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Onset of Soret convection in a nanoparticles-suspension heated from above

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Abstract−The onset of Soret-driven convection in a nanoparticles suspension heated from above is analyzed theoretically based on linear theory and relative instability concept. A new set of stability equations are derived and solved by using the dominant mode method. The dimensionless critical time τ_c to mark the onset of instability is presented here as a function of the Rayleigh number, the Lewis number and the separation ratio. Available experimental data indicate that for large Rayleigh number convective motion is detected starting from a certain time $\tau\approx 3\tau_c$. This means that the growth period of initiated instabilities is needed for convective motion to be detected experimentally. It seems evident that during $\tau_c\le\tau\le 3\tau_c$ convective motion is relatively very weak and the primary diffusive transfer is dominant.

Key words: Soret Convection, Nanoparticles Suspension, Dominant Mode Method

INTRODUCITON

Thermal convection in binary mixtures shows quite different characteristics from those in pure fluids [1-4]. If the suspension of nanoparticles is under consideration, the spatiotemporal properties of convection are much more complex than those of pure fluids or molecular solutions due to the influence of thermal diffusion, i.e., Soret-induced concentration gradients and also the extremely small particle mobility which can be reflected by the Lewis number $Le\le 10^{-4}$ [5-8]. Here $Le(=D_C/\alpha)$ is the Lewis number, D_C the diffusion coefficient, α the thermal diffusivity, respectively. The relative importance of the Soret effect with respect to weak solutal diffusion is measured by the separation ratio $\psi(=(\beta_C/\beta_T)(D_T/D_C))$, where D_T is the Soret diffusion coefficient, and $\beta_T(=-\rho^{-1}(\partial\rho/\partial T))$ and $\beta_C(=\rho^{-1}(\partial\rho/\partial C))$ are the thermal and the solutal expansion coefficient, respectively. For the case of $\psi=0$ double diffusive convection sets in when $Ra+Ra_s\ge 1708$ [9], where $Ra(=g\beta_T\Delta T d^3/\alpha\nu)$ is the thermal Rayleigh number and $Ra_s=(g\beta_C\Delta C d^3/D\nu)$ the solutal Rayleigh number, respectively.

For non-vanishing ψ, however, the external temperature gradient induces concentration variations and these create the buoyancy force. Depending on the sign of the separation ratio ψ, the solutal buoyancy force may act in the same direction as the thermal buoyancy force (for the case of positive ψ), but it counteracts this force for the case of negative ψ. For the case of negative ψ, Soret effects can induce buoyancy-driven motion even in initially uniform concentration and thermally stable configuration. For the fully developed linear temperature field the critical Rayleigh number to represent the convective instability is $Ra_c=720(Le/\psi)$ for the linear concentration field, considering the relative time scale of mass diffusion with respect to thermal diffusion [10].

In the present study the onset of Soret-driven convection in the horizontal fluid layer heated from above with the large concentration difference, that is, $Ra(Le/\psi)^{-1}>720$, will be analyzed by using the dominant mode method. And resulting predictions will also be compared with available experimental results [11].

THEORETICAL ANSLYSIS

1. Governing Equations

The problem considered here is a horizontal fluid layer confined between two rigid plates separated by the vertical distance “d”. The schematic diagram of the basic system of pure diffusion is shown in Fig. 1. The fluid layer, 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 $t\ge 0$ the fluid layer is heated suddenly from above with a constant temperature T_u, that is, Ra has a negative value. For a high $Ra(Le/\psi)^{-1}$, 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 [12]:

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

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

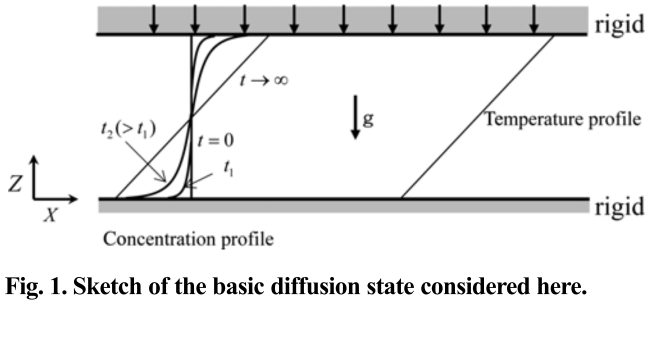

Fig. 1. Sketch of the basic diffusion state considered here.

