Analysis of onset of Soret-driven convection by the energy method

Min Chan Kim[1,*] and Chang Kyun Choi[2]

[1]*Department of Chemical Engineering, Cheju National University, Cheju 690-756, Korea*

[2]*School of Chemical and Biological Engineering, Seoul National University, Seoul 151-744, Korea*

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The Soret-driven instability in binary mixture heated from above is analyzed by using the energy method and its modification. The horizontal fluid layer placed between two plates is in initially quiescent state but the Soret diffusion can induce buoyancy-driven convection in the case of the negative Soret coefficient. For the case of highly unstable density stratification the buoyancy-driven motion sets in during the transient diffusion stage. Here the stability limits which are related to the onset time of instabilities are presented as a function of the Rayleigh number Ra, the Lewis number Le, and the separation ratio ψ. The present stability analysis predicts that the onset time of convective instability decreases with increasing buoyancy parameter $\mathrm{Ra}(\mathrm{Le}/\psi)^{-1}$. The relaxed energy method shows that the first visible motion can be detected from a certain time five times larger than the predicted onset time and the critical wave number is not zero but a finite value.

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INTRODUCTION

Buoyancy-driven convection due to the thermal diffusion in multicomponent systems such as polymer solution, polymer blend, ethanol-water mixture, or various gas mixtures has attracted many researchers' interests since it shows quite different characteristics from those in pure fluids [1–4]. Recently the onset of Soret-driven instability (SDI) in nanoparticles-suspension systems has been studied intensively [5–9].

If suspension of nanoparticles is under consideration, the unexpected spatiotemporal properties of convection are observed due to the extremely small particle mobility. This is usually reflected by the small Lewis number $\mathrm{Le}(=D_C/\alpha)\leqslant 10^{-4}$, where D_C is the mass diffusion coefficient and α is the thermal diffusivity. The separation ratio $\psi[=(\beta_C/\beta_T)\times(D_T/D_C)]$ is used as the measure of the relative importance of the Soret effect with respect to weak solutal diffusion and it represents the coupling between the temperature and concentration fields [5–9]. Here $\beta_C[=\rho^{-1}(\partial\rho/\partial C)]$ is the solutal expansion coefficient, $\beta_T[=-\rho^{-1}(\partial\rho/\partial T)]$ the thermal one, D_T the thermal diffusion (Soret) coefficient, ρ density, C concentration, and T temperature. For binary mixture of negative ψ, the Soret effect can induce buoyancy-driven motion even in initially uniform concentration and thermally stable configuration. For the fully developed linear fields of temperature and concentration the critical Rayleigh number to represent the onset of SDI is given by the critical Rayleigh number $\mathrm{Ra}_c=720(\mathrm{Le}/\psi)$, considering the relative time scale of mass diffusion with respect to thermal diffusion [10]. Recently Shevtsova *et al.* [11] analyzed the onset of SDI by employing the computational fluid dynamics (CFD) technique.

In the present study the onset of SDI in the horizontal fluid layer heated from above, with large buoyancy forces driven by the Soret effect, i.e., $\mathrm{Ra}<0$ but $\mathrm{Ra}_s[=\mathrm{Ra}(\mathrm{Le}/\psi)^{-1}]\geqslant 720$, is investigated by using the conventional energy method and its modification. The energy method has a long history [12] and it is widely used in the hydrodynamic stability analysis [13]. This method gives the necessary condition of instability, i.e., a lower Ra_c bound. It does not require initial conditions for the disturbances, which are not known, and therefore, we concentrate this method. We relax the conventional strong stability criteria by introducing the relaxed stability criterion into the energy method. For the present problem the new stability equations are derived. The resulting predictions are compared with previous predictions based on the linear stability theory and also available experimental results.

*mckim@cheju.ac.kr

THEORETICAL ANALYSIS

Governing equations and base system

The problem considered here is a horizontal fluid layer confined between two rigid plates separated by the vertical distance d. The fluid layer of binary mixture, of which ψ has a large negative value, is initially quiescent at a constant concentration C_i and a constant temperature T_i. For time $t\geqslant 0$ the fluid layer is heated suddenly from above with a constant temperature T_u, that is, the Rayleigh number $\mathrm{Ra}[=g\beta_T(T_i-T_u)d^3/(\alpha\nu)]$ has a negative value. For a high Ra_s, buoyancy-driven convection will set in at a certain time and the governing equations of motion, temperature, and concentration fields are expressed by employing the Boussinesq approximation [14]

