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Instability analysis of Marangoni convection for absorption process accompanied by heat transfer

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Abstract

The objective of this paper is to analyze the onset of Marangoni convection for absorption process accompanied by heat transfer using propagation theory. Ammonia–water is used as a representative solution pair for the absorption process and 2-ethyl-1-hexonal is considered an additive. The re-modified solutal Marangoni number *MBs* is linearly related to the modified solutal Biot number Bs^*. Rearranging this relation, a useful correlation of the solutal Marangoni number *Ms* with respect to the solutal Biot number *Bs* is obtained in a linear form. It is found that there is a critical *Bs* causing the most unstable state of fluid layer. The critical time τ_c to mark the onset of Marangoni convection can be predicted using the correlation developed in the present study. The results from the present study will provide a guideline to enhance heat and mass transfer performance during the absorption process.

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Keywords: Marangoni convection; Absorption; Heat transfer; Propagation theory; Additive

1. Introduction

When an initially quiescent fluid experiences absorption or desorption of a solute through a free surface, the surface tension of the fluid varies with the concentration and temperature of the fluid at the surface. This variation of the surface tension can cause Marangoni convection. After Pearson's [1] theoretical analysis of the thermal Marangoni convection, many studies on Marangoni instability have been performed theoretically or experimentally in the heat transfer systems with different boundary conditions [2–4]. Under the linear stability theory, Sternling and Scriven [5] analyzed solutal Marangoni instabilities of two immiscible fluid layers with mass transfer through the interface. Their onset criteria of Marangoni convection for linear basic concentration profiles have been applied to other systems [6,7]. Using propagation theory for evaporation system, Kang et al. [8] analyzed Brian's [7] system and showed that theoretical results obtained by their numerical simulation agree well with experimental ones obtained by Brian et al. [9].

When certain surfactants are added to the liquid in small quantities, the heat and mass transfer rate can be significantly enhanced beyond the diffusion and conduction controlled rates of the stable system. Many studies on these phenomena have been performed in LiBr/H_2O absorption process [10–12]. Kang et al. [13] conducted experiments on the visualization of Marangoni convection in NH_3/H_2O absorption system with several additives and reported the meaningful results of the effect of additives on the initiation of solutal Marangoni convection.

The objective of this study is to clarify the onset criteria of Marangoni convection for absorption process accompanied by heat transfer based on the propagation theory. The results from the present study will provide a guideline to enhance heat and mass transfer performance during the absorption process.

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Nomenclature

a	dimensionless wave number
A, B	coefficients in Eq. (48)
Bs	solutal Biot number
Bt	thermal Biot number
C	concentration
C_{eq}	equilibrium concentration
d	liquid depth
D	differential operator
D_l	solutal diffusivity
h	transfer coefficient in gas phase
i	imaginary number
Le	Lewis number
MBs	re-modified solutal Marangoni number
MBt	re-modified thermal Marangoni number
Ms	solutal Marangoni number
Mt	thermal Marangoni number
P	pressure
S	surface tension
Sc	Schmidt number
t	time
T	temperature
$\vec{U}$	velocity vector
$\vec{u}$	dimensionless velocity vector
W	vertical velocity
w	dimensionless vertical velocity
x, y, z	coordinates

Greek symbols

α	thermal diffusivity
β	basic concentration gradient
Δ_ϕ	solutal penetration depth
ϕ	dimensionless concentration
γ	surface tension gradient
μ	viscosity
ν	kinematic viscosity
θ	dimensionless temperature
ρ	density
τ	dimensionless time
ζ	similarity variable

Subscripts

0	basic state
1	perturbed state
c	critical condition
i	initial state
s	solutal quantity
t	thermal quantity

Superscript

*	amplitude function

2. Marangoni convection

In general, the solute in solution lowers the surface tension at the liquid–vapor interface. However in solution involving certain additives, as the concentration of the solute increases, the surface tension of the liquid also increases. When this happens, Marangoni convection driven by surface tension variation occurs resulting in significant enhancement of the heat and mass transfer rate. Using the linear stability theory, Sternling and Scriven [5] and Brian [6] reported that both the sign of the surface tension gradient with respect to the solute concentration and the direction of the mass transfer mainly affect the onset of Marangoni convection.

