Int. J. Heat Mass Transfer. Vol. 31, No. 10, pp. 1979–1982, 1988
Printed in Great Britain

0017-9310/88 $3.00+0.00

Free surface convection in a bounded cylindrical geometry: note on the role of surface adsorption and solute accumulation at the air–liquid interface

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(*Received* 8 *September* 1987 *and in final form* 24 *February* 1988)

Abstract—The onset of steady two-component Bénard convection in a cylindrical geometry with a free surface is studied with due consideration given to the adsorption and eventual accumulation of a solute at this free surface. To a first approximation the effect of the latter processes upon the critical Marangoni and Rayleigh number is given.

1. INTRODUCTION

RECENTLY, Narayanan and co-workers [1–3] studied the onset and weakly non-linear development of steady Bénard convection in a cylindrical geometry. They reported some interesting mathematical findings when buoyancy and surface tension gradients act simultaneously. On the other hand, one of the present authors has also, recently, treated the onset of buoyancy-driven instability in a two-component fluid cylindrical layer [4, 5]. It was shown that the aspect ratio and the second component either compete or cooperate in delaying the onset of instability thus enhancing or reducing the buoyancy force. In this paper we extend the results reported in refs. [4, 5] by letting the upper surface of the cylinder become free. As we shall restrict consideration to the onset of steady convection and not consider the development of cellular patterns, the assumption of initially axisymmetric disturbances for flat cylinders is compatible with experiment [6, 7] and with the assumptions used by Narayanan and co-workers [1, 2]. Then we point out up to what extent adsorption and eventual accumulation of solute, the second component, at the free surface affect the onset of instability.

2. FORMULATION OF THE PROBLEM

We consider a motionless binary liquid mixture in a vertical cylinder of height L and horizontal diameter $2r$. When the initially homogeneous layer is heated from below the Soret effect distributes the components in accordance with the value given to the thermal gradient ($\Delta T/L$, where T denotes temperature) until the mass and heat fluxes balance each other. If the upper air–liquid free surface is impervious to mass transfer and we neglect evaporation the solute may, however, be adsorbed and may, eventually accumulate there.

In the simplest case the thermohydrodynamic equations describing the evolution of small disturbances upon the quiescent state are [8]

$$\operatorname{div}\mathbf{u} = 0 \tag{1}$$

$$\partial\mathbf{u}/\partial t = -(1/\rho_0)\operatorname{grad}p+(\delta\rho/\rho_0)g\mathbf{k}+\nu\nabla^2\mathbf{u} \tag{2}$$

$$\partial\theta/\partial t - w(\Delta T/L) = \kappa\nabla^2\theta \tag{3}$$

$$\partial n/\partial t - w(\Delta N_1/L) = \mathbf{D}\nabla^2 n + N_1^0 N_2^0 \mathbf{D}'\nabla^2\theta \tag{4}$$

together with the following boundary conditions (b.c.):

at the bottom ($z = 0$)

$$\mathbf{u} = 0 \tag{5}$$

$$\theta = 0 \tag{6}$$

$$\mathbf{D}(\partial n/\partial z) + N_1^0 N_2^0 \mathbf{D}'(\partial\theta/\partial z) = 0; \tag{7}$$

at the free surface for simplicity assumed undeformable ($Z = L$)

$$w = 0 \tag{8}$$

$$\partial\theta/\partial z = 0 \tag{9}$$

$$-\mathbf{D}(\partial n/\partial z) - N_1^0 N_2^0 \mathbf{D}'(\partial\theta/\partial z) = k_1 n - k_{-1}\gamma \tag{10}$$

$$\rho_0\nu(\partial\mathbf{u}^s/\partial z) = \operatorname{grad}_s\sigma \tag{11}$$

$$(\partial\gamma/\partial t) + \Gamma^0\operatorname{div}_s\mathbf{u}^s - \mathbf{D}_s\nabla^2\gamma = k_1 n - k_{-1}\gamma. \tag{12}$$

We also assume the lateral walls to be rigid, i.e. $\mathbf{u} = 0$ at the periphery, and the following two equations of state

$$\rho = \rho_0(1-\alpha\theta+\boldsymbol{\beta} n) \tag{13}$$

$$\sigma = \sigma_0 + (\partial\sigma/\partial T)\theta + (\partial\sigma/\partial N_1)n. \tag{14}$$

For later convenience and universality in the argument we rescale the problem in order to have dimensionless quantities and to sort out the relevant physical parameters. As in ref. [4] we choose the following scales: unit length, r; unit time, r^2/κ, with κ the thermal diffusivity (thermometric conductivity); velocity, $\kappa L/r^2$; temperature, ΔT; concentration (mass fraction) of, say, component 'one', ΔN_1; surface con-