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

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

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

$$\left\{\frac{\partial}{\partial t}+\mathbf{U}\cdot\nabla\right\}C=D_C\nabla^2 C+D_T\nabla^2 T, \tag{4}$$

with the following initial and boundary conditions:

$$\mathbf{U}=\mathbf{0}, \quad T=T_i, \quad C=C_i \quad \text{at } t=0, \tag{5a}$$

$$\mathbf{U}=\mathbf{0}, \quad T=T_i, \quad D_C\frac{\partial C}{\partial Z}+D_T\frac{\partial T}{\partial Z}=0 \quad \text{at } Z=0, \tag{5b}$$

$$\mathbf{U}=\mathbf{0}, \quad T=T_u, \quad D_C\frac{\partial C}{\partial Z}+D_T\frac{\partial T}{\partial Z}=0 \quad \text{at } Z=d, \tag{5c}$$

where $\mathbf{U}[=(U,V,W)]$ is the velocity vector, P the dynamic pressure, ν the kinematic viscosity, and $\mathbf{g}$ the gravitational acceleration vector. The present system is thermally stable due to the heating from above, i.e., $T_u > T_i$ and therefore $\mathrm{Ra}<0$.

Under the stable temperature gradient, for the case of $D_T=0$, i.e., $\mathrm{Ra}_s=\psi=0$, the concentration field does not change and keeps the initial concentration $C=C_i$ from Eqs. (1)–(5). Therefore, the system is unconditionally stable when there is no Soret effect. However, for the case of $D_T<0$, i.e., $\psi<0$, the stable temperature gradient makes the solute move upward and the system be potentially unstable. In the present study the system of $\psi<0$, $\mathrm{Ra}<0$, and $\mathrm{Le}\to 0$ will be considered. For another extreme case of large positive ψ and small Le, binary mixtures such as ferrofluids heated from below with $\mathrm{Ra}>0$ was already analyzed by Ryskin *et al.* [10].

For the present system of $\psi<0$, $\mathrm{Ra}<0$, and $\mathrm{Le}\to 0$, if the temperature gradient is small, i.e., $\mathrm{Ra}_s<720$, the layer is not only initially stable but also in its equilibrium state. However, for the case of $\mathrm{Ra}_s>720$, while the layer is initially stable, it is unstable for a fully developed diffusive concentration profile, as summarized by Ryskin *et al.* [10]. Therefore, the convective motion occurs during the transient diffusion process and the related stability problem becomes transient. Its critical time t_c to mark the onset of buoyancy-driven motion is not fully understood. For this transient stability analysis we define a set of dimensionless variables τ, z, $\theta_0[=(T_i-T)/\Delta T]$ by using the scale of time d^2/D_C, length d, and temperature $\Delta T(=T_i-T_u)$. The heat conduction equation can be solved by using the separation of variables

$$\theta_0 = z + 2\sum_{n=1}^{\infty}\frac{(-1)^n}{n\pi}\sin(n\pi z)\exp\left(-\frac{n^2\pi^2}{\mathrm{Le}}\tau\right). \tag{6}$$

When the Lewis number is very small, e.g., $\mathrm{Le}\approx 10^{-4}$ for nanoparticles suspension systems, the basic temperature field can be approximated by

$$\theta_0 = z \quad \text{for } \tau \geqslant \mathrm{Le}. \tag{7}$$

This means that the establishment of the conductive heat profile during the time $0\leqslant\tau\leqslant\mathrm{Le}$ 0 will not be considered in the present study (see Fig. 1). In the experimental system of the nanopartles suspension in water (Cerbino *et al.* [6]), where $\mathrm{Le}=1.48\times10^{-4}$, $\psi=-3.41$, and $\mathrm{Sc}=3.7\times10^{4}$, the conduction time $t_{\mathrm{cond}}(=d^2/\alpha)$ is much shorter than the onset time of SDI t_c. For the case of Fig. 1 of Cerbino *et al.* [6], where $Rs=8.33\times10^{6}$ and $d=0.98$ mm, $t_{\mathrm{cond}}=6.5$ s, and $t_c=390$ s. For another experimental case of ethanol-water mixture system of La Porata and Surko [3], where $\mathrm{Le}=7.75\times10^{-3}$, $\psi=-0.24$, $\mathrm{Sc}=1.16\times10^{3}$ and $d=4$ mm, $t_{\mathrm{cond}}=124$ s,

