WETTING-LAYER EFFECTS ON HELE-SHAW FLOW

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Chan and Liang noticed a new instability phenomenon related with the displacing process in a vertical Hele-Shaw cell. When the inner part of the Hele-Shaw plates are coated with a thin surfactant layer and a more viscous fluid is displacing a less viscous one, the interface between the fluids becomes unstable if the surface velocity exceeds a critical value. This is in contradiction with the Saffman-Taylor criterion observed in the clean case (no surfactant on the plates). We show that the variation and the magnitude of the surface tension are the main causes of this phenomenon, by using a simplified form of the boundary conditions on the interface. This new kind of instability appears also for displacements in horizontal Hele-Shaw cells with a preexisting layer of surfactant on the inner part. For this, we use a model with slip conditions on the plates.

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1. INTRODUCTION

One of the simplest models for the flow in a porous medium is the Hele-Shaw approximation. A Stokes liquid flows between two parallel plates at a very small distance, compared with the plates length. The flow is produced by a given velocity far upstream or by a pressure gradient. The average (across the plates) of the Poiseuillle solution gives us an equation similar with the Darcy's law for the flow in a porous medium – see [11]. The viscosity of the filtration fluid is the real viscosity of the initial liquid.

An important problem related with the Hele-Shaw model is the flow of two immiscible fluids which are displacing each other. In a real porous medium, the interface between the immiscible fluids is very difficult to describe and in fact it is not possible to get an effective equation. The simplest assumption is to consider that the displacing fluids remain completely separated along a definite interface. This is the main assumption considered in the Hele-Shaw model: a "sharp" interface exists between the two immiscible fluids and in every point of the equivalent porous medium we have only one fluid. An important application of the Hele-Shaw model is related with the secondary recovery process: the oil from a porous medium is displaced by using a second (forerunner) viscous liquid (usually water) – see [1]. The first results concerning the stability of the immiscible displacement in Hele-Shaw cells are the papers [4] and [14]. Here, a liquid of viscosity μ_1 is displacing a liquid of viscosity μ_2. The gravity is neglected for the horizontal Hele-Shaw cells. In the two above cited papers, a basic steady solution with a straight interface is pointed out. The linear stability analysis of this basic solution is performed. The Saffman-Taylor criterion gives a stable interface when the displacing fluid μ_1 is more viscous and instability when $\mu_1 < \mu_2$. In this last case, the fingering phenomenon appears. In the case of water displacing oil, fingers of water appear in oil and a "trapped" quantity of oil is lost for recovery.

In [2] was noticed a new type of instability phenomenon, in the case of a vertical Hele-Shaw cell filled by air, with a very thin surfactant layer on the plates. The cell is dipped into a pool filled by the same surfactant. Then the interface air-surfactant becomes unstable if the displacing velocity exceeds a critical value. This is in contradiction with the Saffman-Taylor criterion (air is less viscous). An attempt to explain this new phenomenon was given in [2]: a new state equation of the surface tension on the interface was considered, in terms of the deformation of the interface and of the diffusion of surfactant molecules from the bulk on the interface. An important assumption followied the advection of the surfactant from the wetting-layer on the interface between displacing fluids is proportional with the interface velocity. This assumption can be related with the experimental results of Sheng and Smith (1999).

The new instability for the flow in vertical Hele-Shaw cells was partially confirmed in [7] and [8]. In [7] is pointed out that "the displacing velocity is not appearing in the stability parameter of the flow" (beginning of Section 4.3). The variable surfactant concentration on the interface seems to be the principal mechanism producing the instability. A transition region between the constant-thickness region and the meniscus of the pool interface is considered. The flow in the transition region is governed by the 3D Stokes equations. A linear stability analysis of a basic solution in the transition region is performed, in the range of low capillary numbers. The new instability phenomenon is revealed as the "effect on the thickening of the wetting-layer caused by a significant Marangoni stress, which, in turn, originate in the accumulation of surfactants in the cap and transition regions" (see the Conclusions, page 121).

In this paper we perform a linear stability analysis of a basic state, for both vertical and horizontal Hele-Shaw cells with a wetting-layer on the inner part of the plates.

In the case of a vertical cell, we use a simplified form of the boundary conditions on the interface and get a simpler explanation for the new-instability reported in [2]. We prove in a simple way that the principal element producing the new-instability is the variation of the surfactant concentration along the wetting-layer surface. The variation of the surfactant concentration gives (determines) the variation of the surface tension σ. However, we show that the magnitude of σ is also an important element producing the wetting-layer instability – see the last part of Section 2. For this, we perform an analysis of the boundary conditions verified by the perturbations $\overline{v}'$, $\overline{p}'$, $\overline{h}'$,$\overline{\sigma}'$, $\overline{\gamma}'$ of the basic state on the free surface (see *Remark* 1). Our original result follows the instability can appear for an *almost constant* σ, if the magnitude of the surface tension is large enough. As we use a simplified form of the boundary conditions on the interface, our dispersion relations (32), (36) for the growth rate of perturbations are of first order. Moreover, (32) and (36) are related with the basic displacing velocity. The dispersion relation in [7] is of second order.