NOMENCLATURE

A_1, A_1^*	adsorption number (according to units used)
$\mathbf{D}$	mass diffusivity
$\mathbf{D}'$	Soret mass diffusivity
$\mathbf{D}_s$	surface mass diffusivity
E, E^*	elasticity (solutal Marangoni) number (according to units used)
f	aspect ratio
g	gravitational acceleration
H, H^*	surface excess-solute number (according to units used)
h	dimensionless height
J_n	nth-order Bessel function
k, k^*	Fourier wave number (according to units used)
$\mathbf{k}$	unit vector in the z-direction
k_1, k_{-1}	adsorption, desorption coefficients
L	height of liquid layer
M, M^*	thermal Marangoni number (according to units used)
N, N^*	concentration near surface relative to solute gradient
N_1	mass fraction of component 'one'
N_1^0, N_2^0	mass fraction reference values
n	disturbance of N_1
P	Prandtl number
p	pressure field disturbance
R, R^*	Rayleigh number (according to units used)
r	radius of cylinder
r_D	inverse Lewis number
r_D^s	surface inverse Lewis number
S	Soret number (buoyancy ratio)
T	temperature
$\mathbf{u}$	velocity field, components (u, v, w) or (u_ρ, u_ϕ, w)
$\mathbf{u}^s$	surface velocity.

Greek symbols

α	thermal expansion coefficient of liquid mixture
β	volume expansion due to variation of the mass fraction N_1
Γ	mass fraction of component 'one' at free surface
γ	disturbance of Γ
Θ	temperature disturbance
κ	thermal diffusivity of liquid mixture
ν	kinematic viscosity of liquid mixture
σ	air–liquid surface tension
ρ	density of fluid or radial coordinate (in different paragraphs).

centration, $\Gamma^0\Delta N_1/N_1^0$; and pressure, $\rho_0\nu\kappa L/r^3$. Thus in dimensionless form the equations are (no confusion is expected although we use the same notation for dimensional and dimensionless quantities)

$$\operatorname{div}\mathbf{u} = 0, \quad \mathbf{u} = (u, v, w) \tag{15}$$

$$P^{-1}(\partial\mathbf{u}/\partial t) = -\operatorname{grad} p + R^*\theta\mathbf{k} + R^*Sn\mathbf{k} + \nabla^2\mathbf{u} \tag{16}$$

$$\partial\theta/\partial t = \nabla^2\theta + w \tag{17}$$

$$\partial n/\partial t = r_D\nabla^2(n-\theta) + w \tag{18}$$

together with the b.c.

at $z = 0$

$$\mathbf{u} = \theta = \partial(n-\theta)/\partial z = 0; \tag{19}$$

at $z = L/r = h$

$$w = \partial\theta/\partial z = 0 \tag{20}$$

$$\partial n/\partial z = A_1^*(\gamma - n) \tag{21}$$

$$\partial\mathbf{u}^s/\partial z = -M^*\operatorname{grad}_s\theta - E^*r_D\operatorname{grad}_s\gamma \tag{22}$$

$$H^*(\partial\gamma/\partial t + N^*\operatorname{div}_s\mathbf{u}^s - r_D^s\nabla^2\gamma) = n - \gamma; \tag{23}$$

where we have introduced the following groups: $P = \nu/\kappa$; $r_D = \mathbf{D}/\kappa$; $R^* = \alpha g r^4\Delta T/\nu\kappa L$, $S = -\beta\Delta N_1/\alpha\Delta T$, $A_1^* = k_1 r/\mathbf{D}$, $H^* = \kappa\Gamma^0/k_1 N_1^0 r^2$, $N^* = LN_1^0/r\Delta N_1$, $h = L/r$, $r_D^s = \mathbf{D}_s/\kappa$, $M^* = -(\partial\sigma/\partial T)r^2\Delta T/\rho\nu\kappa L$ and $E^* = -(\partial\sigma/\partial\Gamma)\Gamma^0 r^2\Delta N_1/\rho\nu\mathbf{D}LN_1^0$. H^* accounts for the excess-solute accumulation at the air–liquid interface.

