

CONCENTRATION-DEPENDENT, SURFACE-TENSION DRIVEN INSTABILITY IN A HORIZONTAL LIQUID LAYER WITH A DEFORMABLE FREE SURFACE

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The subject of the paper is the problem of the onset of Marangoni-type convection in a thin viscous liquid layer with deformable surface, induced by surface gradients due to the liquid concentration gradients.

1. INTRODUCTION

Surface tension-driven instability problems are among the most interesting ones not only due to their intrinsic academic value but also to their technological importance in fluid phase separation processes such as distillation, absorption and extraction [1, 2], coating and drying processes [3], in the containerless processing of materials in the reduced-gravity environment of a spacecraft [4] as well as in biological systems as a biological cell [5], a tear film on the eye [6] etc. Since the first systematic experiments of thermal convection in a thin liquid layer were performed by BERNARD [7], substantial progress has been made in the study of thermal and/or concentrational instabilities in liquids. Following the pioneering theoretical works of PEARSON [8] on thermal convection in a layer with a free surface and of STERLING and SCRIVEN [9] on interfacial turbulence due to mass transfer across the surface between two liquids, a number of investigators have modified and improved their models. NIELD [10] has included buoyancy effects on thermal convection. SCRIVEN and STEMLING [11] have accounted for capillary waves, SMITH [12] – for gravity waves, and ZEREN and REYNOLDS [13] – for both gravity and buoyancy effects. The liquid layer with two free surfaces has been also considered [14]. The stability of a surface between two layers with finite thickness has been studied [15]. The influence of surface active agents at the interface [16], of nonlinearity of the temperature profile [17], of the surface deformation [18] as well as of the overstability [19, 20] has also been considered.

The main goal of the small disturbance stability analysis is to determine the condition expressed as a critical value of the Marangoni number

$Ma_T = (-d\sigma/dT)\beta_T d^2/\mu D_T$ (in the thermal case) under which the surface tension acting at the surface will produce cellular convection. Here T is the temperature, β_T the thermal gradient across the liquid layer, d the depth of the layer, μ the dynamic viscosity and D_T the thermal diffusivity of the liquid. For example, Ma_T is about 80 in the case of the isothermal wall on which the layer is spread [8].

The onset of solutal Marangoni convection has also been extensively studied by many authors (for references see [1, 3] etc). Here we will mention some papers which are related to the present report. The dynamics and instability of fluid interface was the main topic of the meeting held in 1978 at the Technical University of Denmark in Lingby [21]. The majority of the papers reported there have been translated into Russian and published in [22]. The editors of the publication [22] present a review of the latest important works, including those published in Russian.

Considering the surface-tension driven convection due to concentration gradients, we shall dwell on BRIAN'S works [23, 24], where the effect of Gibbs adsorption is incorporated into hydrodynamic stability analysis. It is shown that Gibbs adsorption can have a profound stabilizing influence on the Marangoni convection. BRIAN [23] proves that the gas-liquid system is always stable to transfer the surface tension-lowering solute from the gas to the liquid phase. Moreover, in the case of desorbing the same kind of solute, the fluid layer is completely stable when the so-called adsorption number $N_A = \Gamma_0/(C_w - C_i)d$ (Γ_0 is the initial excess surface concentration of the solute, $C_w - C_i$ the concentration difference across the layer) exceeds a value equal to 0.05 at which the critical Marangoni number tends to infinity. BRIAN and ROSS [24] have also introduced a penetration-type mass transfer in the stability analysis taking the "frozen" brokenline concentration profile in the unperturbed state. It has been demonstrated that the solute storage in the Gibbs layer may have an important effect on the rate of transfer from the liquid before instability occurs. The theory shows much better agreement with the experimental results obtained by BRIAN *et al.* [25] than the theory based on the constant profile. But the experimental value of the liquid-to-gas phase transfer ratio is still eight times larger than the theoretical one.