In the case of a horizontal Hele-Shaw cell, we consider a less viscous displacing liquid μ_1. We use the lubrication approximation for a Stokes flow in a Hele-Shaw cell, but with a slip condition on the surfactant-layer surface. An average procedure gives us a Darcy-type equation with a new term – see (46). A linear stability analysis is performed, similar with the Saffman-Taylor approach. The dispersion relation (53) also contains a new term. The surfactant layer can be obtained by the coating process studied in [16]. Therefore the thickness (55) is given in terms of μ (the viscosity of surfactant) and U (the coating velocity). The viscosities μ_1, μ_2 of the immiscible liquids are considered larger than the surfactant viscosity μ. Then we can use the boundary conditions given in [12] and [13]. We use a discretization of the normal derivatives of the velocity and the thickness (53), in the range of low capillary numbers. In this way, we get the new term in the dispersion relation (53) as function of the Marangoni stress on the surfactant layer – see the relation (64). This new term allows us to explain a new stability phenomenon: the interface between the liquids μ_1, μ_2 is stable if the interface velocity V is less than a critical value (67), even if the displacing liquid is less viscous. However, the surface tensions of the displacing liquids on the wetting-layer must verify the relationship given in *Remark* 6.

The outline of the paper is the following. In Section 2, we describe the mathematic model and perform the stability analysis in the case of vertical Hele-Shaw cell. The horizontal case is studied in Section 3. We conclude in Section 4.

2. THE VERTICAL HELE-SHAW CELL

The experiment noticed in [2] can be described as follows. A Hele-Shaw cell is dipped into a pool with a surfactant. Therefore we can consider that the surfactant from the pool is displacing the air between the Hele-Shaw plates. The interface air-surfactant is stable, according with the Saffman-Taylor criterion (the surfactant is more viscous). The cell is pulled out from the pool, and a thin layer of surfactant is adhering on the plates, according with the Landau-Levich theory – see [12]. When the "contaminated" Hele-Shaw cell is dipped again in the pool, the interface air-surfactant becomes unstable, even if the surfactant is more viscous, in contradiction with the Saffman-Taylor criterion.

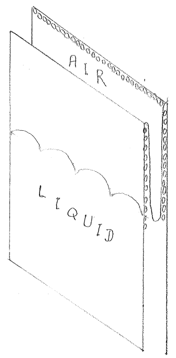

Fig. 1 – Vertical Hele-Shaw cell, with surfactant-adherent layer on inner walls, dipped in a surfactant pool.

We consider a "half" of the Hele-Shaw cell - more precisely we consider a plate contained in the plane xOz, where the axis Ox is pointed down, in the gravity direction; the axis Oy is orthogonal on the plate. The symmetry axis of the cell is $y = 0$. The plate is moving down with the velocity V, into the pool with surfactant. Far up on the plate, there exists a thin layer of the same surfactant, with a constant thickness. A transition region is matching this thin layer with the meniscus on the pool surface. The flow in the transition region is described by the 3D Stokes equations.

We study the case when the wetting-layer (on the plates) was formed by pulling out the plate from the pool, then a variable quantity of surfactant exists on the layer surface. Therefore the surfactant concentration on the layer surface is increasing (recall the Ox axis is down). As a consequence, the surface tension on the wetting-layer surface is not constant, but decreasing as function on the surfactant concentration.

The following notations are used: σ_0, Γ_0, h_0 are the surface tension, the surfactant concentration and the layer thickness far up on the plate; d, ρ, μ, g are the half gap length of the cell, the pool-liquid density and viscosity and the gravity acceleration.

The spatial coordinates are non-dimensionalized by d and the velocities by the dipping speed V. The surface tension and surfactant concentration are scaled by σ_0, Γ_0. The characteristic time is d/V and the pressure is scaled with σ_0/d.

The important dimensionless parameters of our analysis are the *Capillary, Reynolds* and *Bond* numbers given by

$$Ca = \mu V/\sigma_0, \quad Re = \rho V d/\mu, \quad Bo = d^2 \rho g/\sigma_0. \tag{1}$$

As we pointed before, the flow in the transition region (which relates the constant thin layer with the pool surface) is described by the Stokes equations (see also [7]):

$$\begin{aligned} &u_x + v_y + w_z = 0, \\ &ReCa \cdot u_t - Bo + p_x = Ca\Delta u, \\ &ReCa \cdot v_t + p_y = Ca\Delta v, \\ &ReCa \cdot w_t + p_z = Ca\Delta w, \end{aligned} \tag{2}$$

where $(u, v, w), p$ are the velocity components on the axis Ox, Oy, Oz and the pressure. The indices denote the partial derivatives with respect to the time t and with the spatial coordinates x, y, z. The free surface of the surfactant layer is $y =^+_- h(x, z, t)$. The solutions must verify the following boundary conditions (see [7] – page 109, 110, and [12] – page 384):

$$y = -1 \Rightarrow u = 1, \quad v = 0, \tag{3}$$

$$y =^+_- h(x, z, t) \Rightarrow h_t + uh_x + v + wh_z = 0, \tag{4}$$

$$y =^+_- h(x, z, t) \Rightarrow p - \sigma(h_{xx} + h_{zz}) = 2Ca\ v_y, \tag{5}$$

$$y =^+_- h(x, z, t) \Rightarrow \sigma_x = Ca\ u_y, \quad \sigma_z = Ca\ w_y, \tag{6}$$

where σ is the surface tension on the wetting-layer surface and the air pressure is considered equal to zero. The relations (5), (6) are simplified forms of the boundary conditions given in [7]: we neglect the second order terms appearing in [7].

