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Classification

Physics Abstracts

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Critical wetting phenomena : observation of hydrodynamic instabilities

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Résumé. — Dans le cadre de phénomènes critiques de mouillage, on observe souvent qu'un liquide plus dense peut rester sur un liquide plus léger. Nous avons étudié ici les conséquences du mode d'instabilité de Rayleigh-Taylor dans la couche de mouillage semicritique d'un mélange binaire de liquides. Pour soutenir nos hypothèses, nous rapportons des observations expérimentales dans un mélange binaire de cyclohexane et acétonitrile.

Abstract. — In critical wetting phenomena, the situation of a heavier liquid residing on top of lighter liquid is observed. The consequences of the Rayleigh-Taylor mode of instability in the semicritical wetting layer in a binary liquid mixture are investigated. We also report experimental observations in support of our theory, in the critical binary liquid mixture cyclohexane + acetonitrile.

1. Introduction.

The interesting phenomenon of complete wetting of a semicritical interface by one of the critical phases in a binary liquid mixture has now been conclusively established [1-7]. The boundary between the liquids and the vapour phase above the liquids and the boundary of the container wall in contact with the liquids are the two non-critical interfaces, while the surface of separation between the two liquids forms the critical interface. Complete wetting, either of the vapour or of the wall of the container has been observed in systems like methanol + cyclohexane [2], 2,6 lutidine + water [3], perfluoromethyl cyclohexane + methylcyclohexane [4], cyclohexane + acetonitrile [5], isopropanol + perfluoromethyl cyclohexane [6] and a few other mixtures. The transition from complete wetting to partial wetting as the system is brought away from the critical temperature has also been observed. The two situations are schematically described in figure 1.

We have already reported the occurrence of complete wetting and the transition from complete wetting to partial wetting at temperatures away from the critical point, in the cyclohexane acetonitrile liquid mixtures [5]. This system has a very small density difference between the two constituent liquids and consequently the above phenomena are quite conspicuous and enable visual observations.

In the present paper we discuss the possibility of hydrodynamic instabilities in wetting phenomena.

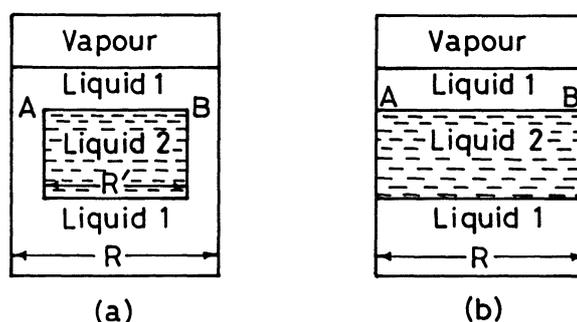


Fig. 1. — A schematic sketch of complete wetting and partial wetting. The figure (a) shows complete wetting situation and (b) shows the partial wetting situation.

In view of the fact that in the wetting interface a heavier liquid layer resides over a lighter one, the possibility of the Rayleigh-Taylor instability deserves consideration. Our observations concerning the effect of cell geometry on the stability of the wetting-layer can be qualitatively identified as the manifestations of the Rayleigh-Taylor instability. Some theoretical studies on the stability of fluid layers were performed by Maxwell [8] and some experimental work was carried on earlier by Duprez.

The criteria for the occurrence of the above instability vis-à-vis the dimensions of the containing vessel are theoretically discussed in the section 2 of the paper. Section 3 gives an account of the observations regarding the stability of the wetting layer with refe-

rence to the dimensions of the container. Since our present system shows a density inversion at about 42 °C, diverse meniscus shapes are observed as the system is progressively cooled below the critical temperature [5]. As is well known [2] cyclohexane + methanol is another system having very low density difference between its constituents but exhibits no density inversion in the temperature range of our interest. For comparison, the meniscus shapes in this system (for the critical concentration) are also reported. The concluding section of the paper is devoted to a qualitative comparison between the predictions of the theory and our experiments.

2. Theory.

2.1 STABILITY OF FLUID SURFACES IN PRESENCE OF CAPILLARY-GRAVITY WAVES. — The stability condition for interfaces between fluids, in presence of capillary-gravity waves is discussed on the basis of the familiar hydrodynamic equations of the fluid [9-11]. In the following we adopt the method given in reference [10, 11] and repeat their calculations in the case of a system in which the upper liquid is heavier than the lower one. As a matter of simplification, we shall consider the interface to be flat and the layer to be of uniform thickness. When the interface is given infinitesimal perturbations, the equations of fluid motion are [10, 11]