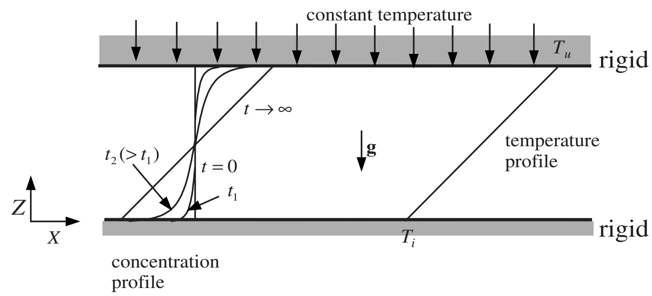

FIG. 1. Schematic diagram of the basic system considered here. The fully developed, linear temperature profile is assumed for $\tau>\mathrm{Le}$ in the present case of $\mathrm{Le}\to 0$.

and $t_c=1600$ s for $\mathrm{Ra}_s=1.6\times10^{5}$. From these experimental data, the transient thermal conduction effect might be negligible for the limiting case of $\mathrm{Le}\to 0$. Therefore, as shown by Ryskin and Muller [10] and Ryskin and Pleiner [9], the appropriate initial state to investigate the onset of convective instability is a state where the temperature profile is fully developed and linear, as given by Eq. (7) while concentration just starts to build up the layers near the boundaries. For the limiting case of $\mathrm{Le}\to 0$ and $\psi<0$, based on the above temperature distribution, the dimensionless concentration field is given by [14]

$$c_0(\tau,z) = z - \frac{1}{2} + \sum_{n=1}^{\infty}\frac{4}{\lambda_n^2}\cos\lambda_n z\exp(-\lambda_n^2\tau) \tag{8}$$

or

$$c_0(\tau,z) = \sqrt{4\tau}\sum_{n=0}^{\infty}\left\{-\operatorname{ierfc}\left(\frac{n}{\sqrt{\tau}}+\frac{z}{2\sqrt{\tau}}\right)-\operatorname{ierfc}\left(\frac{n+1}{\sqrt{\tau}}-\frac{z}{2\sqrt{\tau}}\right)+\operatorname{ierfc}\left(\frac{n+1/2}{\sqrt{\tau}}-\frac{z}{2\sqrt{\tau}}\right)+\operatorname{ierfc}\left(\frac{n+1/2}{\sqrt{\tau}}+\frac{z}{2\sqrt{\tau}}\right)\right\}, \tag{9}$$

where $c_0=D_C(C-C_i)/(j_S d)$, $j_S=D_T\Delta T/d$, $\lambda_n=(2n-1)\pi$ and ierfc is the integral of the complementary error function. For the case of negative ψ the Soret flux j_S has a positive value. The above concentration profiles satisfy the impermeable conditions for concentration at both boundaries. Based on these temperature and concentration fields given by Eqs. (7) and (9), the schematic diagram of the basic system of pure diffusion is shown in Fig. 1. For the deep-pool system of small $\tau(\mathrm{Le}\leqslant\tau\leqslant 0.01)$ and $\operatorname{ierfc}(\infty)=0$, the basic concentration field is approximated by

$$c_0 = -\sqrt{4\tau}\operatorname{ierfc}\left(\frac{z}{2\sqrt{\tau}}\right) \quad \text{for } z\leqslant 0.5. \tag{10}$$

Here $c_0(\tau,0)=-\sqrt{4\tau/\pi}$. Over a certain time interval the concentration gradient, which is propagating from both boundaries, does not feel the presence of the other one. Based on this, Shliomis and Souhar [5] assumed the concentration gradient as

$$\frac{\partial c_0}{\partial z} = \mathrm{erfc}\left(\frac{z}{2\sqrt{\tau}}\right). \tag{11}$$