In this paper, we do not consider the equations of the interfacial surfactant concentration, which are not necessary in our analysis for proving the instability effect due to the wetting-layer effect.

When the plate was pulled out from the liquid-pool with a vertical negative velocity V_d, the thickness of the wetting-layer adhering on the plate is given by the Landau-Levich theory (see [16] and references therein). Therefore we have

$$h_0 \approx Ca_d^{2/3}\sqrt{\sigma_0/\rho g}, \quad Ca_d = \mu V_d/\sigma_0. \tag{7}$$

Then some adimensionalizations of the previous equations and boundary conditions are necessary. However, instead of dealing with *two* capillary numbers, we consider (as in [7]) that the capillary numbers defined by (1) and (7) are almost the same, therefore we consider

$$Ca \approx Ca_d. \tag{8}$$

This assumption is based on the fact that all considered velocities are small enough. We follow [7, 12] and introduce the following non-dimensional quantities, denoted by *overline*

$$\bar{t} = \frac{t}{Ca^{1/3}}, \quad \overline{x} = \frac{x+l}{Ca^{1/3}}, \quad \overline{z} = \frac{z}{Ca^{1/3}}, \quad \overline{y} = \frac{y+1}{Ca^{2/3}}, \quad \overline{h} = \frac{1-h}{Ca^{2/3}} \tag{9}$$

$$\overline{p} = p, \quad \overline{v} = \frac{v}{Ca^{1/3}}, \quad \overline{u} = u, \quad \overline{w} = w, \quad \overline{\gamma} = \frac{\gamma}{Ca^{2/3}}, \tag{10}$$

where γ is the surfactant concentration on the wetting-layer surface. The location of the origin l is determined when we perform the matching procedure with the meniscus on the pool-surface. In the new dimensional quantities, the flow is governed by the following equations:

$$\begin{aligned}
&\overline{u}_{\overline{x}} + \overline{v}_{\overline{y}} + \overline{w}_{\overline{z}} = 0,\\
&ReCa \cdot \overline{u}_{\overline{t}} - Bo\,Ca^{1/3} + \overline{p}_{\overline{x}} = Ca^{2/3}(\overline{u}_{\overline{xx}} + \overline{u}_{\overline{zz}}) + \overline{u}_{\overline{yy}}\ ,\\
&ReCa^{5/3} \cdot \overline{v}_{\overline{t}} + \overline{p}_{\overline{y}} = Ca^{8/3}(\overline{v}_{\overline{xx}} + \overline{v}_{\overline{zz}}) + Ca^{2/3}\overline{v}_{\overline{yy}}\ ,\\
&ReCa \cdot \overline{w}_{\overline{t}} + \overline{p}_{\overline{z}} = Ca^{2/3}(\overline{w}_{\overline{xx}} + \overline{w}_{\overline{zz}}) + \overline{w}_{\overline{yy}}\ ,
\end{aligned} \tag{11}$$

and boundary conditions (3)–(6) on the interface (in the linear approximation) become

$$\overline{h}_{\overline{t}} + \overline{u}\overline{h}_{\overline{x}} - \overline{v} + \overline{w}\overline{h}_{\overline{z}} = 0, \tag{12}$$

$$\overline{p} + \sigma(\overline{h}_{\overline{xx}} + \overline{h}_{\overline{zz}}) = 2Ca^{2/3}\ \overline{v}_{\overline{y}}\ , \tag{13}$$

$$\sigma_{\overline{x}} = Ca^{2/3}\overline{u}_{\overline{y}}, \quad \sigma_{\overline{z}} = Ca^{2/3}\overline{w}_{\overline{y}}\ . \tag{14}$$

The relation (13) is obtained from the equation (5). The change of the sign in front of σ follows from the transformation $(9)_5$.

As in [7], we consider $Ca \approx 10^{-3}, Bo \approx 10^{-2}$ (for $d = 300\mu m$), then we can develop a low-capillary theory in the thin (adhering) film approximation. We assume also the condition $g(\overline{h}_\infty/\nu V) << 1$, where ν is the kinematic viscosity and $\overline{h}_\infty$ is the (constant) thickness of adhering film far-up on the plate. That means the gravity effects play a negligible role and $BoCa^{1/3}$ can be neglected.

We consider now the basic solution corresponding to $ReCa \approx 0$ and $\overline{w} = 0$, that means

$$\text{(15)} \qquad \begin{aligned} &\overline{u}_{\overline{x}} + \overline{v}_{\overline{y}} = 0, \\ &\overline{p}_{\overline{x}} = \overline{u}_{\overline{yy}} \ , \\ &\overline{p}_{\overline{y}} = 0, \\ &\overline{p}_{\overline{z}} = 0, \end{aligned}$$

subject to the boundary conditions (12)–(14).

Σ, Γ, H are denoting the basic surface tension, the basic surfactant concentration and the basic free surface. The basic pressure is not depending on $\overline{y}, \overline{z}$.