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

$$\left(\frac{\partial}{\partial t}+\mathbf{U}\cdot\nabla\right)C=-\nabla\cdot\mathbf{j}, \tag{4}$$

$$\mathbf{j}=\mathbf{j}_D+\mathbf{j}_S=-D_C\nabla C-D_T\nabla T, \tag{5}$$

with the following initial and boundary conditions,

$$\mathbf{U}=0,\ T=T_i,\ C=C_i \qquad \text{at} \qquad t=0, \tag{6a}$$

$$\mathbf{U}=0,\ T=T_i,\ D_C\frac{\partial C}{\partial Z}+D_T\frac{\partial T}{\partial Z}=0 \qquad \text{at} \qquad Z=0, \tag{6b}$$

$$\mathbf{U}=0,\ T=T_u,\ D_C\frac{\partial C}{\partial Z}+D_T\frac{\partial T}{\partial Z}=0 \qquad \text{at} \qquad Z=d, \tag{6c}$$

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 Ra<0.

At the steady state the basic temperature and concentration profiles are linear and time-independent, and its critical condition is well summarized by Ryskin et al. [10]. However, for the case of $Ra(Le/\psi)^{-1}>>720$ convective motion can occur 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 remains unsolved. For this transient stability analysis we define a set of nondimensionalized variables τ, z, $\theta_0(=(T_i-T)/\Delta T)$ by using the scale of time d^2/D_C, length d and temperature $\Delta T(=T_i-T_u)$. Then the basic conduction state is represented in dimensionless form of

$$\frac{\partial\theta_0}{\partial\tau}=\frac{1}{Le}\frac{\partial^2\theta_0}{\partial z^2}, \tag{7}$$

with the following initial and boundary conditions,

$$\theta_0(0, z)=\theta_0(\tau, 0)=\theta_0(\tau, 1)-1=0. \tag{8}$$

The above equations can be solved by using the separation of variables:

$$\theta_0=z+2\sum_{n=1}^{\infty}\frac{(-1)^n}{n\pi}\sin(n\pi z)\exp\left(-\frac{n^2\pi^2}{Le}\tau\right). \tag{9}$$

For the case of nanoparticle suspension systems, the Lewis number is very small ($Le\approx10^{-4}$) and, therefore, the basic concentration field can be approximated by

$$\theta_0=z \qquad \text{for} \qquad \tau\geq Le. \tag{10}$$

Based on the above temperature distribution, the dimensionless concentration field is given [12]:

$$\frac{\partial c_0}{\partial\tau}=\frac{\partial^2 c_0}{\partial z^2} \qquad \text{for the region of } \tau\geq Le, \tag{11}$$

under the following initial and boundary conditions,

$$c_0(0,z)=0,\ \frac{\partial c_0}{\partial z}(\tau,0)=1,\ \frac{\partial c_0}{\partial z}(\tau,1)=1, \tag{12}$$

where $c_0=D_C(C-C_0)/(j_S d)$ and $j_S=-D_S\Delta T/d$. For the case of negative ψ the Soret flux j_S has a positive value. The boundary conditions have been obtained by the impermeable conditions for concentration at both boundaries, that is, j=0 at Z=0 and d. The above equations can be solved by using the Laplace transform:

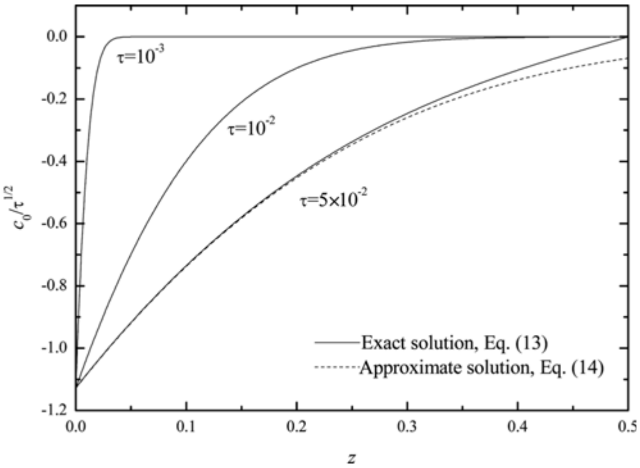

Fig. 2. Comparison of Eq. (9) with (8). For $\tau\leq0.05$ the difference between two equations becomes negligible.

$$c_0(\tau,\zeta)=\sqrt{4\tau}\sum_{n=0}^{\infty}\left\{-\operatorname{ierfc}\left(\frac{n}{\sqrt{\tau}}+\frac{\zeta}{2}\right)-\operatorname{ierfc}\left(\frac{n+1}{\sqrt{\tau}}-\frac{\zeta}{2}\right)\right.$$
$$\left.+\operatorname{ierfc}\left(\frac{n+1/2}{\sqrt{\tau}}-\frac{\zeta}{2}\right)+\operatorname{ierfc}\left(\frac{n+1/2}{\sqrt{\tau}}+\frac{\zeta}{2}\right)\right\}, \tag{13}$$

where $\zeta=z/\sqrt{\tau}$. For the deep-pool system of small τ ($Le\leq\tau\leq0.01$) the basic concentration field is approximated by

$$c_0=\sqrt{4\tau}\begin{cases}-\operatorname{ierfc}\left(\dfrac{z}{2\sqrt{\tau}}\right) & z\leq0.5,\\ \operatorname{ierfc}\left(\dfrac{1-z}{2\sqrt{\tau}}\right) & z\geq0.5.\end{cases} \tag{14}$$

Here $-c_0(\tau, 0)=c_0(\tau, 1)=\sqrt{4\tau/\pi}$. The agreement with the exact solution is very good, as shown in Fig. 2.

2. Stability Equations

Under linear stability theory the nondimensionalized conservation equations incorporating the Soret effect are constituted as follows:

$$\left(\frac{1}{Sc}\frac{\partial}{\partial\tau}-\nabla^2\right)\nabla^2 w=\nabla_1^2 c_1+\psi\nabla_1^2\theta_1, \tag{15}$$

$$\frac{\partial c_1}{\partial\tau}+Rs\frac{\partial c_0}{\partial z}w=\nabla^2(c_1-\theta_1), \tag{16}$$

$$Le\left(\frac{\partial\theta_1}{\partial\tau}+Raw\right)=\nabla^2\theta_1, \tag{17}$$

under the following boundary conditions,

$$w_1=\frac{\partial w_1}{\partial z}=\frac{\partial c_1}{\partial z}=\theta_1=0 \qquad \text{at } z=0 \qquad \text{and } z=1, \tag{18}$$

where w_1, c_1 and θ_1 represent dimensionless quantities of vertical velocity, concentration and temperature disturbances, respectively. Here $\nabla^2=(\partial^2/\partial x^2)+(\partial^2/\partial y^2)+(\partial^2/\partial z^2)$, $\nabla_1^2=(\partial^2/\partial x^2)+(\partial^2/\partial y^2)$, $Rs(=Ra(\psi/Le))=(g\beta_C j_S d^4/D_C^2\nu)$ and the disturbance velocity, concentration and temperature are nondimensionalized by D_C/d, $\alpha\nu/(g\beta_T d^3)$ and $D_C\nu/(g\beta_C d^3)$, respectively.