Stability equations

Consider the following velocity, pressure, temperature, and concentration perturbations $\mathbf{U}_1=\mathbf{U}-\mathbf{U}_0$, $P_1=P-P_0$, $T_1=T-T_0$, and $C_1=C-C_0$. Introducing these perturbations into Eqs. (1)–(4), we can obtain the following dimensionless equations:

$$\nabla \cdot \mathbf{u}_1 = 0, \tag{12}$$

$$\frac{1}{\mathrm{Sc}}\left\{\frac{\partial}{\partial \tau} + \mathbf{u}_1 \cdot \nabla\right\}\mathbf{u}_1 = -\nabla p_1 + \nabla^2 \mathbf{u}_1 + \mathbf{k}\mathrm{Ra}_s(c_1 + \psi\theta_1), \tag{13}$$

$$\frac{\partial \theta_1}{\partial \tau} = \frac{1}{\mathrm{Le}}\nabla^2\theta_1 - w_1\frac{\partial \theta_0}{\partial z} - \mathbf{u}_1 \cdot \nabla\theta_1, \tag{14}$$

$$\frac{\partial c_1}{\partial \tau} = \nabla^2(c_1 - \theta_1) - w_1\frac{\partial c_0}{\partial z} - \mathbf{u}_1 \cdot \nabla c_1 \tag{15}$$

with the following boundary conditions:

$$\mathbf{u}_1 = \theta_1 = \frac{\partial c_1}{\partial z} = 0 \text{ at } z=0 \text{ and } z=1, \tag{16}$$

where $\mathrm{Sc}(=\nu/D_C)$ is the Schmidt number, velocity scale is D_C/d, and the subscripts 0 and 1 represent the base and the perturbation quantities, respectively. Here we retain the nonlinear convective term of $\mathbf{u}_1 \cdot \nabla c_1$, which are usually neglected in the linear stability theory.

For the limiting case of $\mathrm{Le}\to 0$, from Eqs. (14) and (16), the temperature perturbation has a trivial solution of $\theta_1=0$ and therefore the thermal effect can be negligible. For mixtures of large Sc the inertia terms having 1/Sc in Eq. (13) are also negligible. In this case the above perturbation equations reduce to

$$0 = -\nabla p_1 + \nabla^2\mathbf{u}_1 + \mathbf{k}\mathrm{Ra}_s c_1, \tag{17}$$

$$\frac{\partial c_1}{\partial \tau} = \nabla^2 c_1 - w_1\frac{\partial c_0}{\partial z} - \mathbf{u}_1 \cdot \nabla c_1, \tag{18}$$

with the following boundary conditions:

$$\mathbf{u}_1 = \frac{\partial c_1}{\partial z} = 0 \quad \text{at } z=0 \quad \text{and } z=1. \tag{19}$$

Now, we will follow the standard procedure of the conventional energy method [13]. By multiplying Eq. (17) by $\mathbf{u}_1$ and Eq. (18) by c_1 and integrating them over the system volume Ω, for the limiting case of $\mathrm{Le}\to 0$, the kinetic energy and the buoyancy energy relations can be obtained as

$$0 = -\int_\Omega \mathbf{u}_1 \cdot \nabla\left(p_1 + \frac{1}{2}\mathbf{u}_1^2\right)d\Omega + \int_\Omega \mathbf{u}_1 \cdot \nabla^2\mathbf{u}_1 d\Omega + \mathrm{Ra}_s\int_\Omega c_1 w_1 d\Omega, \tag{20}$$

$$\int_\Omega \frac{1}{2}\frac{\partial c_1^2}{\partial \tau}d\Omega = -\int_\Omega c_1\mathbf{u}_1 \nabla c_1 d\Omega + \int_\Omega c_1\nabla^2 c_1 d\Omega - \int_\Omega w_1 c_1\frac{\partial c_0}{\partial z}d\Omega. \tag{21}$$

Using the divergence theorem, the following relations are obtained:

$$0 = -\langle|\nabla \cdot \mathbf{u}_1|^2\rangle + R\langle w_1 c_1'\rangle, \tag{22}$$

$$\frac{1}{2}\frac{\partial\langle|c_1'|^2\rangle}{\partial \tau} = -\langle|\nabla c_1'|^2\rangle - R\left\langle w_1 c_1'\frac{\partial c_0}{\partial z}\right\rangle, \tag{23}$$

where $R=\sqrt{\mathrm{Ra}_s}$, $c_1'=\sqrt{\mathrm{Ra}_s}c_1$, and $\langle(\cdots)\rangle=\int_\Omega(\cdots)d\Omega$. In the above derivation, the boundary conditions (19) and the periodicity in the x and y direction are used.