$$\frac{-\gamma(\eta_{xx} + \eta_{yy})}{(1 + \eta_x^2 + \eta_y^2)^{3/2}} + g(\rho_U - \rho_L) \eta - \Phi_t + \frac{1}{2} \bar{\rho}(\Phi_x^2 + \Phi_y^2 + \Phi_z^2) = \text{constant} \dots \quad (1)$$

$$\eta_t + \Phi_z = \Phi_x \eta_x + \Phi_y \eta_y \dots \quad (2)$$

$$\Phi_{xx} + \Phi_{yy} + \Phi_{zz} = 0 \dots \quad (3)$$

where γ is the surface-tension of the interface, ρ_U and ρ_L denote the densities of the upper and lower liquids respectively, $\bar{\rho}$ the mean density of the two liquids (assumed to be of nearly equal density), $\eta(x, t)$ denotes the vertical displacement of the interface along the z -direction, where the mean position is considered to be $z = 0$, with Φ representing the velocity potential of the fluid for a three-dimensional non-viscous, incompressible and irrotational flow. The $+z$ being vertically upwards, g has a value -9.8 m/s^2 .

In the following we consider a system with circular geometry, symmetric about the vertical z -axis. The solution of equation (3) in such a case, has a form

$$\Phi(r, t) = \sum_n A_n J_n(kr) (C_n \cos n\phi + D_n \sin n\phi) \times \frac{\cosh k(l-z)}{\sinh kl} e^{i\omega t} \dots \quad (4)$$

where the layer is bounded between $z = 0$ and $z = l$.

From the boundary condition that the radial flow must vanish at the walls of the vessel i.e. at $r = R$ we get,

$$J'_n(kR) = 0 \quad (5)$$

which determines the eigen values k for the system.

The solution of equations (1-4) is sought perturbatively by expanding [10]

$$\begin{aligned} \eta &= \eta_0 + \varepsilon \eta_1 + \varepsilon^2 \eta_2 + \dots \\ \Phi &= \Phi_0 + \varepsilon \Phi_1 + \varepsilon^2 \Phi_2 + \dots \\ \omega &= \omega_0 + \varepsilon \omega_1 + \varepsilon^2 \omega_2 + \dots \end{aligned} \quad (6)$$

which yield in the linearized limit,

$$\omega_0^2 = \frac{g(\rho_U - \rho_L) k + \gamma k^3}{\bar{\rho}} \tanh(kl) \quad (7)$$

When $\rho_U < \rho_L$, the system is stable (recall that g is -9.8 m/s^2) and this corresponds to stable ripples on the interface.

We consider now $\rho_U > \rho_L$. It is thus clear from equation (7) that the system can be unstable for $\omega_0^2 < 0$, which corresponds to

$$k^2 < \frac{(-g)(\rho_U - \rho_L)}{\gamma} \quad (8)$$

Hence if k_m be the minimum eigenvalue of (5), no instability can appear in any wave-vector provided k_m satisfies the inequality

$$k_m^2 > \frac{(-g)(\rho_U - \rho_L)}{\gamma} \quad (9)$$

Taking into account the non-linear effects it is seen that multiple hydrodynamic instabilities can appear at wave-vectors k_n , given by,

$$\begin{aligned} k_n^2 [g(\rho_U - \rho_L) + n^2 k_n^2 \gamma] \sinh(nk_n l) &= \\ = nk_n [g(\rho_U - \rho_L) k_n + \gamma k_n^3] \tanh(k_n l) \end{aligned} \quad (10)$$

where $n = 2, 3, 4 \dots$

Algebraic simplifications show that equation (10) does not admit solutions in the range $k_n^2 > (-g)(\rho_U - \rho_L)/\gamma$; possible solutions, if any, lie in the range $k_n^2 < (-g)(\rho_U - \rho_L)/\gamma$. It is thus clear that if the condition $k_m^2 > (-g)(\rho_U - \rho_L)/\gamma$ (11) be satisfied non-linearities cannot produce instabilities in any of the permissible wave vectors of the system. Thus condition (9) guarantees hydrodynamic stability of the system even when non-linearities are included. Thus, in our discussion of the stability of the wetting layer, it suffices if we consider condition (9) alone. In our following calculations, we shall discuss the results in the linearized limit.

Violation of (9) creates an instability in the system, due to which the displacement $\eta(x, t)$ grows exponentially with time, which may disrupt the flat configu-

ration of the liquid layer [12, 13]. These ideas have been applied by one of us (E. S. R. Gopal) in reference [14] to describe the dripping of liquids from the surfaces of metals. Although the stability criteria are derived in the inviscid case, inclusion of viscosity does not affect the stability condition for the surface waves [9-11]. However, it is to be noted that the stability analysis is valid for small displacements of the initially plane interface. In the following we examine similar possibilities for liquid-liquid interfaces close to the critical point.

