

Stability of convection in a vertical binary fluid layer with an adverse density gradient

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When a horizontal temperature gradient is imposed across a vertical layer of a binary fluid with a negative Soret coefficient, the coupling of convection and the Soret effect give rise to a steady-state adverse density gradient along the layer. In this paper we shall experimentally analyze, using different mixtures of ethanol and water, the stability of this situation. We find that it remains stable for density gradients several orders of magnitude higher than the critical value corresponding to an isothermal fluid provided that the Grashof number exceeds a critical value. The stability diagram is delineated. [S1063-651X(99)00101-4]

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When a temperature difference is imposed between the two vertical sidewalls of a thin rectangular cavity, a Hele-Shaw cell, enclosing a binary mixture, the horizontal separation due to the Soret effect combines with the vertical convective currents giving rise to a steady-state concentration gradient along the cell. This phenomenon is known as the thermogravitational effect [1,2]. If the Soret coefficient of the mixture is negative, an adverse density gradient is established. Although many studies have been devoted to convection in a Hele-Shaw cell [3–5], the analysis of the stability of the thermogravitational steady state with an adverse stratification has not yet been accomplished. In this paper, we experimentally study this problem. The obtained results may be relevant in oceanography and crystal growth processes.

Let us consider a vertical layer of binary fluid enclosed in a slender Hele-Shaw slot of height h , gap width d , and breadth b , being $h \gg b \gg d$. If a temperature difference is imposed between the vertical sidewalls, the mass flux referred to the less dense component is given by

$$\mathbf{J} = \rho D [\nabla c - S_T c (1 - c) \nabla T], \quad (1)$$

where c is the concentration of this component in the mass fraction, T is the temperature, ρ is the density, D is the ordinary diffusion coefficient, and S_T is the Soret coefficient. For small separations, the vertical diffusion flux in this equation is negligible and also the product $c(1 - c)$ can be considered constant. On the other hand, for small Grashof numbers, with the exception of small regions near the cell ends, the regimen is purely conductive. Under these conditions, the equations that govern the thermogravitational steady state in the

Boussinesq approximation are

$$-\frac{\partial p}{\partial z} - \rho g + \eta \frac{\partial^2 w}{\partial x^2} = 0, \quad (2)$$

$$\frac{\partial^2 T}{\partial x^2} = 0, \quad (3)$$

$$w \frac{\partial c}{\partial z} = D \frac{\partial^2 c}{\partial x^2}, \quad (4)$$

$$\rho = \rho_0 [1 - \beta(T - T_0) - \gamma(c - c_0)], \quad (5)$$

where x and z are, respectively, the horizontal and vertical coordinates, p is the pressure, g is the gravitational acceleration, η is the dynamic viscosity, and w is the velocity in the vertical direction. The coefficients β and γ are the coefficients of thermal expansion and of the variation of density with concentration, respectively. All thermophysical properties are to be taken at the mean temperature.

Equations (1)–(5), together with the corresponding boundary conditions, have been addressed in previous works [2,6]. According to the results obtained, the steady-state vertical concentration gradient is uniform along the fluid layer and is given by

$$\text{Ra} = 504SA^4, \quad (6)$$

where A is the aspect ratio, $A = h/d$, and Ra and S are, respectively, the solutal Rayleigh number and the buoyancy ratio:

$$\text{Ra} = \frac{\gamma g h^3 \Delta c}{\nu D} \quad \text{and} \quad S = \frac{S_T c (1 - c) \gamma}{\beta} \quad (7)$$

where Δc is the separation between the upper and lower ends of the cell and ν is the kinematic viscosity.

Expressions (6) and (7) show that the Rayleigh number and the separation are independent of the imposed temperature difference ΔT between the walls of the cell. Experimental results from previous works [6,7] show that these equations correctly describe the thermogravitational steady state.

