of Eqs. (19) and (20). The resulting stability criteria become independent of Pr and τ_c is obtained for a given Ma. By employing the frozen-time model, for planar geometry Vidal and Acrivos [9] obtained the following assymtotic relations for the domain of small τ,

$$\mathrm{Ma}\,\tau_c=2 \text{ and } \mathrm{a}\sqrt{\tau_c}=0 \text{ for } \tau_c\to 0, \tag{30}$$

which can be applied for the present spherical geometry for $\tau_c\to 0$. In the spherical geometry, the spherical wavenumber l should be used, instead of the horizontal wavenumber a.

3. Solution Procedure

The stability Eqs. (23)-(25) are solved by employing the outward shooting scheme. To integrate these stability equations the proper values of Dw^*, $\mathrm{D}^3\mathrm{w}^*$ and Θ^* at $\zeta=0$ are assumed for a given Pr and l^*. Since the stability equations and their boundary conditions are all homogeneous, the value of $\mathrm{Dw}^*(0)$ can be assigned arbitrarily and the value of the parameter Ma^* 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 be proceeded numerically.

Integration is performed from $\zeta=0$ to a fictitious upper boundary with the fourth order Runge-Kutta-Gill method. If the guessed value of Ma^*, $\mathrm{D}^3\mathrm{w}^*(0)$ and $\Theta^*(0)$ are correct, w^*, Dw^* and Θ^* will vanish at the lower isothermal boundary. To improve the initial guesses the Newton-Raphson iteration is used. When convergence is achieved, the upper boundary for computation is increased by a predetermined value and the above procedure is repeated. Since the temperature disturbances decay exponentially outside the thermal penetration depth, the incremental change of Ma^* also decays fast with increasing a fictitious upper boundary thickness. This behavior enables us to extrapolate the eigenvalue to the infinite depth. The marginal stability curve for $\mathrm{Pr}\to\infty$ is shown in Fig. 3. In this limiting case, the governing equations are reduced to simpler forms because the inertia terms involving Pr in Eq. (22) are negligible.

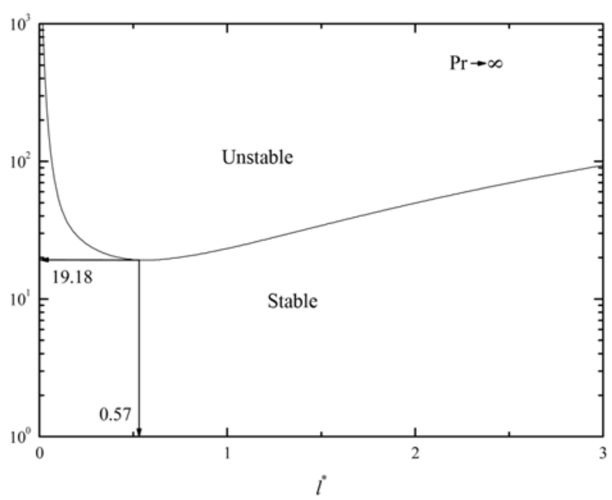

Fig. 3. Marginal stability curve for deep-pool systems of $\mathrm{Pr}\to\infty$ under the propagation theory.

DISCUSSION

The critical conditions predicted by propagation theory for a deep-pool system of $\tau\to 0$ are identical with those for the planar geometry of Kim et al. [25]. According to their results, the critical conditions to mark the onset of convective motion are correlated as

$$\tau_c=19.18\left[1+\left(\frac{0.31}{\mathrm{Pr}}\right)^{0.65}\right]^{1/0.65}\mathrm{Ma}^{-1} \text{ for } \tau_c<0.01. \tag{31}$$