2.2 RATE OF GROWTH OF INSTABILITIES. — For $k^2 < (-g)(\rho_U - \rho_L)/\gamma$ one finds

$$\omega = \pm i/\tau(k) \tag{12}$$

where

$$\frac{1}{\tau(k)} = \left| \frac{(-g)(\rho_U - \rho_L)k + \gamma k^3}{\bar{\rho}} \tanh kl \right| \tag{13}$$

or the disturbance $\eta(k, t)$ grows as,

$$\eta(k, t) \sim \eta(k, 0) \exp(t/\tau(k)) \tag{14}$$

The fastest rate of growth is observed in the case of thin layers (i.e. $kl \rightarrow 0$) for the wave-vector

$$k^2 = \frac{-g(\rho_U - \rho_L)}{2\gamma} \equiv k_0^2 \tag{15}$$

(the more commonly quoted result $k_0^2 = -g(\rho_U - \rho_L)/3\gamma$ applies in the thick layer limit).

Solution of equation (5) show

$$k_m \sim 2/R \tag{16}$$

so that the Rayleigh-Taylor instability is absent if

$$R^2 < \frac{4\gamma}{(-g)(\rho_U - \rho_L)} = 2a^2 \tag{17}$$

where $a = [2\gamma/(-g)(\rho_U - \rho_L)]^{1/2}$ is called the capillarity parameter. For a binary liquid mixture close to the critical point, with $T < T_c$, we know, $\gamma \sim \gamma_0 t^\mu$ and $(\rho_U - \rho_L) \sim |\widetilde{\Delta\rho}| t^\beta$ (where $t = |(T - T_c)/T_c|$) so that for a stable wetting layer to be present, the inequality,

$$R^2 < \frac{4\gamma_0}{(-g)|\widetilde{\Delta\rho}|} t^{\mu-\beta} \tag{18}$$

must be satisfied. From the stability condition in equation (18) it is clear that there cannot be a stable wetting layer very close to T_c . This is because γ goes to zero faster than $|\Delta\rho|$ and close to $T_c - g|\Delta\rho|/\gamma$ becomes very large and violates condition (9).

From the known values $\mu \sim 1.33$, $\beta \sim 0.33$, it is seen from (13) that for a vessel of any arbitrary radius the Rayleigh-Taylor instability is suppressed when a value of t satisfying (18) is reached. Similarly, with reference to equation (13) we conclude that if condi-

tion (18) is not maintained the instability grows at a faster rate in a vessel with smaller k_m i.e. with higher R .

In obtaining (18) we have used a scaled equation for γ as suggested by Van der Waals [15], Cahn and Hilliard [16]. The scaled behaviour is expected to breakdown beyond the critical region i.e. for t large. However, in the large t region, mean-field theory being valid, we get from the Cahn-Hilliard prediction [15], $\gamma \sim (\Delta C)^2/\chi_T^{1/2}$ where ΔC is the concentration difference between the two coexisting phases and χ_T the osmotic compressibility. With $(\rho_U - \rho_L) \propto \Delta C$ and χ_T decreasing with increasing t we find that the capillarity parameter must increase with increasing t . It thus ensures that at a certain value of t , for which the capillarity parameter exceeds the radius of the vessel, the system attains stability against the growth of the surface waves.

Furthermore, we note that though the above conclusions are drawn for the case of partial wetting they are equally valid for the complete wetting situation. In the latter, represented in figure 1, the stability condition for the stability of the AB interface against the Rayleigh-Taylor instability is similar to that given in (18) where R on the L.H.S. is to be replaced by R' .

In summary, the stability of the wetting layer is expected to be drastically affected by the horizontal dimensions of the container, such that the stable layer is observed only if $R^2 < 4\gamma/(-g)(\rho_U - \rho_L)$. This suggests from equation (5) that for a cell of a given diameter, stability against the Rayleigh-Taylor perturbations is ensured as we progressively lower the temperature below T_c . Further, the stability condition for a smaller cell is satisfied at a smaller value of t , as compared, with a cell of wider diameter.

Qualitative support for these ideas is sought experimentally. It should be noted, however, that our above analysis is true for a plane interface. For an unstable interface instabilities at wave vectors close to k_0 give rise to large displacements distorting the interface to a non-planar configuration. Complications in mathematical treatment do not allow analytical calculations to be presented in this case. However, it is to be emphasized that the instability towards the formation of non-planar interface is an evidence of the growth of the Rayleigh-Taylor instability.

3. Observations.

The observations include the study of the effect of cell geometry on the stability of the wetting layer and the study of the meniscus shapes as different compositions of the liquid are filled into identical cells.