According to Eqs. (6) and (7), for positive S_T values $\Delta c > 0$ and hence the less dense component becomes enriched in the upper part of the column. Conversely, for $S_T < 0$ it is the denser component that becomes enriched in the upper part, giving rise to an adverse density gradient. Analysis of the stability of this situation has not yet been accomplished either theoretically or experimentally. In the case of a quiescent isothermal fluid layer, the correspond-

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TABLE I. Rayleigh number and critical Grashof number for different mixtures of ethanol+water. c , concentration in mass fraction of ethanol.

c (%)	$Ra / 10^{11}$	$\Delta T / K$	Gr_c
6.41	2.60	16.5	226.0
11.51	3.27	23.0	276.0
22.04	1.58	13.5	144.0
25.87	1.13	9.9	100.0

ing critical Rayleigh number is given [3] by $Ra_c = (12\pi^2 d^2/b^2)A^4$. For the dimensions of our cell (see below), $Ra_c = 1.48 \times 10^9$. In what follows, we shall experimentally determine the dependence of the critical Rayleigh number on the Grashof number ($Gr = \beta g d^3 \Delta T / \nu^2$) in the thermogravitational problem.

Experiments were performed in an annular slot with a mean circumference length much larger than the gap width. The slot is arranged between two stainless-steel coaxial cylinders of height $h = 42$ cm and inner and outer radii of 0.317 cm and 0.510 cm, respectively. The annular gap width is thus $d = 0.193$ cm and the aspect ratio is $A = 218$. Different mixtures of ethanol and water were used as working liquids. The vertical concentration gradient was determined from the difference in refractive indices of the removed samples at the upper and lower ends of the cell, using the value of the derivative of the index with respect to the concentration at the temperature of the refractometer. This derivative was determined for each mixture from the adjustment of the measurements of the refractive index with concentration. A Pulfrich-type refractometer with a nominal accuracy of 5×10^{-6} was used. More details about the apparatus and the experimental method can be found in Ref. [7].

The Soret coefficient of water and ethanol mixtures has recently been measured by Kolodner *et al.* [8] and by Zhang *et al.* [9]. The values reported by both authors are in agreement and show that for alcohol concentrations below 30% the Soret coefficient is negative. Using the data of Kolodner [8], we determined from Eqs. (6) the Rayleigh number corresponding to the steady state for the four concentrations considered in this work at the mean temperature of 25 °C. The obtained values are shown in Table I. As can be seen, these values are two orders of magnitude higher than the critical value for an isothermal fluid layer.

For each concentration we measured the time evolution of the separation for different values of the Grashof number corresponding to different values of ΔT . Figure 1 shows the results obtained for the mixture of 22% in ethanol. The dotted line indicates the theoretical value of the steady-state separation calculated from Eqs. (6) and (7) using for S_T and D the data of Kolodner *et al.* [8] and those reported by Irving [10] for viscosity. As may be seen, for $Gr = 45$, separation increases with time until a maximum value lower than that predicted by theory is reached; thereafter it decreases. For $Gr = 95$, it does reach the theoretical value but after a few minutes separation begins to decrease. A similar situation occurs for $Gr = 115$. Finally, increasing the Grashof number we find that at $Gr = 144$ steady state remains stable. Therefore, for the Rayleigh number corresponding to this mixture the approximate value of the critical Grashof number is Gr_c

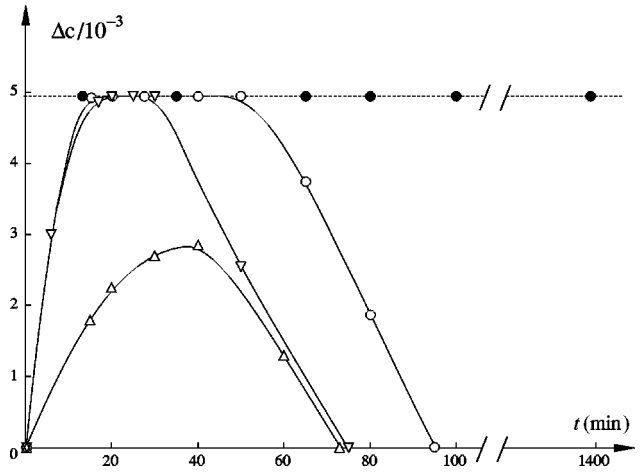


FIG. 1. Time evolution of separation for different Grashof numbers: Δ , $Gr = 45$; ∇ , $Gr = 95$; \circ , $Gr = 115$; \bullet , $Gr = 144$.