In this present study, the absorbate transfers from the gas phase to the liquid phase as shown in Fig. 1. Applying the results of the above works to the absorption system considered here, the basic condition of the onset of Marangoni convection is expressed as follow:

$$\beta\gamma_s < 0, \tag{1}$$

where $\beta(= \partial C/\partial Z)$ and $\gamma_s(= \partial S/\partial C)$ denote the basic concentration gradient and the surface tension gradient with respect to the solute concentration.

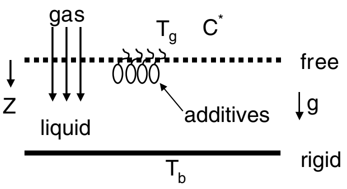

Fig. 1. Schematic diagram of system considered here.

3. Formulation of problem

A horizontal liquid layer considered here is bounded an upper free surface at $Z = 0$ and a lower rigid plane at $Z = d$, as shown in Fig. 1. At time $t = 0$ the absorbate at different temperature from liquid layer, i.e., ammonia in the gas phase begins to be absorbed into the absorbant with additives, i.e., water involving 2-ethyl-1-hexanol. The bulk liquid is incompressible and Boussinesque solution. Both the Soret and Rayleigh effects caused by absorption process are neglected. No absorption heat is generated at the surface.

Under the above approximations, the governing equations to represent Marangoni convection are given by

$$\nabla \cdot \vec{U} = 0, \tag{2}$$

$$\rho\left(\frac{\partial}{\partial t} + \vec{U} \cdot \nabla\right)\vec{U} = -\nabla P + \mu \nabla^2 \vec{U}, \tag{3}$$

$$\left(\frac{\partial}{\partial t} + \vec{U} \cdot \nabla\right)C = D_1 \nabla^2 C, \tag{4}$$

$$\left(\frac{\partial}{\partial t} + \vec{U} \cdot \nabla\right)T = \alpha \nabla^2 T, \tag{5}$$

$$S = S_i + \gamma_s(C - C_i) + \gamma_t(T - T_i), \tag{6}$$

where $\vec{U}$, ρ, P, μ, C, D_1, T, α, S and $\gamma_t(= \partial S/\partial T)$ denote the velocity vector, the liquid density, the pressure, the liquid viscosity, the solute concentration, the solutal diffusivity in the liquid, the temperature, the thermal diffusivity of the liquid, the surface tension and the surface tension gradient with respect to the temperature, respectively. The subscript i, s and t represent the quantity of the initial state, the solutal quantity and the thermal one.

In the present system, the basic profiles of concentration and temperature of the solution can be represented as the following dimensionless equation

$$\frac{\partial \phi_0}{\partial \tau} = \frac{\partial^2 \phi_0}{\partial z^2}, \quad \frac{\partial \theta_0}{\partial \tau} = Le \frac{\partial^2 \theta_0}{\partial z^2}, \tag{7-1,2}$$

with the following proper initial and boundary conditions,

$$\phi_0 = 0, \quad \theta_0 = 0 \qquad \text{at } \tau = 0, \tag{8}$$

$$-\frac{\partial \phi_0}{\partial z} = Bs(1 - \phi_0), \quad -\frac{\partial \theta_0}{\partial z} = Bt(1 - \theta_0) \quad \text{at } z = 0, \tag{9}$$

$$\frac{\partial \phi_0}{\partial z} = 0, \quad \theta_0 = 0 \qquad \text{at } z = 1, \tag{10}$$

where z, τ, ϕ_0 and ϕ_0 denote the dimensionless vertical distance, the dimensionless time, the dimensionless basic concentration of the solute, and the dimensionless basic temperature, respectively. The subscript 0 denotes the basic quantity. Length is non-dimensionalized by the liquid depth d, the time by d^2/D_1, and the basic concentration by C_{eq} and the basic temperature by $\Delta T(= T_{\mathrm{g}} - T_{\mathrm{b}})$. In the case of no-absorption heat generation at the surface, the boundary conditions of temperature and concentration are given as Eq. (9), which is a general expression at the surface. It can be applied for both cases of equilibrium and non-equilibrium states. The dimensionless parameters Le, Bs and Bt are defined as

$$\text{Lewis number} \quad Le = \frac{\alpha}{D_1}, \tag{11}$$

$$\text{solutal Biot number} \quad Bs = \frac{h_s d}{D_1}, \tag{12}$$

$$\text{thermal Biot number} \quad Bt = \frac{h_t d}{\alpha}, \tag{13}$$

where h_s and h_t denote the mass-transfer coefficient and the heat-transfer one, respectively.