In the present system the dimensionless energy identity is defined by adding Eqs. (22) and (23) with the coupling constant $\gamma>0$:

$$E(\tau) = \frac{1}{2}\gamma\langle c_1'\rangle^2. \tag{24}$$

This is connected to the kinetic energy [see Eqs. (13), (20), (22), and (23)], which is neglected for very large Sc. Now, the following equation is derived from Eqs. (22) and (23):

$$\frac{dE}{d\tau} = -\gamma\langle|\nabla c_1|^2\rangle - \gamma R\left\langle w_1\frac{\partial \theta_0}{\partial \zeta}c_1\right\rangle + R\langle w_1 c_1\rangle - \langle|\nabla \cdot \mathbf{u}_1|^2\rangle, \tag{25}$$

where the primes have been dropped. By setting $\hat{c}_1=\sqrt{\gamma}c_1$ the above equation is expressed as

$$\frac{d\hat{E}}{d\tau} = -\langle|\nabla\hat{c}_1|^2 + |\nabla \cdot \mathbf{u}_1|^2\rangle + R\left\langle w_1\frac{\hat{c}_1}{\sqrt{\gamma}} - w_1\frac{\partial c_0}{\partial z}\sqrt{\gamma}\hat{c}_1\right\rangle, \tag{26}$$

where $\hat{E}=\frac{1}{2}\langle\hat{c}_1\rangle^2$. The above relation can be represented as

$$\frac{dE}{d\tau} = RI - D = -D\left(1 - \frac{I}{D}R\right), \tag{27}$$

where

$$I = \left\langle w_1\frac{c_1}{\sqrt{\gamma}} - w_1\frac{\partial c_0}{\partial z}\sqrt{\gamma}c_1\right\rangle, \tag{28}$$

$$D = \langle|\nabla c_1|^2 + |\nabla \cdot \mathbf{u}_1|^2\rangle, \tag{29}$$

where the hats have been dropped. Under the strong instability concept of $dE/d\tau=0$, the onset time τ_s is defined as the critical time. In other words, for a given τ the strong stability limit R_s is determined as

$$\frac{1}{R_s} = \max\left(\frac{I}{D}\right). \tag{30}$$

From the procedure given in Straughan [13], under the normal mode analysis, this maximum problem can be reduced as the following Euler-Lagrange equations:

$$\left(\frac{\partial^2}{\partial z^2}-a^2\right)^2 w_1=-\frac{1}{2}R_s\left(\frac{1}{\sqrt{\gamma}}-\sqrt{\gamma}\frac{\partial c_0}{\partial z}\right)a^2 c_1, \tag{31}$$

$$\left(\frac{\partial^2}{\partial z^2}-a^2\right)c_1=\frac{1}{2}R_s\left(\frac{1}{\sqrt{\gamma}}-\sqrt{\gamma}\frac{\partial c_0}{\partial z}\right)w_1, \tag{32}$$

with the following boundary conditions:

$$w_1=\frac{\partial w_1}{\partial z}=\frac{\partial c_1}{\partial z}=0 \text{ at } z=0 \text{ and } 1, \tag{33}$$

where a is the horizontal wave number. The strong stability conditions are independent of the Schmidt number. The strong stability limit $\text{Ra}_s(\tau_s)$ is given by

$$\sqrt{\text{Ra}_s}=\max_{\gamma}\min_{a} R_s. \tag{34}$$

Now, the strong stability limits are relaxed. Under the relaxed stability concept the critical time τ_r is determined according to the following criterion [15]:

$$\frac{1}{E}\frac{dE}{d\tau}=\frac{1}{E_0}\frac{dE_0}{d\tau} \text{ at } \tau=\tau_r, \tag{35}$$

where E_0 is the basic energy identity, i.e., $E_0=\langle c_0^2\rangle/2$, since $\mathbf{u}_0=\mathbf{0}$. For the limiting case of $\tau\to 0$ [based on the concentration profile (10), $(1/E_0)(dE_0/d\tau)=3/2\tau$] the relaxed energy identity becomes

$$\frac{3E}{2\tau}=RI-D \text{ for } \tau\to 0. \tag{36}$$

The relaxed stability limit is obtained as

$$\frac{1}{R_r}=\max\left[\frac{I}{D+3E/2\tau}\right] \text{ for } \tau\to 0. \tag{37}$$

Under the normal mode analysis, for the limiting case of $\tau\to 0$, the Euler-Lagrange equations for the relaxed stability limit are obtained similar to Eqs. (31) and (32):

$$\left(\frac{\partial^2}{\partial z^2}-a^2\right)^2 w_1=-\frac{1}{2}R_r\left(\frac{1}{\sqrt{\gamma}}-\sqrt{\gamma}\frac{\partial c_0}{\partial z}\right)a^2 c_1, \tag{38}$$

$$\left(\frac{\partial^2}{\partial z^2}-a^2\right)c_1=\frac{1}{2}R_r\left(\frac{1}{\sqrt{\gamma}}-\sqrt{\gamma}\frac{\partial c_0}{\partial z}\right)w_1+\frac{3}{4\tau}c_1. \tag{39}$$