For the limiting case of very small Lewis numbers the temperature disturbance governed by Eqs. (17) and (18) has the solution of $\theta_1=0$. Therefore, the disturbance equations reduce to

$$\left\{\frac{1}{Sc}\frac{\partial}{\partial\tau}-\nabla^2\right\}\nabla^2 w_1=\nabla_1^2 c_1, \tag{19}$$

$$\frac{\partial c_1}{\partial\tau}+Rsw_1\frac{\partial c_0}{\partial z}=\nabla^2 c_1, \tag{20}$$

with the following boundary conditions,

$$w_1=\frac{\partial w_1}{\partial z}=\frac{\partial c_1}{\partial z}=0 \qquad \text{at } z=0 \text{ and } z=1. \tag{21}$$

3. Dominant Mode Method

For the limiting case of $\tau\rightarrow 0$, the stretched vertical coordinate $\zeta(=z/\sqrt{\tau})$ is more suitable to describe the systems havig boundary layer characteristics. Therfore, it may be natural that $c_1(\tau, z)=c_1^*(\tau, \zeta)$ and $w_1(\tau, z)=w_1^*(\tau, \zeta)$. Now, Eqs. (19)-(21) are transformed by using the similarity variable of the base state. Then, for the limiting case of $Sc\rightarrow\infty$ the perturbation equations can be expressed as

$$\left(\frac{1}{\tau}\frac{\partial^2}{\partial\zeta^2}-a^2\right)^2 w_1^*=a^2 c_1^*, \tag{22}$$

$$\frac{\partial c_1^*}{\partial\tau}-\frac{1}{\tau}\left(\frac{\partial^2}{\partial\zeta^2}+\frac{1}{2}\zeta\frac{\partial}{\partial\zeta}-a^2\tau\right)c_1^*=-Rsw_1^*\frac{\partial c_0}{\partial\zeta}, \tag{23}$$

where $(\partial c_0/\partial\zeta)=\sqrt{\tau}\,\mathrm{erfc}(\zeta/2)$. The corresponding boundary conditions are

$$w_1^*=\frac{\partial w_1^*}{\partial\zeta}=\frac{\partial c_1^*}{\partial\zeta}=0 \qquad \text{at } \zeta=0 \text{ and } \zeta\rightarrow\infty. \tag{24}$$

For the case of $\tau\rightarrow 0$, Eq. (23) may be further approximated by

$$\tau\frac{\partial c_1^*}{\partial\tau}=\left(\frac{\partial^2}{\partial\zeta^2}+\frac{\zeta}{2}\frac{\partial}{\partial\zeta}\right)c_1^*, \tag{25}$$

And, therefore, the initial evolution of perturbation is independent of the wave number. Because of the linearity and the homogeneity of Eq. (25), the concentration perturbation can be expanded:

$$c_1^*(\tau,\zeta)=\sum_{n=0}^{\infty}A_n(\tau)\phi_n(\zeta), \tag{26}$$

under the separation of variables concept. By separating the variables, there follows

$$\frac{\tau}{A_n}\frac{dA_n}{d\tau}=\frac{1}{\phi(\zeta)}L\phi_n(\zeta)=\lambda_n, \tag{27}$$

where $L=((d^2/d\zeta^2)+(\zeta/2)(d/d\zeta))$ and λ_n's are the separation variables. This relation leads to the Sturm-Liouville problem

$$L\phi_n=\lambda_n\phi_n(\zeta), \qquad \frac{d\phi_n}{d\zeta}=0 \qquad \text{at } \zeta=0 \text{ and } \zeta\rightarrow\infty. \tag{28}$$

From the Sturm-Liouville theory, the weighting function is $\exp(\zeta^2/4)$, the eigenfunctions ϕ_n of L are $H_n(\zeta/2)\exp(-\zeta^2/4)$, here H_n's are the Hermite polynomials, and the eigenvalues are $\lambda_n=-(n+1)/2$ for n=0, 2, 4, …. From Eqs. (26)-(28), the amplitude coefficients of concentration perturbation represented by Eq. (25) can be expressed as

$$\frac{dA_n}{d\tau}=-\frac{(n+1)}{2\tau}A_n \qquad \text{for } \tau\rightarrow 0 \text{ and } n=0, 2, 4, \ldots. \tag{29}$$

This relation means that the zeroth mode decays with time while the rest of the spectrum decays more rapidly, i.e., the zeroth mode is the dominant one.