Under the semi-infinite approximation, the similarity solutions for the above equations are well known [14],

$$\begin{aligned}\phi_0 &= \operatorname{erfc}\left(\frac{z}{2\sqrt{\tau}}\right) - \exp\left(Bs \times z + \tau Bs^2\right) \\ &\quad \times \operatorname{erfc}\left(\frac{z}{2\sqrt{\tau}} + Bs\tau^{1/2}\right) \\ &= \operatorname{erfc}\left(\frac{\zeta}{2}\right) - \exp\left(Bs^*\zeta + Bs^{*^2}\right)\operatorname{erfc}\left(\frac{\zeta}{2} + Bs^*\right) \\ &\quad \text{for } \tau < 0.01,\end{aligned} \tag{14}$$

$$\begin{aligned}\theta_0 &= \operatorname{erfc}\left(\frac{z}{2\sqrt{Le\tau}}\right) - \exp\left(Bt \times z + Le\tau Bt^2\right) \\ &\quad \times \operatorname{erfc}\left(\frac{z}{2\sqrt{Le\tau}} + Bt\sqrt{Le\tau}\right) \\ &= \operatorname{erfc}\left(\frac{\zeta}{2\sqrt{Le}}\right) - \exp\left(Bt^*\zeta + LeBt^{*^2}\right) \\ &\quad \times \operatorname{erfc}\left(\frac{\zeta}{2\sqrt{Le}} + \sqrt{Le}Bt^*\right) \quad \text{for } \tau < 0.01,\end{aligned} \tag{15}$$

where similarity variable $\zeta = z/\tau^{1/2}$, modified solutal Biot number $Bs^* = \tau^{1/2}Bs$ and modified thermal Biot number $Bt^* = \tau^{1/2}Bt$. Here Bs^* and Bt^* are assumed to be constant. The above similarity solutions are almost the same with the exact solutions obtained without any approximation for $\tau < 0.01$. By this reason they have been used to study the convective instability analysis of the horizontal fluid for deep-pool systems. Over a certain absorbing rate exceeding the critical value, Marangoni convection will set in. The important dimensionless parameters are identified as

$$\text{solutal Marangoni number} \quad Ms = \frac{\gamma_s d}{\mu D_1} C_{\mathrm{eq}}, \tag{16}$$

$$\text{thermal Marangoni number} \quad Mt = \frac{\gamma_t d}{\mu \alpha} \Delta T, \tag{17}$$

$$\text{Schmidt number} \quad Sc = \frac{\nu}{D_1}. \tag{18}$$

The Marangoni numbers Ms and Mt are a measure of the ratio of the driving (the imposed concentration difference C_{eq}; the temperature one ΔT) and the damping (the viscosity and the solute diffusion; the viscosity and the conduction) in the system and are the most important parameters in Marangoni convection.

4. Linear stability theory

Under the normal mode approximation, the dimensionless disturbance quantities $\vec{u}_1$, ϕ_1 and θ_1 can be expressed as follow:

$$(\vec{u}_1, \phi_1, \theta_1) = [\vec{u}_1^*(\tau, z), \phi_1^*(\tau, z), \theta_1^*(\tau, z)] \exp[\mathrm{i}(a_x x + a_y y)], \tag{19}$$

where $\vec{u}_1$, ϕ_1 and θ_1 are the amplitude functions, and i, a_x and a_y denote the imaginary number, the wave number in the x-direction, and the wave number in the y-direction. x and y denote the dimensionless horizontal coordinates. The perturbed velocity has the scale of D_1/d. The perturbed concentration has been non-dimensionalized by C_{eq} and the perturbed temperature by ΔT with the same manner of the basic ones. Under the linear stability theory, the dimensionless amplitude equations for the vertical velocity amplitude w_1^*, concentration ϕ_1^* and temperature θ_1^* are obtained respectively as follow

$$\left[\frac{1}{Sc}\frac{\partial}{\partial \tau} - \left(\frac{\partial^2}{\partial z^2} - a^2\right)\right]\left(\frac{\partial^2}{\partial z^2} - a^2\right) w_1^* = 0, \tag{20}$$

$$\frac{\partial \phi_1^*}{\partial \tau} + w_1^* \frac{\partial \phi_0}{\partial z} = \left(\frac{\partial^2}{\partial z^2} - a^2\right)\phi_1^*, \tag{21}$$