We consider now the perturbations of the basic pressure, velocity, free surface, surface tension and surfactant concentration, denoted by

$$\text{(16)} \qquad \overline{p}', \quad \overline{u}', \quad \overline{v}', \quad \overline{w}', \quad \overline{h}', \quad \sigma', \quad \overline{\gamma}'.$$

In the general case when the basic surfactant concentration Γ is not constant, the basic surface tension Σ is depending on Γ and we suppose

$$\text{(17)} \qquad \Sigma_{\overline{x}} = \Sigma_\Gamma \Gamma_{\overline{x}} \, , \quad \Sigma_{\overline{z}} = \Sigma_\Gamma \Gamma_{\overline{z}},$$

where $_\Gamma$ is denoting the partial derivative in terms of Γ. A linear approximation around the basic state yields

$$\text{(18)} \qquad \sigma'_{\overline{x}} = \Sigma_\Gamma \overline{\gamma}'_{\overline{x}} \, , \quad \sigma'_{\overline{x}} = \Sigma_\Gamma \overline{\gamma}'_{\overline{x}} \, .$$

Recall that $(11)_4$ for $ReCa \approx 0$ at the leading order in Ca is giving $\overline{p}_{\overline{z}} = \overline{w}_{\overline{yy}}$, then for the perturbations we get the equation $(19)_4$ below.

Therefore at the leading order in Ca, the relations (11)–(15) give us the following system which governs the perturbations:

$$\text{(19)} \qquad \begin{aligned} &\overline{u}'_{\overline{x}} + \overline{v}'_{\overline{y}} + \overline{w}'_{\overline{z}} = 0 \ , \\ &\overline{p}'_{\overline{x}} = \overline{u}'_{\overline{yy}} \ , \\ &\overline{p}'_{\overline{y}} = 0, \\ &\overline{p}'_{\overline{z}} = \overline{w}'_{\overline{yy}} \ . \end{aligned}$$

The boundary conditions (on the wetting-layer surface), linearized around the basic state, are:

$$\text{(20)} \qquad \overline{h}'_{\overline{t}} + \overline{u}\overline{h}'_{\overline{x}} - \overline{v}' = 0,$$

$$\text{(21)} \qquad \overline{p}' + \Sigma(\overline{h}'_{\overline{xx}} + \overline{h}'_{\overline{zz}}) = 2Ca^{2/3} \ \overline{v}'_{\overline{y}} \ ,$$

$$\text{(22)} \qquad \Sigma_\Gamma \overline{\gamma}'_{\overline{x}} = Ca^{2/3} \overline{u}'_{\overline{y}} \, , \quad \Sigma_\Gamma \overline{\gamma}'_{\overline{z}} = Ca^{2/3} \overline{w}'_{\overline{y}} \, .$$

The relations (19)–(22) are quite similar with the system studied in [7].

Remark 1. Here we perform a careful analysis of the relations (19), (21), (22) and obtain the important relation (26) below. For this, we use the equation $(19)_1$ and get

$$\overline{v}'_{\overline{yyy}} = -\overline{u}'_{\overline{xyy}} - \overline{w}'_{\overline{zyy}}.$$

The derivation with respect to $\overline{x}$ and $\overline{z}$ in $(19)_2$, $(19)_4$ gives us

$$\Delta_{\overline{x},\overline{z}}\overline{p}' = -\overline{v}'_{\overline{yyy}} \ . \tag{23}$$

From (19) we have $\overline{p}'_{\overline{y}} = 0$, then $\overline{p}'$ is not depending on $\overline{y}$. We use the approximation

$$\overline{p}'|_{(\overline{y}=H)} = 0,$$

where H is the basic free surface. Then the relation (21) can be used to obtain the following relation on the free surface.

$$(\Sigma\Delta_{\overline{x},\overline{z}}\overline{h}')|_{(\overline{y}=H)} = 2Ca^{2/3}\ \overline{v}'|_{(\overline{y}=H)} \tag{24}$$

The relations (22) hold only on the free surface. We use the free-divergence relation $(19)_1$, we perform the derivatives with respect to $\overline{x}, \overline{z}$, then from (22) it follows

$$\Sigma_\Gamma\Delta_{\overline{x},\overline{z}}\overline{\gamma}' = -Ca^{2/3}\ \overline{v}'_{\overline{yy}} \ . \tag{25}$$

We consider now the last three relations (23)–(25) near the basic free surface H and obtain

$$\overline{v}'|_{(\overline{y}=H)} = -\frac{H^3}{3}\Delta_{\overline{x},\overline{z}}\overline{p}' = \frac{H}{2Ca^{2/3}}\Sigma\Delta_{\overline{x},\overline{z}}\overline{h}' = -\frac{H^2}{2Ca^{2/3}}\Sigma_\Gamma\Delta_{\overline{x},\overline{z}}\overline{\gamma}' \ . \tag{26}$$

Remark 2. Recall the last relation (4.4) (see page 116) of [7]:

$$\overline{h}'_{\overline{t}} + \overline{h}'_{\overline{x}} = \frac{H^3}{3}\Delta_{\overline{x},\overline{z}}\overline{p}' - \frac{H^2}{2Ca^{2/3}}\Sigma_\Gamma\Delta_{\overline{x},\overline{z}}\overline{\gamma}' \ . \tag{27}$$

The authors specify that the term from the left hand side of (27) is an advection of the interface perturbation by the basic flow. However, in our paper this term is zero, due to relationship (26) and because we used the simplified boundary conditions (32), (36).