Recently, the interfacial turbulence in gas-liquid mass transfer by describing six organic solutes from their aqueous solutions has been studied [26, 27]. The comparison of the experimental results reported there with the theoretical ones based on Brian's theory shows a discrepancy in two or three cases. This is attributed to the time required for the growth of the microturbulence.

Besides, a question still remains. It is not understood why the experimental points of the critical Marangoni number for some solutes like triethylamine lie on the $Ma - N_A$ plot in the area where the adsorption number is more than 0.05 in contrast to Brian's theoretical result pointing that area as an area of absolute stability.

One of the possible reasons for this paradox is the surface deformation which influences the convection and the mass transfer in desorbing processes from a liquid-jet column and wetted-wall column used [26], but it is not taken into account in the stability theory.

In the present paper the effect of the free surface deformation on the onset of the Marangoni convection in a horizontal liquid layer subjected to description or adsorption of a surface-active solute from (to) its surface is studied using the linear stability theory. The main difference between Brian's analysis [23] and the present work is that the boundary condition which represents a solute material balance at the interface consists of additional terms due to its deformability. Besides, the gravity values are also considered. The study shows that as a general rule the surface deformation causes a destabilizing effect and long wave disturbances are most unstable.

2. FORMULATION OF THE PROBLEM

Consider an infinite horizontal layer of viscous liquid of density ρ , kinematic viscosity ν and surface tension σ placed at a rigid wall at $z = 0$. A surface active solute is desorbing from (or adsorbing on) the free surface of the layer into (from) an ambient gas. The motion of the gas is assumed to be negligible. The mass transfer coefficient is supposed to be constant. In the unperturbed state the liquid is at rest and there is a constant concentration gradient of the solute species, i.e.

$$(2.1) \quad \bar{v} = 0, \quad p_0 = p_G + \rho g(d - z), \quad c_0 = c_w - \beta z,$$

where v is the fluid velocity, p_0 the hydrostatic pressure, p_G the gas pressure above the layer, c_0 the unperturbed solute concentration, c_w solute concentration at the bottom of the layer, $(-\beta)$ the concentration gradient, d the liquid depth and g the gravitational acceleration. The coordinate z is measured from the bottom. The constant β is positive in the desorbing case and negative in the adsorbing case.

Restricting our analysis to dilute solutions, we assume all physical properties of the liquid unchangeable except the surface tension which is taken to depend linearly on the surface excess concentration Γ . Then, the governing equations are the equations of mass, momentum, and solute diffusion

$$(2.2) \quad \begin{aligned} (\nabla \cdot \bar{v}) &= 0, \\ \frac{d\bar{v}}{dt} &= \bar{g} - \frac{1}{\rho} \nabla p + \nu \nabla^2 \bar{v}, \\ \frac{dc}{dt} &= D_L \nabla^2 c, \end{aligned}$$

where t is the time, p the dynamic pressure, c the solute concentration in the liquid phase, D_L the diffusion coefficient for the solute in the liquid, ∇ the gradient operator, ∇^2 the Laplacian.

Suppose the equilibrium state is perturbed by a disturbance denoted by primes, i.e.,

$$(2.3) \quad \bar{v} = \bar{v}', \quad p = p_0 + p', \quad c = c_0 + c'.$$

According to the linear stability theory, the vertical component of the velocity w' and the perturbed concentration c' satisfy the following equations:

$$(2.4) \quad \begin{aligned} \left(\frac{\partial}{\partial t} - \nu \nabla^2 \right) \nabla^2 w' &= 0, \\ \left(\frac{\partial}{\partial t} - D_L \nabla^2 \right) c' &= \beta w'. \end{aligned}$$

Employing the cylindrical coordinates (r, φ, z) the Laplacian is expressed here in the form

$$(2.5) \quad \nabla^2 = \frac{\partial^2}{\partial z^2} + L^2, \quad L^2 = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \varphi^2}.$$