By substituting the solution obtained above into Eq. (23), and using the orthogonality between ϕ_n's, we obtain the amplitude variation of the dominant mode as

$$\tau\frac{dA_0}{d\tau}=\sigma\tau A_0=-\left(\frac{1}{2}+a^2\tau\right)A_0-Rs\,\tau^2 A_0\langle w^*\frac{dc_0^*}{d\zeta}\rangle, \tag{30a}$$

where

$$\langle w^*\frac{dc_0^*}{d\zeta}\rangle=\frac{\int_0^\infty w^*\frac{dc_0^*}{d\zeta}\,d\zeta}{\int_0^\infty \exp(-\zeta^2/4)d\zeta}=-\frac{\int_0^\infty w^*\mathrm{erfc}(\zeta/2)d\zeta}{\sqrt{\pi}}. \tag{30b}$$

Here $w^*=w_1^*/(\tau A_0)$, $c_0^*(\zeta)=c_0(\tau, \zeta)/\sqrt{\tau}$ and σ is the growth rate in the (τ, ζ)-domain, i.e., $\sigma=1/A_0(dA_0/d\tau)$. With $c_1^*=A_0(\tau)\exp(-\zeta^2/4)$, for the limiting case of $Sc\rightarrow\infty$, w^* can be obtained analytically from Eq. (22) as

$$\left(\frac{\partial^2}{\partial\zeta^2}-a^2\tau\right)^2 w_1^*=a^2\tau\exp(-\zeta^2/4), \tag{31}$$

under the boundary conditions of Eq. (24). Therefore, the driving force for the instability, the integral $\langle w^* dc_0^*/d\zeta\rangle$, is also a function of time and the wave number.

Now, consider the stability criterion. Under the relative stability concept the critical time τ_r is determined according to the following criterion [13]:

$$\frac{1}{E_0}\frac{dE_0}{d\tau}=\frac{1}{E_1}\frac{dE_1}{d\tau} \qquad \text{at} \qquad \tau=\tau_r. \tag{32}$$

Here, for the case of $Sc\rightarrow\infty$ the dimensionless energy identities are defined as

$$E_0(\tau)=\frac{1}{2}\langle c_0^2\rangle,\ \ E_1(\tau)=\frac{1}{2}\langle c_1^2\rangle, \tag{33}$$

where $\langle\cdot\rangle=\sqrt{\left(\int_V (\cdot)^2 dV\right)/V}$ and V denotes the dimensionless volume considered, e.g., one pair of vortices. Based on Eq. (9) and the dominant mode solution $c_1=A_0(\tau)\exp(-\zeta^2/4)$, the relations of $(1/E_0)(dE_0/d\tau)=3/(4\tau)$ and $(1/E_1)(dE_1/d\tau)=(1/A_0)(dA_0/d\tau)+1/(4\tau)$ can be derived, and therefore, the relative instability time τ_r is determined as

$$\sigma+\frac{1}{4\tau}=\frac{3}{4\tau} \qquad \text{at } \tau=\tau_r. \tag{34}$$

Based on Eqs. (30) and (35), the following relation for the relative instability time τ_r is obtained:

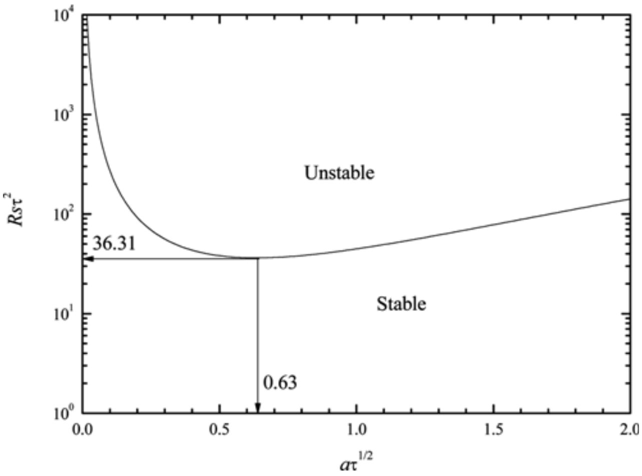

Fig. 3. Neutral stability curves for the nanoparticles-suspension systems.

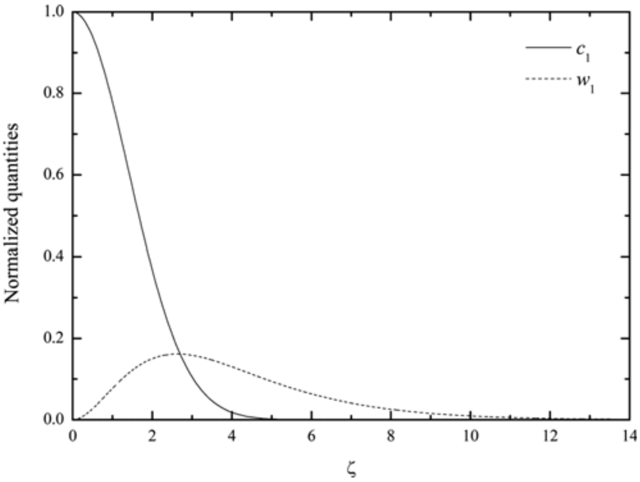

Fig. 4. Normalized amplitude functions at the critical conditions.

$$\mathrm{Rs}\,\tau_r^2 = \frac{\sqrt{\pi}(1+a^2\tau_r)}{\int_0^\infty w^* \mathrm{erfc}(\zeta/2)d\zeta}. \tag{35}$$

For small τ the present neutral stability curve is given in Fig. 3. The critical conditions can be determined analytically from Eq. (35) when

$$\frac{\partial \tau_r}{\partial a} = 0, \tag{36}$$

for a given Rs. The critical wave number a_c yields the minimum τ-value, *i.e.* τ_c. At this critical condition, the disturbance quantities are summarized in Fig. 4.

RESULTS AND DISCUSSIONS

It is well-known that for large τ, the long wave instability of a=0 is the preferred mode and the critical condition is Rs=720 [10]. For small τ the present neutral stability curve given in Fig. 3 yields the critical condtions:

$$\mathrm{Rs}\,\tau_c^2 = 36.31,\ a_c\sqrt{\tau_c} = 0.63 \quad \text{for } \tau \to 0. \tag{37}$$

Eq. (37) can be rewritten as a function of Ra_s:

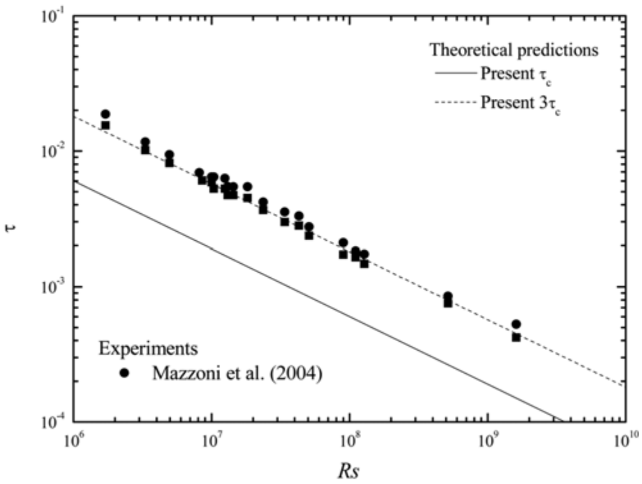

Fig. 5. Comparison of critical times with available experimental data.