With the boundary conditions (32) the relaxed stability limit $Rs(\tau_r)$ is given by

$$\sqrt{\text{Ra}_s}=\max_{\gamma}\min_{a} R_r. \tag{40}$$

For systems of $\tau>0.01$, a similar approach can be applied and the terms containing $3/4\tau$ should be slightly modified. According to the basic concentration profile (8) $dE/d\tau$ can be written as

$$\frac{dE}{d\tau}=\left(\frac{1}{E_0}\frac{dE_0}{d\tau}\right)E = \frac{\sum_{n=1}^{\infty}(16/\lambda_n^2)\exp(-\lambda_n^2\tau)\{1-\exp(-\lambda_n^2\tau)\}}{1/12-\sum_{n=1}^{\infty}(8/\lambda_n^4)\exp(-\lambda_n^2\tau)\{2-\exp(-\lambda_n^2\tau)\}}E. \tag{41}$$

For the limiting case of $\tau\to 0$, from the above equation we obtain $(1/E_0)(dE_0/d\tau)=3/2\tau$. For another limiting case of $\tau\to\infty$, $(dE_0/d\tau)=0$ and the above stability equations (38) and (39) degenerate into the strong stability formulation [see Eqs. (31) and (32)]. Therefore, the present stability equations can cover the whole time region without any assumptions.

Solution method

The stability equations (38) and (39) were solved by employing the outward shooting scheme. In order to integrate them a trial value of the eigenvalue R_r and the boundary conditions $\partial^3 w_1/\partial z^3$ and c_1 at $z=0$ are assumed properly for a given a and γ. Since the boundary conditions (33) are all homogeneous, the value of $\partial^2 w_1/\partial z^2$ at $z=0$ 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, together with the three known conditions at the lower boundary, give all the information to make numerical integration smoothly.

The integration based on the fourth-order Runge-Kutta method is performed from $z=0$ to $z=1$. By using the Newton-Raphson iteration the trial values of R_r, $\partial^3 w_1/\partial z^3$, and c_1 are corrected until the stability equations satisfy the upper boundary conditions within the relative tolerance of 10^{-10}. For the strong stability limits the solution procedure is almost the same as above.

RESULTS AND DISCUSSIONS

For $\text{Le}\to 0$, the present stability criteria obtained from the energy method, strong and relaxed, are shown in Fig. 2 and they are also compared with the available predictions. They all show that for large τ the long wave instability of $a=0$ is the preferred mode and the critical condition is $\text{Ra}_s=720$ [10]. The relaxed stability limit yields the critical time:

$$\tau_r=3.16\text{Ra}_s^{-1/2} \text{ as } \text{Ra}_s\to\infty, \tag{42}$$

of which the constant is a little smaller than 5.57 from the propagation theory of Kim *et al.* [16]. Those from the strong stability limit and the frozen-time model are much smaller.

Very recently, Shevtsova *et al.* [11] conducted three-dimensional, time-dependent CFD analysis on the onset of SDI. They simulated the Soret-driven motion for the system of water (90%)-isopropanol (10%), where $\text{Le}=6.7\times 10^{-3}$ and $\psi=-0.4$ with the Prandtl number of 10.85. Their calculation condition is much closer to La Porta and Surko's [3] experimental one for the ethanol-water system than the present system of $\text{Le}\to 0$. Their stability criterion is

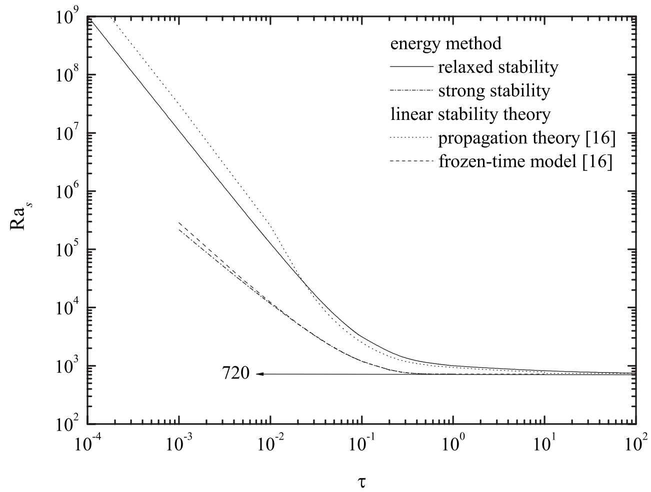

FIG. 2. Comparison of the predicted critical times for Le→0.