$$\frac{\partial \theta_1^*}{\partial \tau} + w_1^* \frac{\partial \theta_0}{\partial z} = Le\left(\frac{\partial^2}{\partial z^2} - a^2\right)\theta_1^*, \tag{22}$$

where the dimensionless wave number a denotes $\left(a_x^2 + a_y^2\right)^{1/2}$. Neglecting the solute accumulation, the surface diffusion and the absorption heat in the absorbed layer, the proper boundary conditions are given by

$$w_1^* = 0 \quad \text{at } z = 0, \tag{23}$$

$$a^2(Ms\phi_1^* + LeMt\theta_1^*) = -\frac{\partial^2 w_1^*}{\partial z^2} \quad \text{at } z = 0, \tag{24}$$

$$\frac{\partial \phi_1^*}{\partial z} = Bs\phi_1^* \quad \text{at } z = 0, \tag{25}$$

$$\frac{\partial \theta_1^*}{\partial z} = Bt\theta_1^* \quad \text{at } z = 0, \tag{26}$$

$$w_1^* = \frac{\partial w_1^*}{\partial z} = \frac{\partial \phi_1^*}{\partial z} = \theta_1^* = 0 \quad \text{at } (z = 1). \tag{27}$$

Eq. (24) means that the tangential shear stress is balanced by surface tension driven by the solute concentration change and the temperature one at the upper free surface.

5. Scaling analysis

Foster [15] showed that for deep-pool system the onset time to mark convective motion is related to the penetration depth other than liquid depth. In the small time region, applying the scaling analysis to Eqs. (21), (22) and (24) the following relations are possible

$$w_1^* \frac{\Delta\phi_0}{\Delta_\phi} \sim \frac{\phi_1^*}{\Delta_\phi^2}, \tag{28}$$

$$w_1^* \frac{\Delta\theta_0}{\Delta_\phi} \sim \frac{\theta_1^*}{\Delta_\phi^2}, \tag{29}$$

$$(Ms\phi_1^* + LeMt\theta_1^*) \sim -\frac{w_1^*}{\Delta_\phi^2}, \tag{30}$$

where Δ_ϕ represents the dimensionless concentration penetration depth. The basic concentration and temperature gradients are obtained from Eqs. (14) and (15) as follow

$$\frac{\Delta\phi_0}{\Delta_\phi} \sim Bs \exp(Bs^* + Bs^{*^2})\mathrm{erfc}(0.5 + Bs^*), \tag{31}$$

$$\frac{\Delta\theta_0}{\Delta_\phi} \sim Bt \exp(Bt^* + LeBt^{*^2})\mathrm{erfc}(0.5/\sqrt{Le} + \sqrt{Le}Bt^*). \tag{32}$$

Substituting Eqs. (31) and (32) into Eqs. (28) and (29) and rearranging Eq. (30) the following relation between non-dimensional parameters yield

$$\begin{aligned} MBs + MBt/Le = {} & Ms^* Bs^* \exp(Bs^* + Bs^{*^2})\mathrm{erfc}(0.5 + Bs^*) \\ & + Mt^* Bt^* \exp(Bt^* + LeBt^{*^2}) \\ & \times \mathrm{erfc}(0.5/\sqrt{Le} + \sqrt{Le}Bt^*), \end{aligned} \tag{33}$$

where the modified solutal Marangoni number $Ms^* = \sqrt{\tau}Ms$ and the modified thermal Marangoni number $Mt^* = \sqrt{\tau}Mt$. The re-modified solutal Marangoni number MBs and the re-modified thermal one MBt represent the first term and the second term times Lewis number Le in the right-hand side of the above equation respectively. These re-modified parameters obtained by the scaling analysis can be identified as

re-modified solutal Marangoni number

$$MBs = \frac{\gamma_s d^2}{\mu D_1}\left(\frac{\partial C_0}{\partial Z}\right)\bigg|_{Z=\Delta_C}, \tag{34}$$

re-modified thermal Marangoni number

$$MBt = \frac{\gamma_t d^2}{\mu \alpha}\left(\frac{\partial T_0}{\partial Z}\right)\bigg|_{Z=\Delta_C}. \tag{35}$$

Considering Eqs. (16), (17), (34) and (35), Marangoni number characterizing the absorption system can be defined with overall concentration difference or the basic concentration gradient for absorption process.