3.1 EFFECT OF CELL GEOMETRY. — Purified samples of the liquids were filled at the critical composition in five cells of different dimensions. First, with the height of the liquid column fixed at 1.5 cm liquids were filled in cylindrical cells of diameter 2 cm, 4.5 cm, 4.9 cm, 6.4 cm and 7.4 cm. They were mounted adjacent to each other on the same stand in a thermo-

statically controlled bath, with a millidegree control. Due to the limitation in the size of the thermostat, only two cells were mounted at a time for the observations. The T_c of the cells agreed within the errors permissible.

The liquids were taken to the one-phase region by heating above the critical temperature and were cooled in steps of a few mK. A few mK below the phase separation temperature, a droplet of the heavier liquid was observed at the liquid vapour interface. We observed the formation of one single droplet in the smallest cell whereas multiple droplets could be seen at the liquid-vapour interface of the larger cells (Fig. 2). This is an evidence of the instability which does not allow a single wetting layer to be stable in a cell of larger diameter.

After the appearance of the droplets the temperature was held fixed and the stability of the droplets was observed as a function of time. In all such cases it was observed that the droplet in the larger cell vanished faster than the corresponding one in the smaller cell. As an example we quote that at 200 mK below T_c the layer in the larger cell was stable for three hours while that in the smaller cell was stable for more than five hours after which the observations were discontinued. The same behaviour was observed at all temperatures upto a temperature 1 K below T_c . We found the layers to be more stable as the system was cooled progressively, e.g. at 1 K below T_c the layer in the larger cell was found to vanish after seventeen hours while that in the smaller cell continued to be stable for more than twenty-four hours. In view of our prediction that far away from T_c the capillarity parameter is large enough to permit stability in both cells, we cooled the system 5 degrees below T_c . At this temperature the layers in both cells were found to be stable for over one hundred hours. These observations are thus in close conformity with the predictions of Rayleigh-Taylor instability.

The densities of the two phases can be approximately calculated from the study of the coexistence curve if the densities of the pure individual components be known. We have used the formula

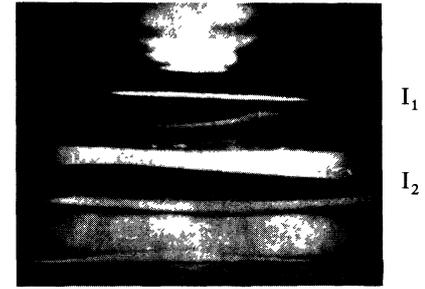
$$\rho_1(t) = v_a(t) \rho_a(t) + (1 - v_a) \rho_b(t)$$

$$\rho_2(t) = v'_a(t) \rho_a(t) + (1 - v'_a) \rho_b(t)$$

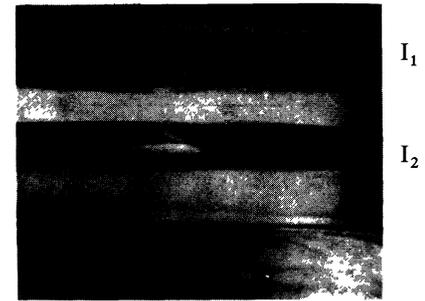
to obtain the densities of the two phases, where $v_a(t)$ and $v'_a(t)$ are the volume fractions of component a in the two coexisting phases and $\rho_a(t)$ and $\rho_b(t)$ denote the densities of the pure individual components. We have measured the densities at room temperature and have used the standard formula [17].

$$\rho(T) = \rho(T_s) + 10^{-3} \alpha(T - T_s) + 10^{-6} \beta(T - T_s)^2 + 10^{-9} \gamma(T - T_s)^3$$

to obtain the densities of the pure components at



(a)



(b)



(c)

Fig. 2. — Three photographs of the cell with a large diameter where the liquid vapour interface has multiple droplets. I_1 indicates the liquid-vapour interface and I_2 indicates the liquid-liquid interface. (a) Many small droplets can be seen at I_1 . (b) Four droplets are seen at I_1 . Another droplet has just dropped down and it can be seen at I_2 . (c) Two droplets can be seen at I_1 . All three photographs were taken at different temperatures as the system was cooled in steps of a few mK from the critical temperature.

other temperatures where the parameters

$$\alpha_{cy} = -0.8879 \quad \beta_{cy} = -0.972 \quad \gamma_{cy} = 1.55$$

$$\alpha_{ace} = -1.055 \quad \beta_{ace} = -0.138 \quad \gamma_{ace} = -6.00$$

are chosen from those given in the standard literature.