$$\tau_c = 12.88 Ra_s^{-2/3} \text{ as } \tau \to 0, \tag{38}$$

since $\mathrm{Rs}=Ra_s\sqrt{\pi/4\tau}$. This equation is less convenient in predicting τ_c; however, Cerbino et al. [8] suggested a similar relation of $\delta^{-1} \sim (\mathrm{Rs}\delta)^{0.35}$ where $\delta = \sqrt{\pi\tau^*}$. Their relation can be reformulated as $\tau^* \sim Ra_s^{-0.70}$, where τ^* is the latency time illustrated later. Their exponent -0.70 is quite similar to the present one $-2/3$.

By using the shadowgraph method Cerbino et al. [6,7] and Mazzoni et al. [11] visualized the Soret-driven convective motion in a colloidal suspension of 22 nm diameter silica particles (LUDOX®) dispersed in water. For this system Le=1.48×10^{-4}, $\Psi=-3.41$ and Sc=3.7×10^4. They obtained the latency time τ^* and the peak time τ_p as the characteristic times when the variance of the intensity of images starts to grow and it shows the maximum value, respectively. They also measured the oscillation period τ_{osc} as a function of Rs. These experimental data are compared in Fig. 5. As shown in this figure, our $3\tau_c$-values bound the experimental τ^*-values quite well. As expected, the peak time τ_p is a little larger than the latency time τ^*. The present results imply that the predicted critical time τ_c is smaller than the detection time τ^*, i.e. the fastest growing mode of instabilities, which sets in at $\tau=\tau_c$, will grow with time until manifest motion is first detected experimentally. To find the growing mechanism, a more refined analysis is now in progress by assuming the dominant mode solution as a proper initial conditions.

CONCLUSIONS

The critical condition to mark the onset of convective motion driven by Soret diffusion in an initially quiescent, horizontal nanoparticle-suspension layer heated from above has been analyzed theoretically. It appears that manifest convection is detected at $3\tau_c$ in comparison with available experimental data. For small time the critical wavelength becomes smaller with increasing Rs, while for large time a long wave mode ($a_c \to 0$) is the most unstable mode.

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NOMENCLATURE

a : dimensionless wavenumber
C : concentration
c_0 : dimensionless base concentration, $D_C(C-C_0)/(j_S d)$
d : distance between the plates
D_C : diffusion coefficient
D_T : Soret diffusion coefficient
g : gravitational acceleration
H_n : n-th Hermite polynomial
j_S : Soret diffusion flux, $-D_S\Delta T/d$
Le : Lewis number, D_C/α
P : pressure
Ra : Rayleigh number, $g\beta_T\Delta Td^3/\alpha\nu$
Ra_s : solutal Rayleigh number, $g\beta_C\Delta Cd^3/D\nu$
Rs : Rayleigh number based on Soret flux, $Ra(Le/\psi)^{-1}$
Sc : Schmidt number, ν/D_C
T : temperature
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 : thermal diffusivity
β_C : solutal expansion coefficient, $(1/\rho)(\partial\rho/\partial C)$
β_T : thermal expansion coefficient, $(-1/\rho)(\partial\rho/\partial T)$
Δ_C : penetration depth
δ_C : dimensionless penetration depth, Δ_C/d
θ_0 : dimensionless base temperature, $(T_i-T)/\Delta T$
ζ : similarity variable, $z/\sqrt{\tau}$
τ : dimensionless time, D_Ct/d^2
ψ : separation ratio, $(\beta_C/\beta_T)(D_T/D_C)$

Subscripts

c : critical conditions
i : initial conditions
u : upper plate conditions
0 : basic quantities
1 : perturbed quantities

Superscript

* : transformed quantities

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