6. Propagation theory

For small time region, using similarity variable ζ, the coordinate of system can be transformed from (τ, z)-frame to (τ, ζ)-frame. In this sense the dimensionless velocity in (τ, ζ)-frame w^* can be expressed as follow

$$w^*(\zeta) = \frac{W_1^*}{W_{\Delta_C}} = \sqrt{\tau} w_1^*(\tau, z), \tag{36}$$

where W_1^* is the disturbance velocity and $W_{\Delta_C}(=\Delta_C/t)$ is the growth velocity of solutal penetration depth in which Δ_C and t denote the solutal penetration depth and time. With this procedure, the following relations are possible:

$$[w_1^*, \phi_1^*, \theta_1^*] = [w^*(\zeta)/\sqrt{\tau}, \phi^*(\zeta), \theta^*(\zeta)]. \tag{37}$$

Inserting the above forms into Eqs. (20)–(27) yield

$$\left[\left(D^2 - a^{*2}\right)^2 + \frac{1}{2Sc}\left(\zeta D^3 - a^{*2}\zeta D + 2a^{*2}\right)\right] w^* = 0, \tag{38}$$

$$\left(D^2 + \frac{1}{2}\zeta D - a^{*2}\right)\phi^* - w^* D\phi_0 = 0, \tag{39}$$

$$Le\left(D^2 + \frac{1}{2Le}\zeta D - a^{*2}\right)\theta^* - w^* D\theta_0 = 0 \tag{40}$$

with the boundary conditions

$$w^* = 0 \quad \text{at } \zeta = 0, \tag{41}$$

$$D^2 w^* = -a^*(Ms^*\phi^* + LeMt^*\theta^*) \quad \text{at } \zeta = 0, \tag{42}$$

$$D\phi^* = Bs^*\phi^* \quad \text{at } \zeta = 0, \tag{43}$$

$$D\theta^* = Bt^*\theta^* \quad \text{at } \zeta = 0, \tag{44}$$

$$w^* = Dw^* = D\phi^* = \theta^* = 0 \quad \text{for } \zeta \to \infty, \tag{45}$$

where $D(\cdot) = \mathrm{d}(\cdot)/\mathrm{d}\zeta$, $a^* = a\tau^{1/2}$. All these dimensionless groups having the superscript $*$ are assumed to be eigenvalues through τ. Then, Eqs. (38)–(45) are functions of the similarity variable ζ only. For a given Sc, Le, Bs^*, Bs^*, Mt^* and a^* the minimum value of Ms^* is found numerically.

7. Results and discussion

With the propagation theory the marginal stability curves for various Sc are obtained as shown in Fig. 2. It is found that the minimum values of Ms^* slightly decrease with increasing Sc for other fixed parameters and the curves become almost the same for $Sc > 500$. This means that the increase of Sc makes the fluid layer be more unstable and the effect of Sc on the criteria of solutal Marangoni convection instability is not significant for a larger value than 500.

For an absorption system without heat transfer, the re-modified solutal Marangoni number MBs is linearly related to the modified solutal Biot number Bs^* as shown in Fig. 3. The following linear correlation is found within the bound of 1% error:

$$MBs = 5.36 + 7.43Bs^*. \tag{46}$$

The above equation for non-linear basic profile system is similar to Nield's result (1964) for linear basic profile system, which is $Mt = 32.073Bt$ for a large Bt.

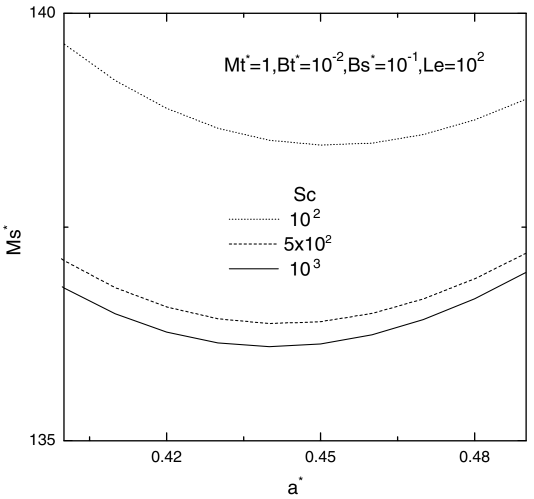

Fig. 2. Marginal stability curves for various Sc.