Taking into account *Remark* 2, we get a new stability analysis of the perturbed interface and show the two elements producing the new-instability: the *variation* Σ_Γ and the *magnitude* Σ of the basic surface tension on the wetting-layer surface. For this we recall the relations (20), (26) and get

$$\overline{h}'_{\overline{t}} = -\overline{u}\overline{h}'_{\overline{x}} + \frac{1}{2}\{\frac{H}{2Ca^{2/3}}\Sigma\Delta\overline{h}'\} - \frac{1}{2}\{\frac{H^2}{2Ca^{2/3}}\Sigma_\Gamma\Delta\overline{\gamma}'\}. \tag{28}$$

We consider two important cases.

1) In the case of an *almost constant* surface tension, that means

$$\Sigma_\Gamma \overline{\gamma}' \approx 0$$

we use the relations (28) on the free surface, and obtain

$$\overline{h}'_{\overline{t}} = -\overline{u}\overline{h}'_{\overline{x}} + \frac{H}{2Ca^{2/3}} \Sigma \Delta_{x,z} \overline{h}' \ . \tag{29}$$

This relation holds in the hypothesis

$$|\Sigma \Delta \overline{h}'| >> |H \Sigma_\Gamma \Delta \overline{\gamma}'|, \tag{30}$$

then we neglect the third term in the right part of (28). Therefore the variation of Σ is considered much smaller compared with its magnitude, but the laplacian of $\overline{h}'$ and $\overline{\gamma}'$ are of the *same* order - see below the relations (31) and (35).

As in [7], we consider the normal disturbance $\overline{h}'$ of the form

$$\overline{h}' \propto exp(\varepsilon x + \mathrm{i}\, kz + at), \tag{31}$$

where a is the growth constant. This kind of "expansion" is used also in [17], related with the stability of the displacement of an Oldroyd-B fluid by air in a Hele-Shaw cell. If we consider here an "amplitude", it will simplify, because the relationship (29) is linear.

Then the relation (29) gives us

$$a = -\overline{u}\varepsilon + \frac{H}{2Ca^{2/3}} \Sigma(\varepsilon^2 - k^2). \tag{32}$$

The *basic* velocity $\overline{u}$ is down, then positive. We consider $\varepsilon > 0$, because the perturbations must decay to zero far from the pool surface. The *first main conclusion* of this section is obtained from (32):

In the case of a very slow variation of the surface tension, the wetting-layer surface is unstable if the following condition holds

$$\frac{H}{2Ca^{2/3}} \Sigma(\varepsilon^2 - k^2) > \overline{u}\varepsilon. \tag{33}$$

The above condition is verified for small enough wavenumbers k and large enough basic surface tension Σ. Moreover, we have $a < 0$ for large wavenumbers.

2) In the case of a *large variation* of the surface tension (that means for relative large variation of the surfactant concentration), we use the relations (28) with two terms involving Σ. It is important to emphasize that the surface tension is decreasing in terms of the surfactant concentration, therefore $\Sigma_\Gamma = -M < 0$, where M can be considered as a Marangoni positive number and (28)

becomes

$$\overline{h}'_{\overline{t}} = -\overline{u}\overline{h}'_{\overline{x}} + \frac{1}{2}\{\frac{H}{2Ca^{2/3}}\Sigma\Delta\overline{h}'\} + \frac{1}{2}\{\frac{M\,H^2}{2Ca^{2/3}}\Delta\overline{\gamma}'\}, \tag{34}$$

Besides the normal mode decomposition (31) for $\overline{h}'$, we consider a similar decomposition for $\overline{\gamma}'$ (used also in [7]):

$$\overline{\gamma}' \propto exp(\varepsilon x + \mathrm{i}\, kz + at), \tag{35}$$

and from the last two relations we get

$$a = -\overline{u}\varepsilon + \frac{H}{4Ca^{2/3}}(\varepsilon^2 - k^2)(\Sigma + M\ H). \tag{36}$$

The last relation gives the *second main conclusion* of this section:

If the surface tension and its variation are of the same order on the wetting layer surface, then the displacing process is unstable if

$$\frac{H}{4Ca^{2/3}}(\varepsilon^2 - k^2)(\Sigma + M\ H) > \overline{u}\varepsilon. \tag{37}$$

Moreover, we have $a < 0$ *for large wavenumbers.*

We see that in this case a Fourier decomposition was necessary for both perturbations of the surface tension and surfactant concentration.

Remark 3. In both formulas (29)–(34) we can add in the right part the term $-\Delta_{\overline{x},\overline{z}}\overline{p}'$. If the perturbation $\overline{p}'$ is decomposed in Fourier modes (in a similar way to $\overline{h}'$, $\overline{\gamma}'$), we see that this third term has a destabilizing effect. Therefore the stability is governed by the competition between three elements: the basic velocity $\overline{u}$, the magnitude of Σ, M and the magnitude of momentum advection due to the perturbations of the second component of the velocity.

Remark 4. From the inequalities (33) and (37) we see that the instability can appear if the basic velocity $\overline{u}$ is less than a critical value, which however depends on the variation and the magnitude of the basic surface tension. This is not in contradiction with the experimental data reported by Chan and Liang (1997), because the basic velocity and surface tension are related and we not know the basic solution. However, in our analysis, the basic velocity is appearing in the stability parameters of the flow.

3. THE HORIZONTAL HELE-SHAW CELL

In this section we study the case of a less viscous displacing fluid, then $\mu_1 < \mu_2$, in a horizontal Hele-Shaw cell with a surfactant-wetting layer deposed on the inner walls – see Figure 2.