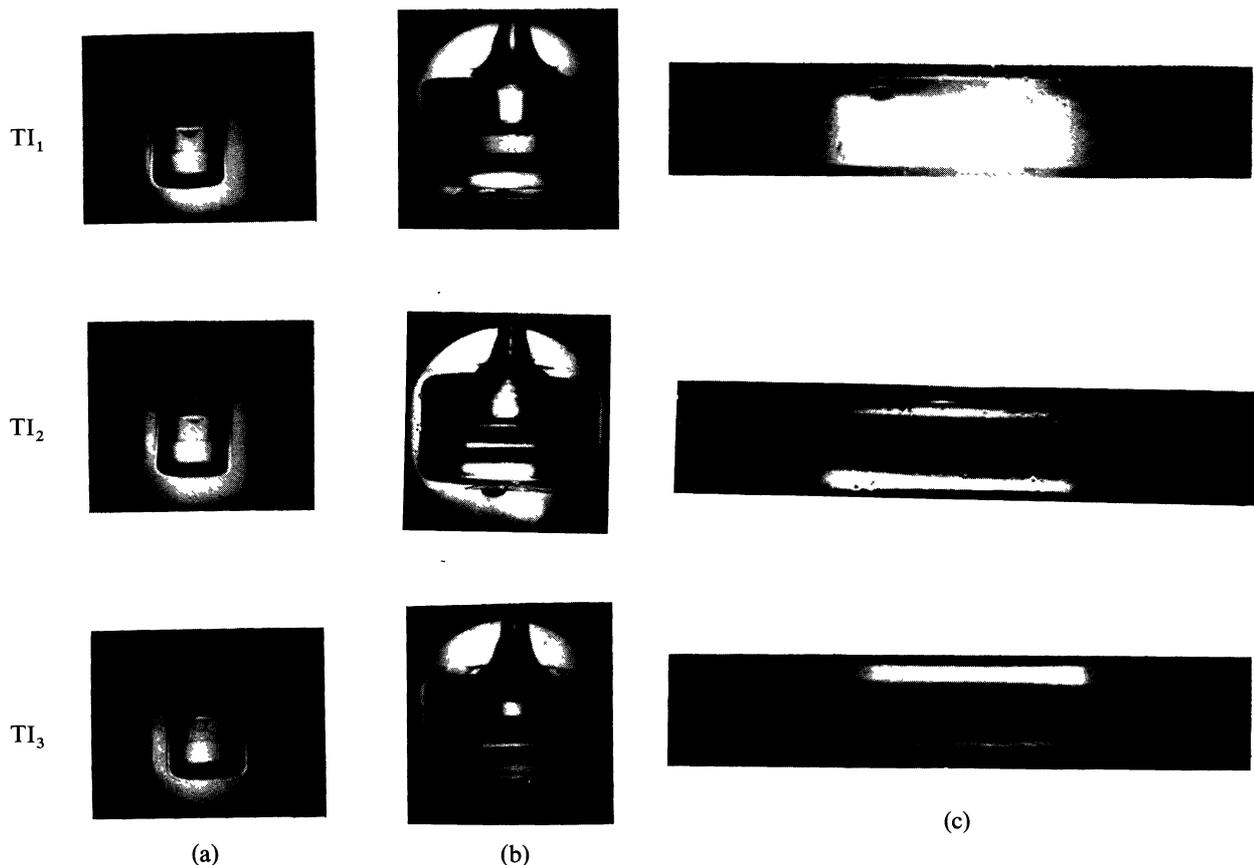


Fig. 3. — The photographs show the presence or absence of a droplet at the l-v interface of both the small and large cells. The photographs were taken at a temperature 1°C from T_c . TI_1 indicates the time when the system settled down after a thorough agitation. TI_2 indicates the time 12 h after the first photograph. TI_3 indicates the time 20 h after TI_1 . Column (a) shows three photographs of the smaller cell at TI_1 , TI_2 and TI_3 . Column (b) shows the larger cell at TI_1 , TI_2 and TI_3 . Column (c) gives an enlarged version of the l-v interface in the larger cell at TI_1 , TI_2 , and TI_3 . A droplet can be seen at the liquid-vapour interface of the smaller cell even at TI_3 , but the larger cell has no droplet at TI_3 . This shows that the droplet is more unstable in the larger cell than in the smaller cell. The smaller cell had the droplet even 24 h after TI_1 .

The calculated values of $\tau(k_m)$ at $t \sim 10^{-5}$ with ~ 0.01 cm are 10 and 3.5 s respectively for the cell diameter 2 cm and 7.4 cm cells. In our observations, the wetting layers were observed to be stable for several hours. This wide deviation between the two is attributed to the part that the wetting layers have non-planar shapes before dripping. This corresponds to a layer of varying l , while $\tau(k)$ has been calculated for constant l . Further at these conditions (i.e. large displacements of the planar interface) strong non-linear effects come into play and the rates of growth cannot be estimated exactly.