$= 144$. Below this value the thermogravitational steady state is unstable.

Similar experiments were conducted for the other concentrations shown in Table I. The critical Grashof numbers obtained are shown in this table, which also offers the corresponding critical values of ΔT . Figure 2 plots the neutral stability curve on the Ra - Gr diagram; the region of stability of the fluid layer is shaded. As it may be seen, the experimental points fit a straight line, indicating that the neutral curve is linear in the range of Gr values considered in this work.

From the above it may be concluded that convection favors the stability of the adverse density gradient originated by the thermogravitational effect. Thus, for sufficiently large values of the Grashof number it is possible to stabilize adverse gradients several orders of magnitude higher than the critical value corresponding to a quiescent layer.

From another point of view, this work shows that it is possible to measure negative Soret coefficients using the thermogravitational technique. This requires that the gap width of the thermogravitational cells and the temperature difference between the walls should be sufficiently large to be able to operate above the value of the critical Grashof number.

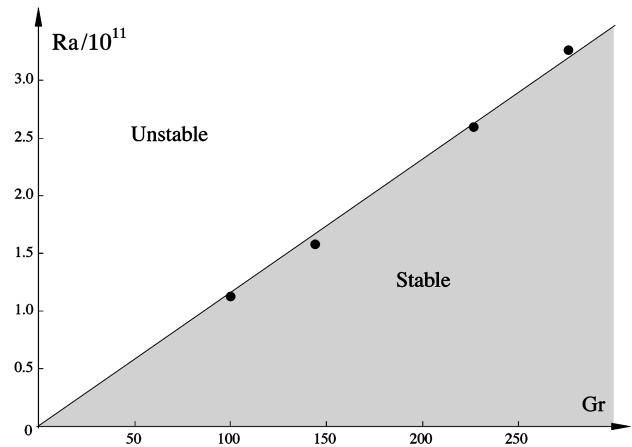


FIG. 2. Neutral stability diagram.

- [1] H. J. V. Tyrrell, *Diffusion and Heat Flow in Liquids* (Butterworths, London, 1961).
- [2] O. Ecenarro, J. A. Madariaga, J. Navarro, C. M. Santamaría, J. A. Carrión, and J. M. Savirón, *J. Phys.: Condens. Matter* **1**, 9741 (1989).
- [3] R. A. Wooding, *J. Fluid Mech.* **7**, 501 (1960).
- [4] J. N. Koster and U. Müller, *J. Fluid Mech.* **139**, 363 (1984); J. N. Koster, P. Ehrhard, and U. Müller, *Phys. Rev. Lett.* **56**, 1802 (1986).
- [5] C. Bizon, A. A. Predtechensky, J. Werne, K. Julien, W. D. McCormick, J. B. Swift, and H. L. Swinney, *Physica A* **239**, 204 (1997); A. A. Predtechensky, W. D. McCormick, J. B. Swift, Z. Noszticzius, and H. L. Swinney, *Phys. Rev. Lett.* **72**, 218 (1994).
- [6] O. Ecenarro, J. A. Madariaga, C. M. Santamaría, M. M. Bou-Ali, and J. Valencia, *Entropie* **198/199**, 71-76 (1996).
- [7] M. M. Bou-Ali, O. Ecenarro, J. A. Madariaga, C. M. Santamaría, and J. Valencia, *J. Phys.: Condens. Matter* **10**, 3321 (1998).
- [8] P. Kolodner, H. Williams, and C. Moe, *J. Chem. Phys.* **88**, 6512 (1988).
- [9] K. J. Zhang, M. E. Briggs, R. W. Gammon, and J. V. Sengers, *J. Chem. Phys.* **104**, 6881 (1996).
- [10] J. B. Irving, Glasgow National Engineering Laboratory Report No. 631, 1977.