3. RESOLUTION OF THE PROBLEM AND RESULTS

As in ref. [4] we have used a Galerkin method with cylindrical coordinates (ρ, ϕ, z) and the following trial solutions:

$$u_\rho = z(3z-2h)\cos n\phi\, U(\rho) \tag{24}$$

$$u_\phi = z(3z-2h)\sin n\phi\, V(\rho) \tag{25}$$

$$w = u_z = -z^2(z-h)\cos n\phi\, W(\rho) \tag{26}$$

$$\theta = z(z-2h)\cos n\phi\, W(\rho) \tag{27}$$

$$n = 2zh\cos n\phi\, W(\rho) \tag{28}$$

$$\gamma = 2h^2\cos n\phi\, W(\rho) \tag{29}$$

with

$$U(\rho) = -[J_n'(k^*\rho) - J_n'(k^*)\rho^{n+1}]/k^*J_n(k^*) \tag{30}$$

$$V(\rho) = n[J_n(k^*\rho)/\rho - J_n(k^*)\rho^{n+1}]/k^{*2}J_n(k^*) \tag{31}$$

$$W(\rho) = [J_n(k^*\rho)/J_n(k^*)] - \rho^n. \tag{32}$$

Note that u_r, u_ϕ and w satisfy continuity equation (1) so that

$$U' + U/\rho + nV/\rho - W = 0. \tag{33}$$

Here and earlier a prime denotes a derivative with respect to the corresponding argument.

Then using the above indicated trial functions the standard Galerkin method [9] yields the neutral stability loci. As the analytical expressions of these loci involve rather cumbersome, albeit elementary relations between all the parameters in the problem we shall report here the results found in table and figure form only. Note that in ref. [4] we used two groups of scales (for tall and flat cylinders, respectively). Here the second set of scales can be obtained by writing

$$f = 1/h, \quad kf = k^*, \quad R = \alpha g L^3 \Delta T/\nu\kappa$$

$$M = -(\partial\sigma/\partial T) L \Delta T/\rho\nu\kappa$$

$$E = -(\partial\sigma/\partial\Gamma)\Gamma^0 L \Delta N/\rho\nu D N_1^0$$

$$H = \kappa\Gamma^0/k_1 N_1^0 L^2$$

$$N = N_1^0/\Delta N_1$$

and

$$A_1 = k_1 L/D.$$

In Table 1 we provide the critical values for the solutal (elasticity) Marangoni number, E, when we set to zero the Rayleigh, R, the thermal Marangoni, M, numbers and the buoyancy ratio, S. Values of E are given as we vary the aspect ratio f as well as the excess-solute number, H. The latter has a slightly stabilizing effect. With data from ref. [10] H is generally in the range 10^{-6}–10^{-1}.

Figure 1 depicts the neutral stability curves in the plane (E, H) as we vary the Rayleigh and thermal Marangoni numbers.

Figure 2 shows how the (E, R) neutral stability

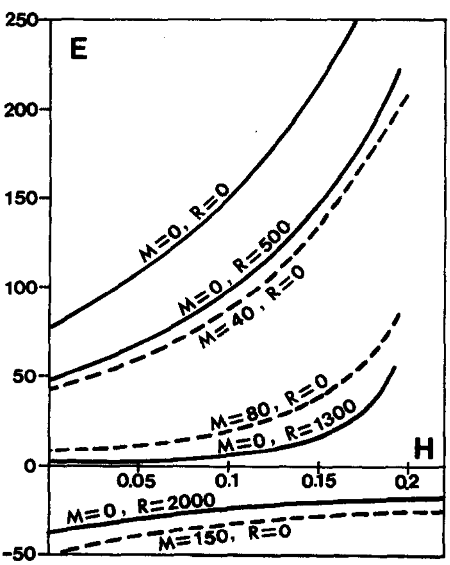

FIG. 1. Thermosolutal convection with a free surface. Neutral stability lines for the onset of solutal convection when we have excess-solute accumulation at a free surface. Values of the solutal (elasticity) Marangoni and excess-solute numbers are given for various choices of the Rayleigh (R) and thermal Marangoni (M) numbers.

Table 1. Critical solutal (elasticity) Marangoni numbers for various aspect ratios of the cylinder, f, and the excess-solute number, H. j is the number of zeros of the Bessel function $J_0(\rho)$. $R = M = S = 0$, $A_1 = r_D = r_D^s = 0.01$ and $N = 10$

	$H = 0.00$		$H = 0.05$		$H = 0.10$	
f	E	j	E	j	E	j
2.8	79.6	2	105.9	1	151.6	1
4	78.8	3	106.9	2	162.4	2
5.2	78.6	4	108.0	3	150.7	2
6.4	78.4	4	109.1	4	150.7	3
7.6	78.2	5	108.2	4	153.6	4
8.8	78.2	6	107.8	5	150.8	4
10	78.2	7	107.8	6	150.7	5

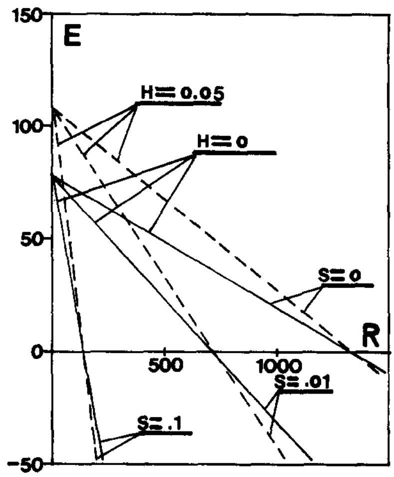

FIG. 2. Thermosolutal convection with a free surface. Neutral stability lines for the onset of instability as we vary the excess-solute number, H, and the buoyancy ratio, S. E is the solutal (elasticity) Marangoni number and R the Rayleigh number.