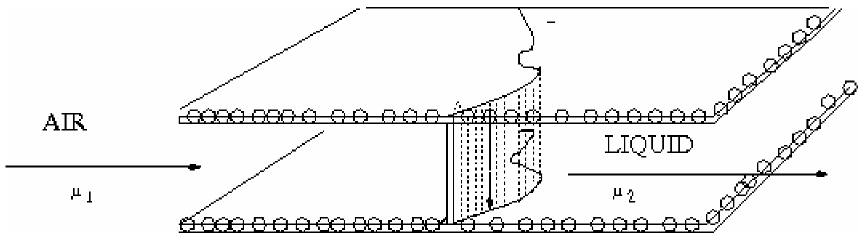

Fig. 2 – Horizontal Hele-Shaw cell with wetting-surfactant adherent layer on inner walls.

We consider here a Hele-Shaw model with a particular slip condition. The plates of the cell are parallel with the plane xOz and the gap is $2d$. The displacing process is in the Ox direction and the axis Oy is orthogonal on plates, described by the equations $y = 0$ and $y = 2d$. The gravity (pointed down in the negative Oy direction) is neglected.

We derive first a Darcy-type equation for a single fluid with viscosity η, subject to non-slip conditions on the plates.

Let (u, v, w) be the velocities components in the directions Ox, Oy, Oz and p the pressure. As usual in this model, we neglect the vertical component v of the velocity. We neglect also u_x, u_{xx}, w_x, w_{xx} and u_z, u_{zz}, w_z, w_{zz} in front of u_y, u_{yy}, w_y, w_{yy}. We consider u and w as functions of x and y only. The Stokes equations become

$$\eta \cdot u_{yy} = p_x, \quad \eta \cdot w_{yy} = p_z, \quad 0 = p_y, \tag{38}$$

where η is the viscosity. It is known that a basic solution exists with a sharp planar interface.

At the distance δ on the both inner walls we consider the following boundary conditions:

$$u(x, y = \delta) = u(x, y = 2d - \delta) = \epsilon, \quad w(x, y = \delta) = w(x, y = 2d - \delta) = 0. \tag{39}$$

The basic steady solution with slip condition (39) is obtained as follows. We neglect all terms containing $2d\delta$ and δ^2. We solve the above problem (38)-(39) and get

$$\eta \cdot u = p_x(y^2/2) + Ay + B; \quad \eta \cdot w = p_z(y^2/2) + Cy + D; \tag{40}$$

$$\eta\epsilon = p_x(\delta^2/2) + A\delta + B; \quad \eta\epsilon = p_x(2d - \delta)^2/2 + A(2d - \delta) + B; \tag{41}$$

$$0 = p_y(\delta^2)/2 + C\delta + D; \quad 0 = p_y(2d - \delta)^2/2 + C(2d - \delta) + D. \tag{42}$$

Subtracting the two relations (41) we get

$$0 = p_x\frac{1}{2}[\delta^2 - (2d - \delta)^2] + A[\delta - (2d - \delta)];$$

$$0 = p_x \frac{1}{2} 2d(2\delta - 2d) + A(2\delta - 2d) \Rightarrow \quad A = -dp_x;$$
$$\eta\epsilon = p_x \delta^2/2 - 2dp_x\delta/2 + B \Rightarrow B = \eta\epsilon + p_x(2d\delta - \delta^2)/2 \approx \mu\epsilon;$$

$$u = \frac{1}{2\eta} p_x [y^2 - 2dy + (2d\delta - \delta^2)] + \epsilon \approx \frac{1}{2\eta} p_x (y^2 - 2dy) + \epsilon. \tag{43}$$

Subtracting the two relations (42) we get

$$w = \frac{1}{2\eta} p_z [y^2 - 2dy + (2d\delta - \delta^2)] \approx \frac{1}{2\eta} p_z (y^2 - 2dy). \tag{44}$$

We consider below the mean velocities $\overline{u}, \overline{v}$ and obtain

$$\begin{aligned} \overline{u} &= \frac{1}{2d} \int_{\delta}^{2d-\delta} u(x,y)\mathrm{d}y = \frac{1}{\eta} p_x [-\frac{d^2}{3}] + \epsilon \frac{d-\delta}{d}, \\ \overline{w} &= \frac{1}{2d} \int_{\delta}^{2d-\delta} v(x,y)\mathrm{d}y = \frac{1}{\eta} p_z [-\frac{d^2}{3}]. \end{aligned} \tag{45}$$

We define the "permeability" $K = d^2/3$, then we get

$$\overline{u} = -\frac{K}{\eta} p_x + \epsilon \frac{d-\delta}{d}, \quad \overline{w} = -\frac{K}{\eta} p_z. \tag{46}$$

$$p_x = -\frac{\eta}{K} \overline{u} + \frac{\mu}{K} \epsilon \frac{d-\delta}{d}, \quad p_z = -\frac{\eta}{K} \overline{w}. \tag{47}$$

The component $\overline{w}$ of the basic velocity is neglected in front of $\overline{u}$, then we have $p_z = 0$.