The perturbed free surface is presented by

$$(2.6) \quad z = d + \zeta(r, \varphi, t).$$

Involving the Gibbs adsorption into hydrodynamic stability analysis, we first suppose the adsorbed layer on the surface with the excess concentration being the sum

$$(2.7) \quad \Gamma = \Gamma_0 + \Gamma',$$

where Γ_0 is a constant unperturbed value. Second, we assume the surface concentration to depend on the bulk concentration at the surface by the adsorption isotherm

$$(2.8) \quad \Gamma = \delta c_i = \frac{\alpha}{RT} c_i \quad \left(\alpha = - \frac{d\sigma}{dc} \right)$$

with the "Gibbs depth" δ being constant. Using the relation (2.8) twice for unperturbed and perturbed quantities and Eqs. (2.3) and (2.1) for c_i , the expression follows from Eq. (2.7):

$$(2.9) \quad \Gamma = \delta(c_i' - \beta \zeta).$$

Since ζ is small compared to d , the boundary conditions at $z = d + \zeta$ can be expanded about $z = d$ using the Taylor series. Applying Scriven's balance equations [29] and retaining only the linear terms in perturbed relations, we

have the kinematic condition

$$(2.10) \quad w' = \frac{\partial \zeta}{\partial t},$$

a normal force balance

$$(2.11) \quad \Gamma_0 \frac{\partial}{\partial t} (L^2 w') = \rho \left[\frac{\partial}{\partial t} - \nu \left(\frac{\partial^2}{\partial z^2} - 3L^2 \right) \right] \frac{\partial w'}{\partial z} (\rho g - \sigma L^2) L^2 \zeta,$$

a tangential force balance

$$(2.12) \quad -\Gamma_0 \frac{\partial}{\partial t} \left(\frac{\partial w'}{\partial z} \right) = \rho \nu \left(\frac{\partial^2}{\partial z^2} + L^2 \right) w' - \frac{\alpha}{\delta} L^2 \Gamma',$$

and a solute material balance

$$(2.13) \quad -D_L \frac{\partial c'}{\partial z} = H k_c (c' - \beta z) + \frac{\partial \Gamma'}{\partial t} - \Gamma_0 \frac{\partial w'}{\partial z} - D_s L^2 \Gamma',$$

where H is Henry's law constant and D_s the diffusivity of the solute in the interface.

The terms on the left-hand side of Eqs. (2.11) and (2.12) represent the inertial force acting on the material surface. The right-hand parts of these balances consist of viscous, gravity and capillary forces. The terms on the left-hand side of the material balance represent diffusion of the solute to (from) the surface from (to) the liquid phase and the first term on the right-hand-side — the transfer of the solute from (to) the interface to (from) the gas phase. The other three terms represent Gibbs' adsorption. The first of them is the solute within the Gibbs layer, the second one the surface convection and the last on the surface diffusion.

On the wall the boundary conditions for the velocity are

$$(2.14) \quad w' = \frac{\partial w'}{\partial z} = 0 \quad \text{at} \quad z = 0,$$

where the second condition is received by utilizing the continuity equation. The condition for the concentration may be in two forms

$$(2.15) \quad c = 0 \quad \text{or} \quad \frac{\partial c}{\partial z} = 0$$

named "conducting" and "insulating", respectively. The experimental realization of both cases is discussed by BRIAN [23]. We restrict our analysis to the "conducting" case in which the bottom wall could be imagined to be a membrane with very high permeability through which the solute diffuses from an intensively stirred solution.