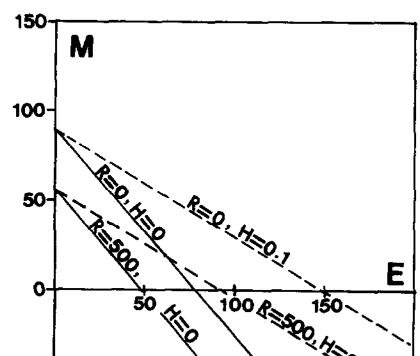

FIG. 3. Thermosolutal convection with a free surface. Neutral stability lines in the plane of both Marangoni numbers, M and E, as we vary the Rayleigh and the excess-solute accumulation numbers, R and H, respectively.

lines are affected by the values taken by the excess-adsorption parameter, H. We also show here the influence of the buoyancy ratio. It appears that H has no influence upon the critical values of the Rayleigh number whereas the buoyancy ratio does not alter the critical elasticity number.

Figure 3 depicts the neutral stability lines in the $(M,$

E) plane as we vary the Rayleigh and the excess-solute numbers. Again the latter plays a stabilizing role whereas the former is indeed destabilizing when the layer is heated from below.

Finally, within a reasonable 15% deviation and limiting ourselves in each reference to the appropriate case, we have recovered earlier reported results [1–3, 8, 11, 12].

Acknowledgements—The first author wishes to express his gratitude to the Spanish Science Policy General Directorate for a predoctoral fellowship that permitted him to carry on the work at U.N.E.D. Both authors also acknowledge the sponsorship of the Stiftung Volkswagenwerk and of CAICYT (Spain).

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CONVECTION AVEC SURFACE LIBRE DANS UNE GEOMETRIE CYLINDRIQUE: NOTE SUR LE ROLE DE L'ADSORPTION DE SURFACE ET DE L'ACCUMULATION DE SOLUTE A L'INTERFACE AIR–LIQUIDE

Résumé—La mise en place de la convection de Benard avec deux composants, dans une géométrie cylindrique, avec une surface libre, est étudiée en considérant l'adsorption et l'accumulation éventuelle d'un soluté à cette surface libre. En première approximation, on donne l'effet de ces mécanismes sur les nombres critiques de Marangoni et de Rayleigh.

KONVEKTION IN EINER BEGRENZTEN ZYLINDRISCHEN GEOMETRIE MIT FREIER OBERFLÄCHE: ANMERKUNG ÜBER DEN EINFLUSS DER OBERFLÄCHENADSORPTION UND DER ANREICHERUNG DES GELÖSTEN STOFFES AN DER LUFT–FLÜSSIGKEIT-GRENZFLÄCHE

Zusammenfassung—Das Einsetzen der stationären Bénard-Konvektion zweier Komponenten in einer zylindrischen Geometrie mit freier Oberfläche wird betrachtet. Besondere Aufmerksamkeit gilt dabei der Adsorption und der möglichen Anreicherung des gelösten Stoffes an der freien Oberfläche. Für den Einfluß des letzteren Prozesses auf die kritische Marangoni- und Rayleigh-Zahl wird eine erste Abschätzung gegeben.

КОНВЕКЦИЯ В ОГРАНИЧЕННОМ ОБЪЕМЕ ЦИЛИНДРИЧЕСКОЙ ФОРМЫ СО СВОБОДНОЙ ПОВЕРХНОСТЬЮ: РОЛЬ ПОВЕРХНОСТНОЙ АБСОРБЦИИ И ПРОЦЕССА НАКОПЛЕНИЯ РАСТВОРЕННОГО ВЕЩЕСТВА НА ГРАНИЦЕ РАЗДЕЛА ВОЗДУХ-ЖИДКОСТЬ

Аннотация—Исследуется возникновение стационарной конвекции Бернара, вызванной двумя механизмами, в объеме цилиндрической формы со свободной поверхностью. Особое внимание обращено на адсорбцию и возможное накопление растворенного вещества на этой поверхности. В первом приближении показано влияние этих процессов на критические числа Марангони и Рэлея.