Consider now two immiscible liquids. Let u_1, u_2 and μ_1, μ_2 be the horizontal velocities, the viscosities and $\overline{u}_i = \nabla \Phi_i$. Then $\eta = \mu_1$ for the displacing liquid, $\eta = \mu_2$ for the displaced liquid and the pressures in both liquids are

$$p_1 = -\frac{\mu_1}{K} \Phi_1 + \frac{\mu_1}{K} \epsilon x \frac{(d-\delta)}{d}; \qquad p_2 = -\frac{\mu_1}{K} \Phi_2 + \frac{\mu_2}{K} \epsilon x \frac{(d-\delta)}{d}. \tag{48}$$

The flow direction of the basic solution is in the positive axis Ox. In the moving frame $X = x - Vt$, the basic interface is $X = 0$. However, we still use $x = 0$ for denoting the basic interface, where we have Laplace's law and consider a constant surface tension T. The basic interface is planar, therefore the pressure is continuous across the interface.

We study now the stability of the above basic solution. Like in [14], the perturbed interface is described by

$$x = b \exp(\mathrm{i}\, nz + at). \tag{49}$$

From the continuity of normal velocity at the planar interface, it follows that near $x = 0$, in the first order approximation, the velocity potentials in the two regions satisfy the relations:

$$(\Phi_1)_x = (\Phi_2)_x = V + a\, b \exp(\mathrm{i}\, nz + at). \tag{50}$$

The above potentials must vanish far downstream and upstream. Therefore we can assume

$$\Phi_1 = Vx + (ab/n) \exp(\mathrm{i}\, nz + at + nx), \quad x < 0; \tag{51}$$

$$\Phi_2 = Vx - (ab/n) \exp(\mathrm{i}\, nz + at - nx), \quad x > 0. \tag{52}$$

We use the approximate form of the Laplace's law: $p_2 - p_1 = T x_{yy}$. Here T is the surface tension on interface and x_{yy} is the first order approximation for the interface curvature. Then, in the first order approximation, from (48)–(52) we get

$$\left[-\frac{\mu_2}{K}(Vx - \frac{xa}{n}) + \frac{\mu_2}{K}\epsilon x \frac{d-\delta}{d}\right] - \left[-\frac{\mu_1}{K}(Vx + \frac{xa}{n}) + \frac{\mu_1}{K}\epsilon x \frac{d-\delta}{d}\right] = -Tn^2 x.$$

Recall the perturbed interface (49). We simplify with x and obtain the formula of the growth constant a:

$$\frac{a}{Kn}(\mu_1 + \mu_2) = \frac{\mu_2 - \mu_1}{K}\left[V - \epsilon\frac{(d-\delta)}{d}\right] - Tn^2. \tag{53}$$

The new term ϵ (containing the slip-velocity) appears, compared with the Saffman-Taylor formula – see [14]. It follows:

$$\mu_1 < \mu_2 \quad \text{and} \quad V - \epsilon\frac{(d-\delta)}{d} < 0 \Rightarrow \quad a < 0. \tag{54}$$

Therefore, if the displacing liquid is *less* viscous and the interface velocity V is less than the critical value $\epsilon(d-\delta)/d$, we have a *stable* flow.

If $\epsilon = 0$, then in the classical Hele-Shaw cell, we obtain always instability, because $V > 0$.

Later on, we obtain the value of ϵ in terms of the variable surface tension on the surfactant-layer surfaces $y = \delta$ and $y = 2d - \delta$, by using appropriate boundary conditions related with the surfactant-driven Marangoni stress.

We study now the effect of preexisting layer of surfactant on the plates, when a surfactant layer of fluid of thickness δ exists on both plates of the Hele-Shaw cell, obtained by the coating procedure of Landau-Levich (see [16] and references therein). In the frame of *low* capillary numbers Ca, the thickness δ of the surfactant layer is

$$\delta \approx Ca^{2/3} = \{\mu U/S\}^{2/3}, \tag{55}$$

where both plates were pulled out with the velocity U from a reservoir filled with a surfactant -fluid of viscosity μ and surface tension S with air. We suppose

$$\mu < \mu_1,\ \mu_2. \tag{56}$$

We derive now the boundary conditions between the Helen-Shaw liquids and the preexisting surfactant layer. Let $\Gamma(x), \sigma(\Gamma)$ be the surfactant concentration on the surface of the surfactant layer and the (variable) surface tension which acts on the layer surface. Then, as in [12, 13], the conditions on the layer surface can be considered of the form

$$p_l - p_i = -\sigma^i \cdot C; \qquad \mu_i \cdot \partial u_i/\partial n = -\sigma^i_x, \tag{57}$$

where p_l is the pressure in the surfactant-layer and p_i is the pressures in the liquid i. C is the curvature of the layer surface, σ^i is the surface tension between the liquid i and the wetting- layer, $\partial u/\partial n$ is the normal derivative of u.

In our case, the preexisting horizontal layer is of almost constant thickness, then the curvature C is zero and from $(57)_1$ we obtain a continuous pressure.

We use the second condition (57) and obtain the following important result:

Even if the fluids in the Hele-Shaw cell are at rest (that means $V = 0$), we still have a slow flow near the plates given by the condition $(57)_2$.

This is known as Marangoni effect: the flow induced by the variation of the surface tension on the interface between the liquids and the preexisting layer of surfactant.

For obtaining the above result, a condition is derived from the second relation (57), by using a discretization formula of the normal derivative of the liquid velocity u_i. It can be seen that the formula $(57)_2$ must be used in terms of the *interior normal* derivative on the plates. Then we obtain the conditions

$$y = \delta, \quad y = 2d - \delta \quad \Rightarrow \mu_i \cdot \partial u_i/\partial n = -\sigma^i_x. \tag{58}$$

Remark 5. We consider $\sigma^i_x < 0$. This is related with an increasing surfactant concentration on the wetting-layer.