The solution of the system (2.4) with the boundary conditions (2.10)–(2.15) is written in the form

$$(2.16) \quad [w', c', \zeta] = [w(z), c(z), z] \mathcal{L}_l \exp \omega t,$$

where $\omega = \omega_r + i\omega_i$ is the wave frequency and \mathcal{L}_l the cylindrical harmonic function of order l satisfying the equation

$$(2.17) \quad (L^2 + l^2) \mathcal{L}_l = 0.$$

Before substituting Eq. (2.16) into Eq. (2.14) and the boundary conditions, they are rewritten in a dimensionless form using d , d^2/D_L , D_L/d and βd as scaling quantities for length, time, velocity and concentration, respectively. Reserving the same notations for the variables, we have ($D = d/dz$)

$$(2.18) \quad [\text{Sc}^{-1} \omega - (D^2 - l^2)] (D^2 - l^2) W = 0,$$

$$(2.19) \quad [\omega - (D^2 - l^2)] C = W$$

with the boundary conditions at $z = 1$

$$W - \omega Z = 0,$$

$$(2.20) \quad -\omega \gamma \text{Cr} \text{Sc}^{-1} l^2 W = \text{Cr} [\text{Sc}^{-1} \omega - (D^2 - 3l^2)] DW + (\text{Bo} + l^2) l^2 Z,$$

$$(D^2 + l^2) W + \text{Ma} l^2 (C - Z) + \omega \gamma \text{Sc}^{-1} DW = 0,$$

$$DC - N_A DW + (\text{Bi} + l^2 S + \delta \omega) (C - Z) = 0,$$

and at $z = 0$

$$(2.21) \quad W = DW = C = 0.$$

The following dimensionless groups are introduced:

$$\gamma = \frac{\Gamma_0}{\rho d}, \quad \delta = \frac{\delta}{d}, \quad S = \frac{D_s \delta}{D_L d},$$

$$N_A = \frac{\Gamma_0}{\beta d^2} \quad \text{the adsorption number,}$$

$$\text{Cr} = \frac{\rho v D_L}{\sigma d} \quad \text{crispation number,}$$

$$\text{Ma} = \frac{\alpha \beta d^2}{\rho v D_L} \quad \text{Marangoni number,}$$

$$\text{Bi} = \frac{H k_G d}{D_L} \quad \text{diffusion Biot number,}$$

$$\text{Bo} = \frac{\rho g d^2}{\sigma} \quad \text{Bond number,}$$

$$\text{Sc} = \frac{\nu}{D_L} \quad \text{Schmidt number.}$$

The parameter γ is the ratio of the solute mass at the surface unit element to the liquid mass of the column of the layer beneath this element. It could be named a surface mass number. This number as well as the Schmidt number and the depth ratio $\bar{\delta}$ disappears in Eq. (2.20) in the case of stationary convection, when $\omega = 0$ and will not affect neutral stability curves and, consequently, the value of the critical Marangoni number. But these groups are expected to influence the rate of growth of an unstable disturbance in the case of oscillatory instability.

The quantity S represents the relative importance of the surface diffusion. As shown by BRIAN [23], the surface diffusion has a small effect on the neutral stability in comparison with the surface convection presented by the adsorption number. This number gives the ratio of the solute flux in the Gibbs layer due to surface convection to the flux from the liquid phase to the surface. Its sign depends on the sign of the product of α and β and always coincides with the sign of the Marangoni number. Both numbers are positive in the case of desorbing a surface-tension-lowering solute (when $\alpha > 0$, $\beta > 0$), or adsorbing a solute which increases the surface tension ($\alpha < 0$, $\beta < 0$). They are negative when a surface-tension-lowering solute adsorbs ($\alpha > 0$, $\beta < 0$) or surface-tension-increasing solute desorbs ($\alpha < 0$, $\beta > 0$).

The deformability of fluid interfaces is characterized by the crispation number which is negligibly small only for thick layers and interfaces with high surface tension. The Bond number shows the relative importance of gravity waves. The diffusion Biot number represents the ratio of the liquid phase mass transfer resistance to the gas phase resistance.