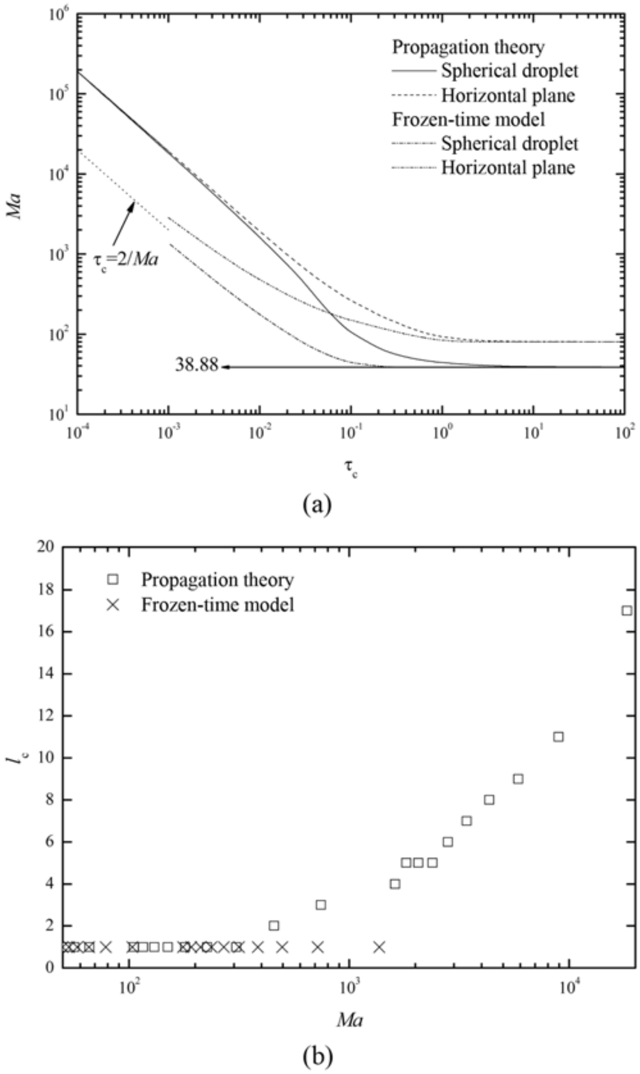

Fig. 4. Comparison of the critical conditions based on the theoretical methods. (a) critical time and (b) critical wavenumber.

within the error bound of 8%. It is believed that for a given Ma and Pr the fastest growing mode of infinitesimal disturbances would be set in at $t=t_c$ with $l=l_c$. The above equations show that τ_c decreases with an increase in Ma and also Pr. The Pr-effect becomes pronounced for Pr<1, which means the inertia terms make the system more stable. Since, τ_c is almost independent of Pr for Pr>10 and the mass transfer systems correspond to $Pr\rightarrow\infty$, the present study was conducted for the limiting case of $Pr\rightarrow\infty$.

Now, the domain of time is extended to $\tau_c>0.01$ by keeping Eqs. (23) and (24) and using Eq. (6a). In condition of Eq. (26b) the upper boundary $\zeta\rightarrow\infty$ is replaced with z=1, i.e., $\zeta=1/\sqrt{\tau_c}$ and in Eqs. (23) and (24) Ma^* and l^* are replaced with $Ma\tau_c$ and $l\sqrt{\tau_c}$. Since τ_c is the fixed parameter, the resulting stability equations are a function of ζ only and the physics of Eq. (20) is still alive. For a given Pr and τ_c the minimum Ma value and its corresponding wavenumber l_c are obtained. The solution procedure is almost the same as in the previous section. The results are summarized in Fig. 4, wherein those obtained from the conventional frozen-time model are also shown. The critical wavenumber l_c increases with increase of Ma. For $\tau_c<0.01$ the former ones are the same as those of the deep-pool system (Eq (31)). For large τ_c they approach the critical condition of $Ma_c^\infty=38.88$ since the basic temperature profile becomes parabolic. It is known that for small τ the frozen-time model yields the lower bounds of τ_c and the terms involving $\partial(\cdot)/\partial\tau$ in Eqs. (9) and (10) stabilize the system. It is interesting that propagation theory yields smoothly the stability criteria over the whole domain of time and the curvature effects are significant for the region of $\tau_c>>0.01$.

At the critical conditions illustrated above, the amplitude functions of w^* and θ^* are featured in Fig. 5, wherein the quantities have been normalized by the corresponding maximum magnitude w^*_{max} and q^*_{max}. It is seen that incipient temperature disturbances are confined mainly within the dimensionless thermal penetration. This implies that the inertia effect makes the velocity disturbances penetrate further into the fluid and therefore may stabilize the system.

In the present study, the time for the amplification of disturbances is not considered. Foster [29] commented that with correct dimensional relations the relation of $\tau_o\cong 4\tau_c$ would be kept for the case of a Rayleigh-Bénard problem. This relation of $\tau_o\cong 4\tau_c$ can be found in the various diffusive systems [10,11,18-25]. This means that the fastest growing mode of instabilities, which set in at $t=t_c$, will grow with time until manifest motion is first detected experimentally. Therefore, it seems evident that the predicted onset time t_c is smaller than the detection t_o. For the present system the Marangoni number based on the temperature difference, M, is $Ma\sqrt{4\tau/\pi}$, so the critical condition of Eq. (31) is rearranged as

$$\tau_c = 367.9\left(M\sqrt{\frac{\pi}{4}}\right)^{-2} \text{ for } \tau_c \leq 0.01 \text{ and } Pr\rightarrow\infty. \tag{32}$$