The time scales involved in these observations are from 30 min to 100 h, where even a visual observation is sufficient to decide whether the droplet vanishes first in the smaller or in the larger cell.

In the next part of our observation, we filled the system in cells of constant diameter 2 cm while the height of the liquid column was varied between 0.8 and 6.5 cm. The change in the shape of the meniscus in the different cells, was observed as the temperature

was lowered. The same qualitative trend as reported earlier [5] was observed in all the cells. The different meniscus shapes observed as the cell height was varied are given in the photograph (Fig. 4).

3.2 STUDY OF MENISCUS SHAPES WITH TEMPERATURE AND COMPOSITION. — The different shapes of the meniscus and the wetting behaviour was also observed as a function of the composition of the liquids. These can be seen in the photograph (Fig. 5). The peculiar meniscus shapes are due to the fact that there is a density inversion between the two liquids in the temperature range of interest. For comparison, we also observed the change in the meniscus shape of the binary liquid mixture cyclohexane + methanol. This system also has a very low density difference between the two liquids but do not exhibit density inversion in the temperature range of our study. This system also shows wetting property. There is no notable change in the meniscus shape as the temperature is lowered. The plot of the density vs. temperature for our liquid mixture can be seen in figure 6.

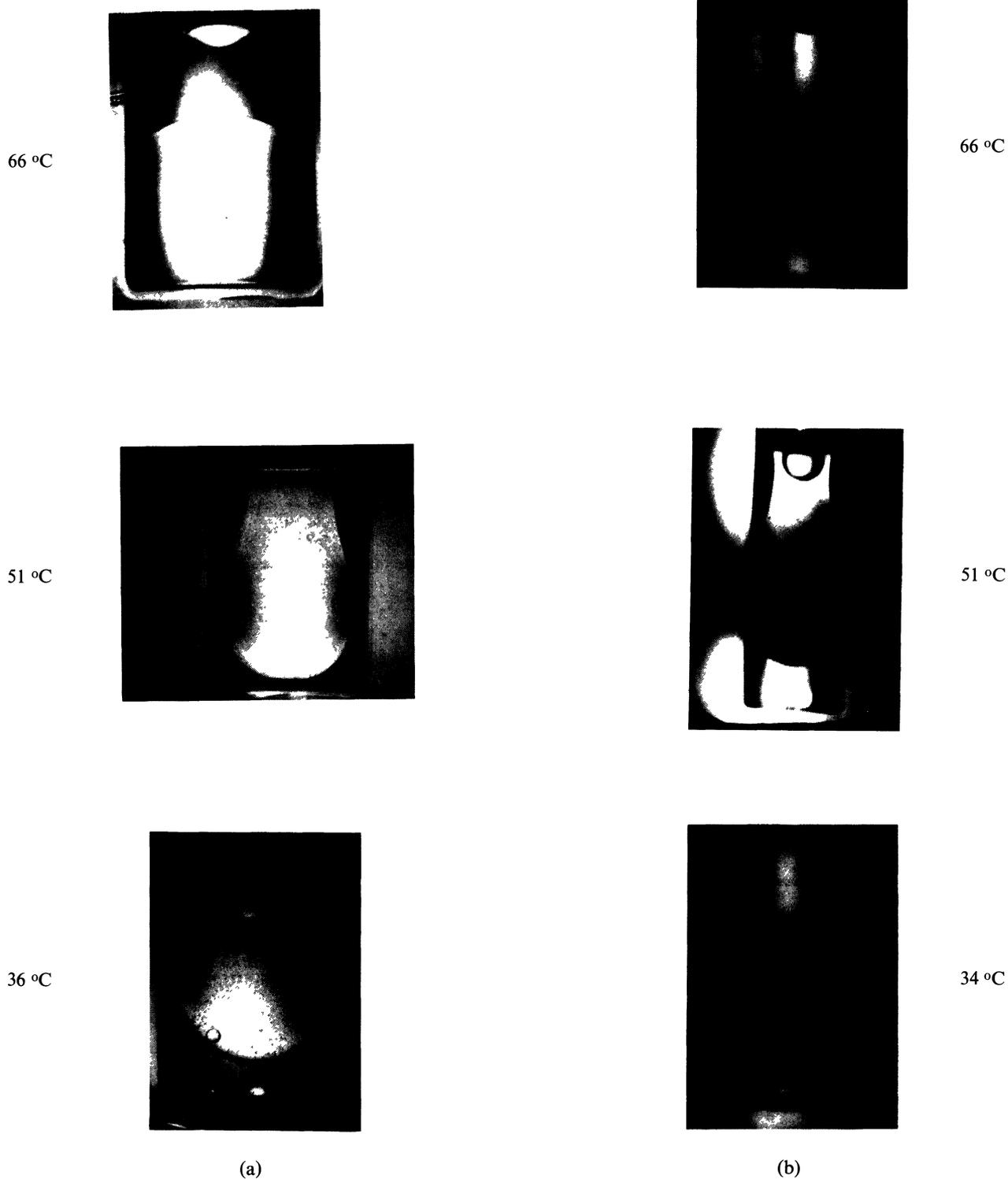


Fig. 4. — Photographs of the meniscus shape when the height of the cell is varied keeping a constant diameter. Figure (a) shows the change of the meniscus shape in a cell of length to diameter ratio 1.435. Figure (b) shows the change in the meniscus shape for a cell have length/diameter = 2.990. No qualitative change is observed in the meniscus shape when the cell height is varied.