3. SOLUTION AND RESULTS

We assume that instability sets in as stationary convection and thus $\omega = 0$. The general solution of the system (2.18)–(2.21) with $\omega = 0$ has been obtained. The solubility condition gives the expression for the Marangoni number

$$(3.1) \quad \text{Ma} = \frac{8l(\sinh l \cosh l - l) [l \cosh l + (\text{Bi} + l^2 S) \sinh l]}{8\text{Cr} l^5 \cosh l + (\sinh^3 l - l^3 \cosh l) - 4N_a l^2 \sinh l (\sinh^2 l - l^2)}$$

As expected, this formula expresses in an explicit form Brian's result for a flat liquid surface when $\text{Cr} = 0$ (then Bo disappears). If $N_a = S = 0$ additionally,

the results is identical to that of PEARSON'S for the conducting case [8]. For $S = Bo = N_A = 0$ the expression (3.1) coincides with the result of LIN and BRENNER (see (2.12) in [6]) due to the fact that by definition their solutal Marangoni number is twice smaller than the number defined here.

To compare the results obtained in the case of a deformable free surface with those for a nondeformable one, we present in Fig. 1 neutral stability

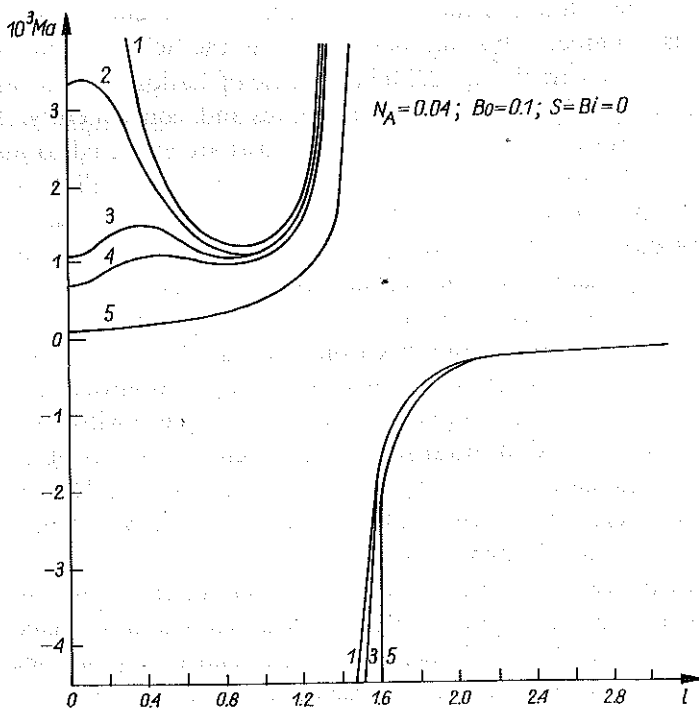


FIG. 1. 1 - $Cr = 0$, 2 - 0.00002, 3 - 0.000063, 4 - 0.0001, 5 - 0.001.

curves for various values of the crispation number and the same values of N_A , Bi and S (i.e., for $N_A = 0.04$, $Bi = S = 0$), as have been taken by BRIAN [23] in his Fig. 1. In our calculations for $Cr \neq 0$, we take $Bo = 0.1$. The branches of the curves corresponding to negative Ma with positive N_A will be disregarded as they have no physical sense. From the other side, for negative values of N_A the Marangoni number is always positive, what also corresponds to a situation of of no physical significance. Hence the gas liquid system with a deformable free surface is always stable to transfer of a surface-tension-lowering solute from the gas to the liquid phase as well as to desorbing a surfactant which raises the surface tension.

It is seen from Fig. 1 that the behaviour of the curves for small wave numbers changes considerably with when the crispation number increases. In

contrast to the case of nondeformable interface, at $Cr \neq 0$ the Marangoni number does not go to infinity when l approaches zero. This follows from the expansion of Ma in a series in the vicinity of $l = 0$ given by the formula

$$(3.2) \quad Ma = \frac{2Bo}{3Cr}(1+Bi) + \frac{2}{3Cr} \left[(1+Bi) \left(1 - \frac{2}{15}Bo \right) + \left(\frac{1}{3} + S \right) Bo - \frac{Bo}{6Cr}(1+Bi) \left(\frac{1}{20} - N_A \right) \right] l^2 + O(l^4).$$