$$\tau_c = \frac{t_p}{t_D} = 1.05\mathrm{Ra}_s^{-0.52} \text{ for } \mathrm{Ra}_s > 3 \times 10^4, \qquad (43)$$

where t_p is the peak time where the vertical velocity attains the maximal value and $t_D(=d/D_C)$ is the diffusion time. Even though this criterion is quite lower than the present stability limit (42) for the present limiting case of Le→0, it shows the same scaling relation as the present prediction of $\tau_r \sim \mathrm{Ra}_s^{-1/2}$.

The present predictions are compared with available experimental data in Fig. 3. By using the shadowgraph method Cerbino *et al.* [6,7] and Mazzoni *et al.* [17] visualized the Soret-driven convective motion in a colloidal suspension of 22 nm diameter silica particles (LUDOX®) dispersed in water. Their experimental condition is that of $\mathrm{Le}=1.48\times10^{-4}$, $\psi=-3.41$, and $\mathrm{Sc}=3.7\times10^4$. They obtained the latency time τ^* and the peak time τ_p as the characteristic times when the

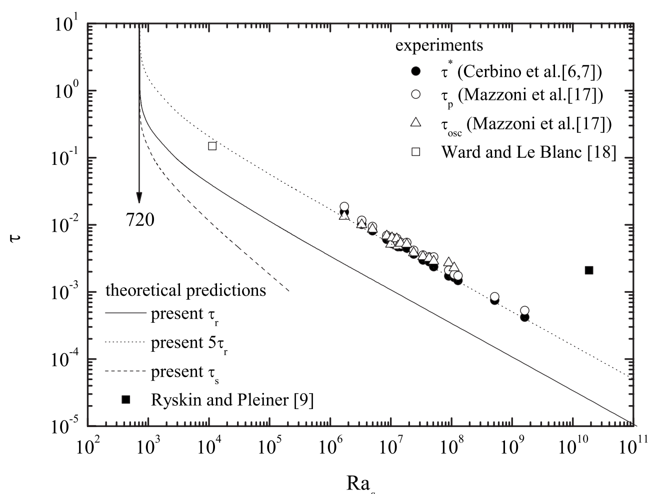

FIG. 3. Comparison of the critical times for Le→0 with available experimental data for the nanopartles suspension system of $\mathrm{Le}=1.48\times10^{-4}$, $\psi=-3.41$, and $\mathrm{Sc}=3.7\times10^4$.

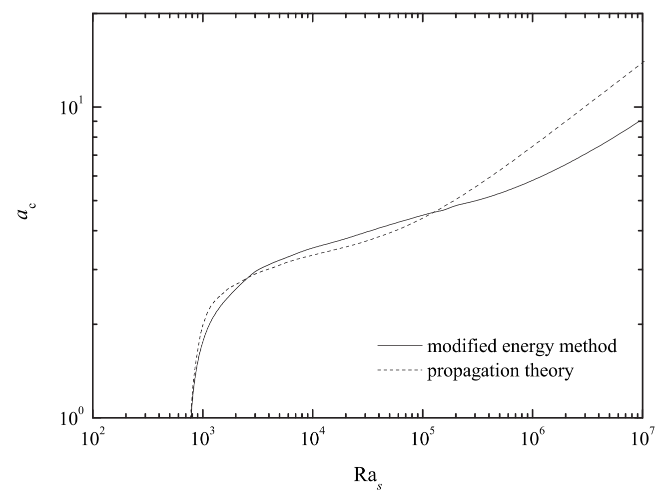

FIG. 4. Comparison of the predicted critical wave numbers for Le→0.