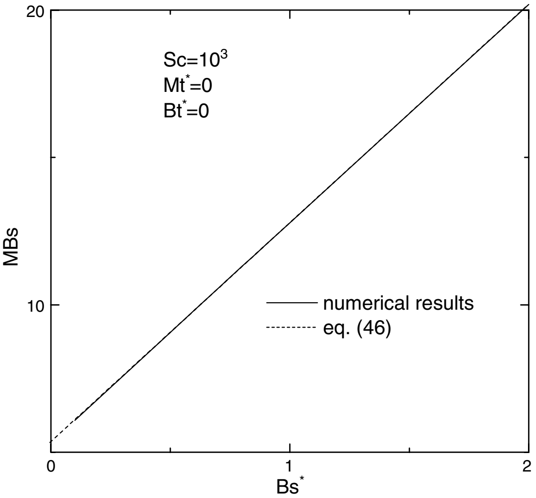

Fig. 3. Re-modified solutal Marangoni number MBs versus modified solutal Biot number Bs^* with no heat transfer.

Rearranging Eq. (46) to Ms^* instead of MBs, the following equation is obtained:

$$Ms^* = \frac{5.36 + 7.43Bs^*}{Bs^* \exp(Bs^* + Bs^{*2})\operatorname{erfc}(0.5 + Bs^*)}. \tag{47}$$

It is plotted in the solid line and the values obtained by simulation are featured with the circles in Fig. 4 respectively. Fig. 4 shows that Ms^* has the minimum at the value of Bs^* near 0.85. From the definition of Ms^*, it is obvious that when Ms^* is minimum, τ_c is also minimum with given conditions. It is known that the shorter τ_c is, the faster Marangoni convection sets in. Therefore the critical Bs^* at which the minimum value of Ms^* is

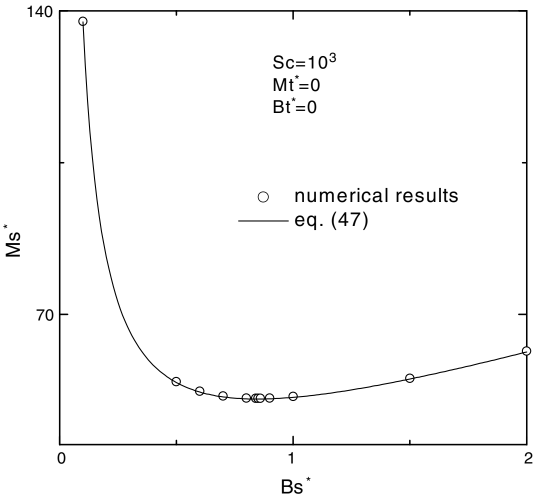

Fig. 4. Modified solutal Marangoni number Ms^* versus modified solutal Biot number Bs^* with no heat transfer.

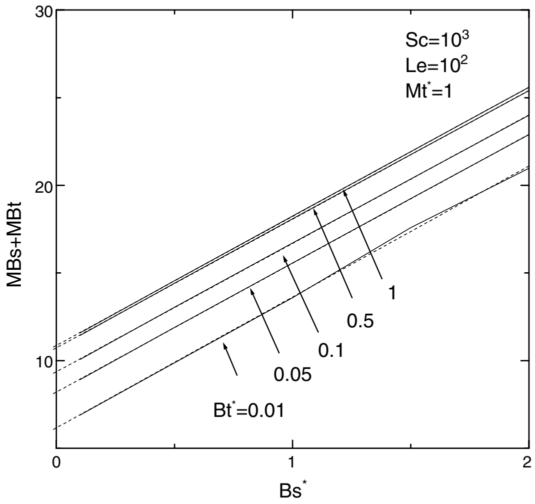

Fig. 6. The sum of re-modified Marangoni numbers versus the solutal Biot number Bs^* for various Bt^*.

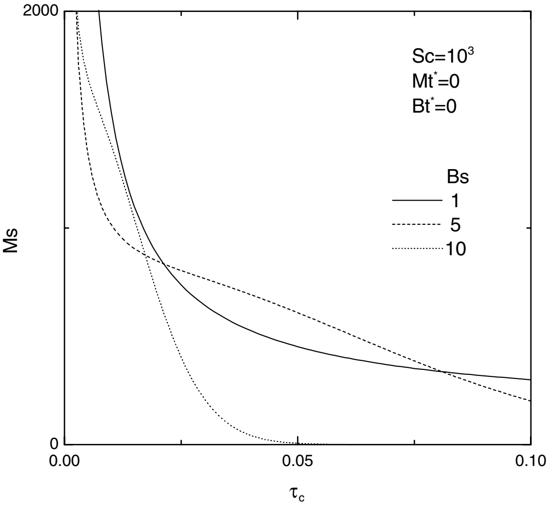

Fig. 5. The effect of Ms on the critical time τ_c for various Bs with no heat transfer.