As the region below each curve represents a stable state, the lowest point on the curve determines the critical value Ma_c of the Marangoni number and the corresponding critical wave number l_c . When the crispation number increases from zero, Ma_c decreases slowly from 1170 at $l_c = 0.91$ to 1057 at $l_c = 0.87$, occurring at $Cr = 0.6305 \times 10^{-4}$. At that value the critical wave number changes abruptly to zero and the minimum value of Ma is given by the first term of the series (3.2):

$$(3.3) \quad Ma_c^* = \frac{2Bo(1+Bi)}{3Cr}.$$

This formula gives in an explicit form the dependence of Ma_c on the governing parameters for every $Cr \geq Cr^*$. It is obvious that this critical Marangoni number decreases quickly when Cr increases further and, for instance, takes the value 66.7 for a relatively large but still reasonable value of the crispation number equal to 10^{-3} (with $Bo = 0.1$ and $Bi = 0$). It is worth noting that Ma_c does not depend on the adsorption number as well as on S . Indeed, the value of Cr at which a sudden change of l_c occurs varies with Bi and N_A . The influence of Bi may be demonstrated in the case under consideration where Cr grows from 0.6305×10^{-4} at $Bi = 0$ to 0.708×10^{-4} at $Bi = 1$ and 0.813×10^{-4} at $Bi = 100$. For these values of the Biot number the critical Marangoni number is plotted versus the crispation number in Fig. 2. It is seen that Ma_c increases with increasing Bi and that the surface deformation is important when the crispation number is of order of 10^{-5} and higher.

This sample case is interesting with another feature. The wave number l (say, l_{inv}) for which the Marangoni number changes a sign varies a little with Cr at fixed N_A , while it goes down rapidly when the adsorption number increases. For example, at $Cr = 10^{-5}$ l_{inv} changes from 1.47 at $N_A = 0.04$ to 0.28 at $N_A = 0.05$ and 0.14 at $N_A = 0.06$. But the main difference between the deformable surface and the flat one is that at $Cr \neq 0$ the inversion wave number l_{inv} does not become zero and the curves do not rise and go to infinity as N_A approaches 0.05 as it occurs for nondeformable interfaces. The point l_{inv} is determined from the condition that the denominator of Eq. (3.1) be zero for fixed Cr , N_A and Bo . It is easy to show that for $Cr = 0$ (with Bo disappearing) l_{inv} goes to zero and Ma tends to infinity when N_A approaches

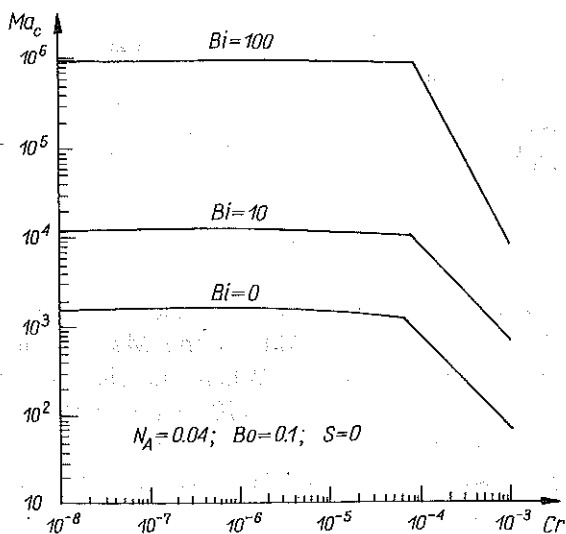


FIG. 2.

0.05. This proves BRIAN's result [23]. But for $Cr \neq 0$, the inversion number is different from zero for any finite N_A . So, in the case of deformable surface there is an interval $[0, l_{inv}]$ in which the Marangoni number takes positive and bounded values.