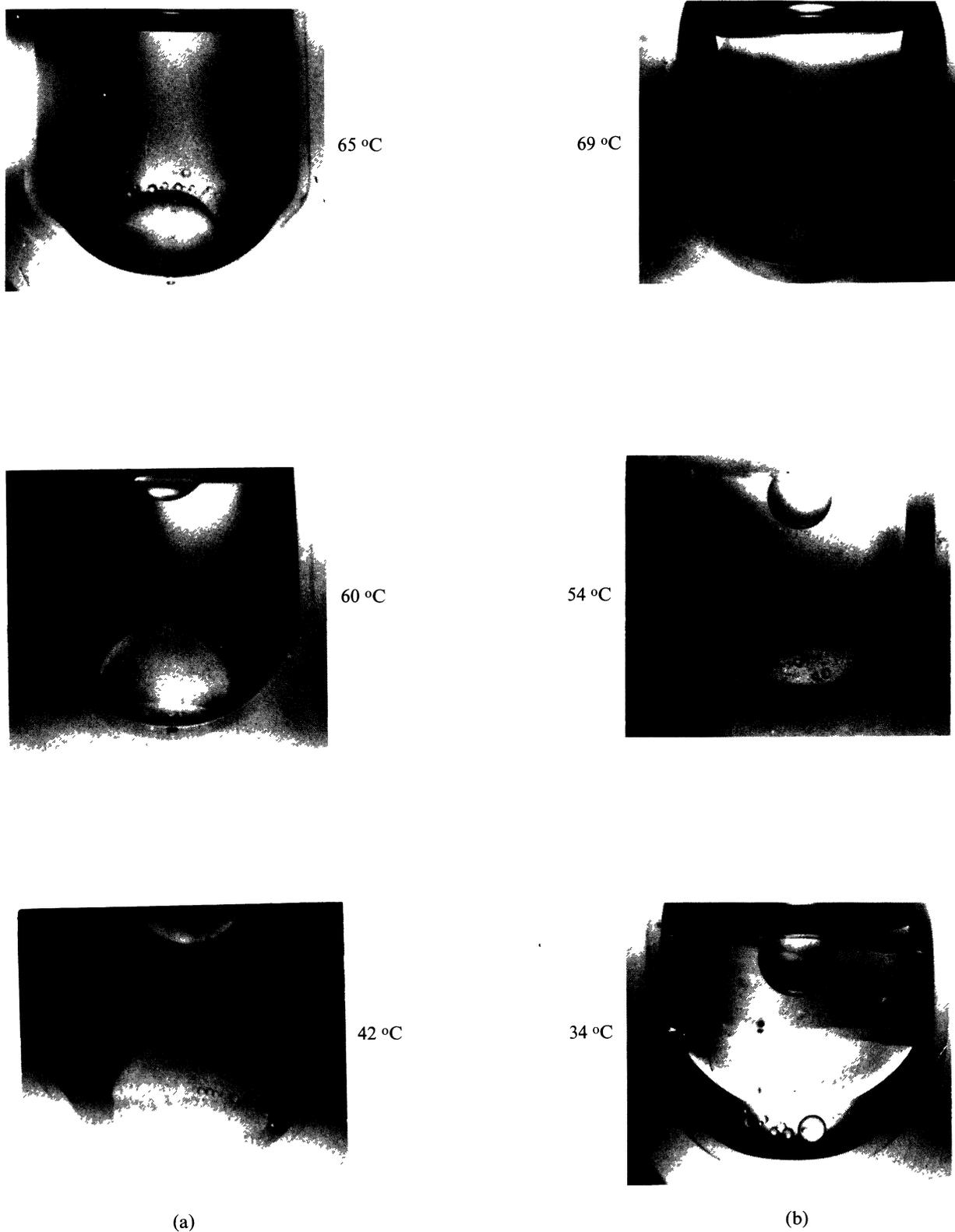


Fig. 5. — Photographs of the meniscus shape when the composition of the liquids are varied. (a) shows the meniscus shapes at different temperature for a composition 0.221 mole fraction of C_6H_{12} ; (b) shows the meniscus shapes at different temperature for a composition 0.650 mole fractions of C_6H_{12} .