From Foster's comment ($\tau_o\cong 4\tau_c$), the following relation can be obtained as

$$\tau_o \cong 4\tau_c = 1471.6\left(M\sqrt{\frac{\pi}{4}}\right)^{-2} \text{ for } \tau_c \leq 0.01 \text{ and } Pr\rightarrow\infty. \tag{33}$$

Since the temperature difference increases continuously during the growth period ($\tau_c\leq\tau\leq\tau_o$), the detection time τ_o may be suggested as

$$\tau_o = 38.36 Ma^{-1} \text{ for } \tau_c \leq 0.01 \text{ and } Pr\rightarrow\infty. \tag{34}$$

where $Ma=M\sqrt{\pi/4\tau_o}$. A similar trend can be extended to $\tau_c>0.01$. This scenario is supported by the previous experimental and theoretical results for propanol (Pr=30) in planar geometry, as shown in Fig. 6. It seems evident that convective motion is very weak during $t_c\leq t\leq t_o$ since the related heat transport is well represented by the conduction state.

CONCLUSIONS

The critical condition to mark the onset of surface tension-driven motion in an initially quiescent, spherical droplet subjected to uniform cooling has been analyzed by using propagation theory. A new set of stability equations is derived under the scaling relation and relative instability concept. The resulting stability criteria compare reasonably well with previous theoretical predictions for the horizontal fluid layer. It is very interesting that propagation theory covers the whole domain of time and the limiting stability criterion of $\tau_c\geq$

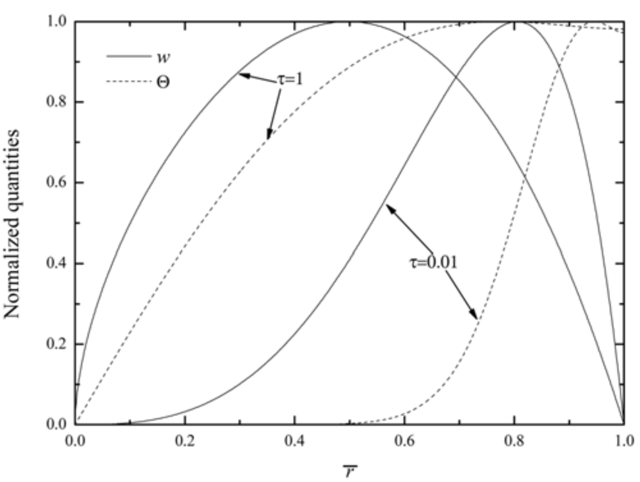

Fig. 5. Amplitude profiles of disturbances at the various critical conditions.

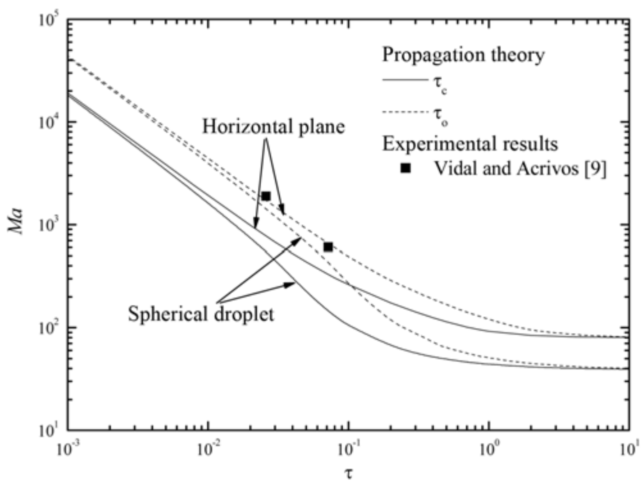

Fig. 6. Comparison of experimental result with the present theoretical predictions.

100 approaches Ma_c^{∞}=38.88.