As the neutral stability curves shift to the left with increasing N_A (and decreasing l_{inv}), the minimum of the function $Ma(l)$ appears at smaller values of l . Then, it may be expected that there is a value of N_A (say, N_A) which, after reaching the absolute minimum of Ma , will be only at the point $l = 0$ irrespective to the crispation number.

The condition for a local minimum of Ma to exist at $l = 0$ is derived easily from the series (3.2). The crispation number must be larger than the quantity

$$(3.4) \quad \overline{Cr} = \frac{Bo^2(1+Bi)(1-20N_A)}{120 \left[(1+Bi) \left(1 - \frac{2}{15} Bo \right) + \left(\frac{1}{3} + S \right) Bo \right]}$$

if the denominator of the fraction is different from zero. This denominator is really positive for physically reasonable values of $Bi \geq 0$, $S \geq 0$ and $Bo < 7.5$. It is seen that \overline{Cr} is not positive for $N_A \geq 0.05$ and hence in this case the local minimum of Ma exists for every $Cr > 0$. A detailed analysis shows that this minimum presented by Eqs. (3.3) is the absolute one when $N_A \geq 0.05$. Thus there are two intervals of changing the adsorption number with a different behaviour of the system. N_A increasing from zero to the value 0.05 has a stabilizing effect on the system only for sufficiently small values of the crispation number. If Cr is larger than \overline{Cr} for which a sudden change of the

critical wave number to zero occurs, the adsorption number doesn't influence more the stability of the layer. In the last case as well as when $N_A \geq 0.05$ the critical Marangoni number is presented by Eq. (3.3) and depends strongly on the crispation number. It is obvious that the surface deformation destabilizes the system and long-length waves are the most unstable ones.

The present analysis makes reasonable the appearance of points presenting critical Marangoni numbers at the $Ma_c - N_A$ plot in the area where the adsorption number is larger than 0.05 as reported by IMASHI et al. [27]. Those critical Marangoni numbers are probably influenced by the surface deformation which, in principle, is not avoidable in their experiments with a liquid-jet column. Regarding the influence of the other parameters, we see that Bo and Bi are directly included in Eq. (3.3) and cause an increase of the critical Marangoni number when they increase. The parameter S has a relatively small effect on the stability of the layer because of the little change of the expression $(Bi + l^2 S)$ in Eq. (3.1) for fixed Bi and variable l in the vicinity of the critical wave number which is smaller and in many cases much smaller than one.

In conclusion, the present analysis shows that in the desorbing processes of a surface-tension-lowering solute from the horizontal liquid layer, the deformation of the gas-liquid surface has a destabilizing effect on the onset of Marangoni convection while the gravity stabilizes the system.

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STRESZCZENIE

**NIESTATECZNOŚĆ POZIOMEJ WARSTWY CIECZY Z POWIERZCHNIĄ
SWOBODNĄ, WYWOŁANA NAPIĘCIEM POWIERZCHNIOWYM W WARUNKACH
ZMIENNEJ KONCENTRACJI**

W pracy omówiono powstawanie konwekcji typu Marangoniego w cienkiej warstwie cieczy lepkiej ze swobodną powierzchnią, wywołanej gradientami napięcia powierzchniowego indukowanymi gradientami koncentracji cieczy.

РЕЗЮМЕ

**НЕУСТОЙЧИВОСТЬ ГОРИЗОНТАЛЬНОГО СЛОЯ ЖИДКОСТИ
СО СВОБОДНОЙ ПОВЕРХНОСТЬЮ ВЫЗВАННАЯ ПОВЕРХНОСТНЫМ
НАПРЯЖЕНИЕМ В УСЛОВИЯХ ПЕРЕМЕННОЙ КОНЦЕНТРАЦИИ**

В работе обсуждено возникновение конвекции типа Марангони в тонком слое вязкой жидкости со свободной поверхностью, вызванной градиентами поверхностного напряжения индуцированными градиентами концентрации жидкости.

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