variance of the intensity of images starts to grow and it shows the maximum value, respectively. Based on their experimental results, they suggested the scaling relation of $\tau^* \sim \mathrm{Ra}_s^{-0.52}$, which is quite close to that of Eq. (42). They also measured the oscillation period τ_{osc} as a function of Ra_s. The experimental data of Ward and Le Blanc [18] are those for the Rayleigh-Bénard convection in the electrochemical redox system. Their stability equations and boundary conditions are identical to the present ones [19]. As shown in the figure, our τ_r value predicts the experimental trend and the $5\tau_r$ values bound the experimental τ^* values quite well. It seems that the instability, which sets in at $\tau=\tau_r$, grows until first detected experimentally at $\tau\approx5\tau_r$. For the nanoparticles suspension system of $\mathrm{Le}=7\times10^{-5}$, $\psi=-10$, and $\mathrm{Sc}=10^5$, Ryskin and Pleiner [9] analyzed the similar problem numerically using the conduction time scale. They introduced an arbitrary small initial disturbance and solved the stability equation. The first overshoot time given in Fig. 6 of Ryskin and Pleiner corresponds to $\mathrm{Ra}_s=1.86\times10^{10}$ and $\tau=2.1\times10^{-3}$. As shown in Fig. 3, their overshoot time is far from the present critical time and the experimental data. It is mentioned that the overshoot time depends on the initial magnitude of the disturbances and pattern [15].

It is well known that the conventional energy method cannot give the information on the critical wave number. For the present system this model yields the critical wave number of $a_c=0$ for the whole range of Ra_s. However, its modification yields the finite wave number at the onset of convection, as shown in Fig. 4. Even though the experimental critical wave number is not reported in the [6] experiment of Cerbino *et al.*, the finite mode rather than long-wave mode of convective motion is preferred. In this viewpoint the relaxed energy method is better than the conventional one. It is mentioned that more refined work should be pursued.

CONCLUSIONS

The critical condition to mark the onset of convective motion driven by the Soret diffusion in an initially quiescent, horizontal fluid layer heated from above has been analyzed by using the energy method and its modification. Here the stability equations are derived based on the relaxed energy method and its critical time τ_r to mark the onset of instability is obtained. It seems that for the present system manifest convection is first detected at $\tau \cong 5\tau_r$ in comparison with available experimental data and for $\tau \leqslant 5\tau_r$ velocity disturbances are too weak to be detected experimentally. The present results show that the relaxed energy method can be applied reasonably well to the analysis of onset of SDI in binary systems having a negative separation ratio and very small solute diffusivity.

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[1] M. Giglio and A. Vendramini, Phys. Rev. Lett. **38**, 26 (1977).
[2] T. Nambu, Y. Yamaguchi, T. Kushiro, and S. Sakura, Faraday Discuss. **128**, 285 (2004).
[3] A. La Porta and C. M. Surko, Phys. Rev. Lett. **80**, 3759 (1998).
[4] J. Liu and G. Ahlers, Phys. Rev. E **55**, 6950 (1997).
[5] M. I. Shliomis and M. Souhar, Europhys. Lett. **49**, 55 (2000).
[6] R. Cerbino, A. Vailati, and M. Giglio, Phys. Rev. E **66**, 055301(R) (2002).
[7] R. Cerbino, S. Mazzoni, A. Vailati, and M. Giglio, Philos. Mag. **83**, 2023 (2003).
[8] R. Cerbino, S. Mazzoni, A. Vailati, and M. Giglio, Phys. Rev. Lett. **94**, 064501 (2005).
[9] A. Ryskin and H. Pleiner, Phys. Rev. E **71**, 056303 (2005).
[10] A. Ryskin, H. W. Muller, and H. Pleiner, Phys. Rev. E **67**, 046302 (2003).
[11] V. M. Shevtsova, D. E. Melnikov, and J. C. Legros, Phys. Rev. E **73**, 047302 (2006).
[12] D. D. Joseph, Arch. Ration. Mech. Anal. **22**, 163 (1966).
[13] B. B. Straughan, *The Energy Method, Stability, and Nonlinear Convection* (Springer, Berlin, 2004).
[14] St. Hollinger, M. Lucke, and H. W. Muller, Phys. Rev. E **57**, 4250 (1998).
[15] J.-C. Chen, G. P. Neitzel, and D. F. Jankowski, Phys. Fluids **28**, 749 (1985).
[16] M. C. Kim, J. S. Hong, and C. K. Choi, AIChE J. **52**, 2677 (2006).
[17] S. Mazzoni, R. Cerbino, A. Vailati, and M. Giglio, Eur. Phys. J. E **15**, 305 (2004).
[18] W. J. Ward III and O. H. Le Blanc, Jr., Science **225**, 1471 (1984).
[19] M. C. Kim, L. H. Kim, and C. K. Choi, Int. Commun. Heat Mass Transfer **31**, 837 (2004).