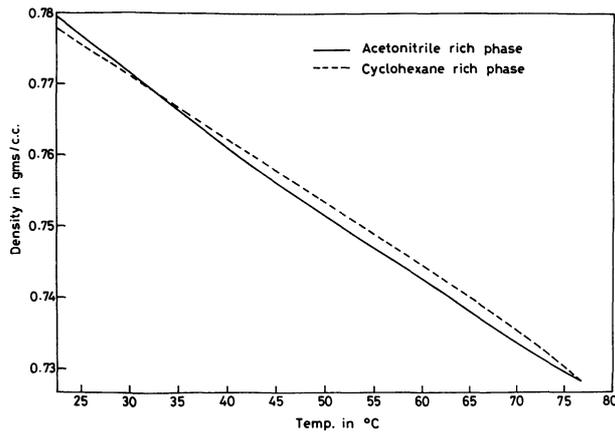


Fig. 6. — Plot of density vs. temperature of the cyclohexane and acetonitrile rich phases. The densities are very closely matched and there is a density inversion as a function of temperature.

4. Discussion.

In the present work we have investigated the appearance of the Rayleigh-Taylor instability in the wetting layer of a critical binary liquid mixture. Our studies show that the stability of the wetting layer depends very crucially on the diameter of the containing vessel as also on the value of t . For example, 1 mK below T_c the value of $[(-g)(\rho_U - \rho_L)/\gamma]^{1/2} = 67.5 \text{ cm}^{-1}$ which corresponds to a cell radius of 0.0304 cm. For a cell of radius larger than this value, we cannot have a stable wetting layer. The stability condition can be found graphically represented in figure 7. Figure 7 shows a semi-log plot of $[(-g)(\rho_U - \rho_L)/\gamma_0 t^\mu]^{1/2}$ vs. t . If any wave vector k_m falls below the curve then the wetting layer is unstable. For known cell diameter the temperature at which a stable layer becomes unstable, is also indicated in the figure. For example, in a cell of radius 0.5 cm, the wetting layer is unstable above 76.5°C. The maximum wave vector is calculated from equation (17).

At a given temperature, the instability is found to grow at a faster rate in a cell of larger diameter. In addition, the wetting layer is found to be stable in the cells when the temperature is sufficiently away from the critical point. These qualitative observations show remarkable consistency with the predictions based on Rayleigh-Taylor instability.

As a matter of detail, we observe that the vibration

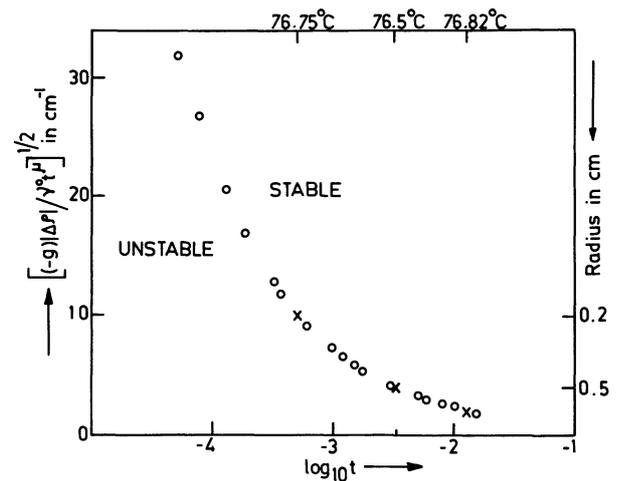


Fig. 7. — Plot of $\log t$ vs. $[(-g)|\Delta\rho|/\gamma_0 t^\mu]^{1/2}$ indicates the boundary between stable and unstable region. The cross mark « x » shows the temperature at which the stable layer becomes unstable for particular cell diameter which is shown at the right side of the graph.

spectrum of the surface oscillations should be calculated by taking into account the shape of the meniscus, whereas we have used the approximation that the interface is flat. This consideration should also incorporate the oscillations of the angle of contact [18]. However, we note that though these factors are necessary to obtain the details, these should not significantly affect the condition of stability. Moreover, the calculation of the meniscus shape is itself a complicated calculation [19].

To derive further justification of our theory, we have undertaken the measurement of the capillarity parameter a of the system at different temperatures. With the help of these data we wish to obtain a quantitative verification of our ideas, the other alternative being to conduct experiments under microgravity conditions e.g. in space. In conclusion, we note that there is qualitative evidence that the gravity-capillary waves affect the stability of the wetting layer. Their effect on the equilibrium value of the wetting layer thickness will be reported in a subsequent work.

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