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NOMENCLATURE

a : dimensionless horizontal wavenumber
D : differential operator, $d/d\zeta$
k : thermal conductivity [$Wm^{-1}K^{-1}$]
P : pressure [Pa]
Pr : Prandtl number, ν/α
R : radius of droplet [m]
q_w : wall heat flux [Wm^{-2}]
l : dimensionless spherical wave number
Ma : Marangoni number based on the heat flux, $\gamma q_w d^2/k\alpha\mu$
Ma^* : modified Marangoni number, $Ma\tau$
($\bar{r}$, θ, ϕ) : dimensionless spherical coordinates
T : temperature [K]
T_i : initial temperature [K]
T_s : surface temperature [K]
t : time [s]
U : velocity vector [ms^{-1}]
u_r : dimensionless radial velocity
w : dimensionless velocity, $\bar{r}u_r$

Greek Letters

α : thermal diffusivity [m^2s^{-1}]
γ : negative rate of change of surface tension [K^{-1}]
Δ_T : thermal penetration depth [m]
δ_T : dimensionless thermal penetration depth, Δ_T/d
Θ_1 : dimensionless temperature disturbance, $T_1\gamma d/(\alpha\nu)$
Θ_0 : dimensionless basic temperature, $k(T-T_i)/q_w d$
μ : viscosity [Pa s]
ν : kinematic viscosity [m^2s^{-1}]
ρ : density [kg m^{-3}]
τ : dimensionless time, $\alpha t/d^2$
ζ : similarity variable, $\zeta=(1-\bar{r})/\sqrt{\tau}$

Subscripts

c : critical state
i : initial state
o : observable
s : surface condition
0 : basic quantities
1 : disturbance quantities

Superscripts

* : amplitude functions

REFERENCES

1. J. R. A. Pearson, *J. Fluid Mech*., **4**, 489 (1958).
2. L. E. Scriven and C. V. Sternling, *J. Fluid Mech.*, **19**, 321 (1964).
3. J. C. Berg and A. Acrivos, *Adv. Chem. Eng*., **6**, 61 (1966).
4. M. K. Smith and S. H. Davis, *J. Fluid Mech.*, **132**, 119 (1983).
5. O. Pirotte and G. Lebon, *Appl. Microgravity Tech.*, **1**, 175 (1988).
6. H. C. Hoefsloot and H. W. Hoogstraten, *Appl. Microgravity Tech.*, **106**, 2 (1989).
7. A. Cloot and G. Lebon, *Microgravity Sci. Tech.*, **3**, 44 (1990).
8. S. K. Wilson, *J. Eng. Math*., **28**, 427 (1994).
9. A. Vidal and A. Acrivos, *I&EC Fund.*, **7**, 53 (1968).
10. K. H. Kang and C. K. Choi, *Phys. Fluids*, **9**, 7 (1998).
11. K. H. Kang, I. G. Hwang and C. K. Choi, *AIChE J.*, **46**, 15 (2000).
12. R. J. Gumerman and G. M. Homsy, *J. Fluid Mech*., **68**, 191 (1975).
13. K. K. Tan and R. B. Thorpe, *Chem. Eng. Sci*., **54**, 775 (1999).
14. K. K. Tan, *Int. J. Heat Mass Transfer.*, **48**, 135 (2005).
15. V. M. Ha and C. L. Lai, *Proc. R. Soc. London A*, **457**, 885 (2001).
16. J. F. Patzer and G. M. Homsy, *Phys. Fluids*, **24**, 567 (1981).
17. V. M. Ha and C. L. Lai, *Int. J. Heat Mass Transfer.*, **47**, 3811 (2004).
18. M. C. Kim, J. H. Park and C. K. Choi, *Chem. Eng. Sci.*, **60**, 5363 (2005).
19. C. K. Choi, J. H. Park and M. C. Kim, *Heat Mass Transfer.*, **41**, 155 (2004).
20. M. C. Kim, J. S. Hong and C. K. Choi, *AIChE J.*, **52**, 2333 (2006).
21. M. C. Kim, C. K. Choi and J. K. Yeo, *Phys. Fluids*, **19**, 084103 (2007).
22. M. C. Kim and C. K. Choi, *Korean J. Chem. Eng*., **26**, 874 (2006).
23. M. C. Kim, T. J. Chung and C. K. Choi, *Theoret. Comp. Fluid Dynamics*, **18**, 105 (2004).
24. M. C. Kim and C. K. Choi, *Chem. Eng. Sci*., **60**, 599 (2005).
25. M. C. Kim, C. K. Choi, D. Y. Yoon and T. J. Chung, *I&EC Res*., **46**, 5775 (2007).
26. H. S. Carslaw and J. C. Jaeger, *Conduction of heat in solids*, 2nd ed., Oxford Univ. Press, UK (1959).
27. J. Crank, *The mathematics of diffusion*, 2nd ed., Oxford Univ. Press, UK (1975).
28. J.-C. Chen, G. P. Neitzel and D. F. Jankowski, *Phys. Fluids*, **28**, 749 (1985).
29. T. D. Foster, *Phys. Fluids*, **12**, 2482 (1969).