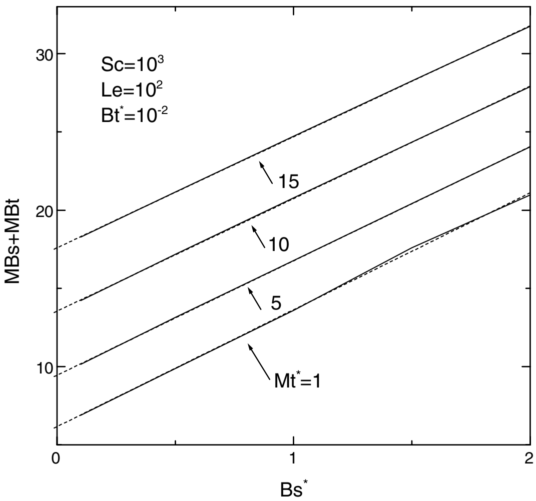

Fig. 7. The sum of re-modified Marangoni numbers versus the solutal Biot number Bs^* for various Mt^*.

obtained makes the fluid layer most unstable with a given condition.

Comparing with Pearson's result [1] Ms becomes infinite instead of a certain value for $Bs = 0$. Pearson used Marangoni number based on the basic gradient. On the other hand, Marangoni number used in the present study is based on the overall concentration difference. Regarding the difference of definition, Rabin [16] pointed out that Marangoni number defined by Pearson included the Biot number.

Fig. 5 shows the effect of Ms on the critical time τ_c for various Bs. It is represented that there is a pair of Ms and a critical value of Bs to make the fluid layer most unstable. For example Bs of 5 is the critical value for the larger values of Ms than 900.

The results in the sense of linear relation of the re-modified parameters to Bs^* are shown in Fig. 6 for various Bt^* and Fig. 7 for various Mt^*, respectively. According to the definition of MBt, it varies with Mt^* and Bt^*. In Fig. 6, the effect of Bs^* on the sum of MBs and MBt are obtained with a fixed Mt^* but a variable Bt^*. In Fig. 7, these are calculated with a fixed Bt^* but a variable Mt^*. Figs. 6 and 7 show that the values of $MBs + MBt$ for given Bt^* and Mt^* with other fixed

Table 1
Coefficients A and B in Eq. (48) for Fig. 6

Bt^*	A	B
10^{-2}	7.47	6.17
5×10^{-2}	7.35	8.21
10^{-1}	7.32	9.37
5×10^{-1}	7.34	10.78
1	7.37	10.86

Table 2
Coefficients A and B in Eq. (48) for Fig. 7

Mt^*	A	B
1	7.47	6.17
5	7.31	9.46
10	7.19	13.55
15	7.10	17.60

parameters are linearly related to Bs^* respectively. The correlations are expressed as follow:

$$MBs + MBt = ABs^* + B, \tag{48}$$

where A and B denote the slop and interceptor of the lines respectively. The coefficients A and B in the above equation are summarized in Table 1 for Fig. 6 and Table 2 for Fig. 7. Using these relations the critical time to mark the onset of convective motion can be found with given conditions.

8. Conclusions

In the present study, the onset of solutal Marangoni convection for absorption process accompanied by heat transfer has been investigated under propagation theory.

The following conclusions are drawn from the present study.

1. Based on the scaling analysis, it is found that the sum of the re-modified Marangoni numbers MBs and MBt have a linear relation to the modified solutal Biot number Bs^*. Using these relations, the critical time τ_c to mark the onset of Marangoni convection can be predicted with given conditions.
2. There is a critical Bs causing the most unstable state of fluid layer. This means that the transport condition of the upper gas phase affects Marangoni instability at the surface.
3. The effect of Sc on the criteria of solutal Marangoni instability is not significant for a larger value than 500.

It is expected that the results from the present study provide a guideline to enhance heat and mass transfer performance during the absorption process. It can be achieved by performing the numerical simulation of Marangoni convection for the whole time domain using the results of the present study, i.e., the velocity and concentration profiles at the critical onset time as the initial condition of numerical analysis and conducting the experiments on the visualization of Marangoni convection.

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