The surfactant surface is orthogonal on Oz. The derivative of u in terms of z is approximated as follows (our analysis is only for *small* δ)

$$u_y(y = \delta) \approx \frac{u(\delta) - u(0)}{\delta} + O(\delta^2). \tag{59}$$

$$u_y(y = 2d - \delta) \approx \frac{u(2d) - u(2d - \delta)}{\delta} + O(\delta^2). \tag{60}$$

The preexisting surfactant layer is adherent on the plates, then we have $u(0) = u(2d) = 0$. Therefore the *interior* normal derivatives (directed into the plates) are

$$u_n(y=\delta) \approx \frac{u(\delta)}{\delta}; \quad u_n(y=2d-\delta) \approx \frac{u(2d-\delta)}{\delta} \tag{61}$$

We use the last four relations and obtain the boundary conditions verified by the displacing liquids *on the surface of the preexisting layer*:

$$\mu_i u_i(\delta) = \mu_i u_i(2d-\delta) = -\delta\sigma_x^i \tag{62}$$

We emphasize here that the above two formulas are giving us a *positive* velocity – see *Remark 5*. This is consistent with our assumption concerning the positive displacement direction.

Remark 6. For a steady solution, we need a constant velocity of the initial straight interface between the displacing liquids μ_1, μ_2. Therefore we need $u_1 = u_2$ and get the necessary condition

$$\frac{\sigma_x^1}{\mu_1} = \frac{\sigma_x^2}{\mu_2}. \tag{63}$$

However, the above condition could be removed in the particular case when μ, μ_1 are very small. In this case, the displacing fluid is not so much affected by the surfactant-driven Marangoni stress.

We use now the relation (62), to get the small velocity on the surfactant layer. With $\epsilon = -\delta\sigma_x^i/\mu_i$, our *slip* conditions on the Hele-Shaw plates are

$$y = \delta \quad \text{and} \quad y = 2d-\delta \quad \Rightarrow u_i = -\delta\sigma_x^i/\mu_i. \tag{64}$$

The new boundary conditions and the formula (53) give the following equation which governs the flow stability:

$$\frac{a}{Kn}(\mu_1+\mu_2) = \frac{\mu_2-\mu_1}{K}\left[V + \frac{\delta\sigma_x^i(d-\delta)}{\mu_i\, d}\right] - Tn^2. \tag{65}$$

The *main result of this section* is obtained from the formula (65), when the displacing liquid is less viscous, according with *Remark* 6.

The Saffman-Taylor analysis gives us an unstable flow, because $\sigma_x^i = 0$, $V > 0$ and we get

$$\max_n(a) > 0. \tag{66}$$

Recall $\sigma_x^i < 0$ – see *Remark* 5. Suppose the condition (63) holds and

$$V < -\frac{\delta\sigma_x^i(d-\delta)}{\mu_i\, d}. \tag{67}$$

Then the right hand side of (65) becomes *negative*, that means the flow is *stable* and we get *a reverse Saffman-Taylor instability.* We can see that the right hand side of the above inequality is positive, then there exists *a small positive* V which satisfies this condition.

Remark 7. As we pointed out in Introduction, the paper [2] is related with this phenomenon. An instability is reported only for a *large* enough V, when the displacing fluid is *the same* as the preexisting liquid layer on the plates, which is not our case. Therefore our result is not in contradiction with the experiment of Chan and Liang [1], and reveals a new effect of surfactants on the flow in Hele-Shaw cells

4. CONCLUSIONS

We study here a particular flow in the case of a vertical and horizontal Hele-Shaw cell, when the inner part of the plates are coated by a thin film of surfactant.

For the vertical cell, the starting point is the experiment of Chan and Liang, described in [2]. In contradiction with the classical Saffman-Taylor criterion (for the clean plates of the Hele-Shaw cell), in [2] was reported an instability of the surface between immiscible fluids, even if the displacing fluid is *more* viscous. This new instability phenomenon, which can be considered as a "reverse Saffman-Taylor instability" is related with the effects of surfactants on the flow in a thin gap (see [5, 6, 12, 13, 15, 16] and references therein).

Before the experiment obtained in [1], some theoretical studies concerning this problem were initiated in [9] and [10], where a non-physical state equation for the surface tension was used – a linear dependence in term of the surface curvature.

An attempt to explain the above new instability phenomenon was given in [3], where a state equation of the surface tension is proposed, in term of the deformation of the interface.

A basic study related with the considered problem was given in [7] – the authors used the 3D Stokes equations to describe the flow in the "contaminated" Hele-Shaw cell and performed a stability analysis of a basic solution. They partially confirmed the experimental result of [1], but pointed out that rather the variation of the surface tension on the wettin-layer is giving the new-observed instability, and not the interface velocity, which is not appearing in the stability parameter of the flow.

In this paper, we perform a stability analysis similar with those of Krechetnikov and Homsy given in [7]. We remark a small error in the stability analysis given in this cited paper – the last equation (4.4) page 116 (see *Remark* 2).

We carefully analyse the boundary conditions verified by the perturbations of the basic solution on the wetting-layer surface (see *Remark* 1). Starting with the relations (23)–(26), we reconsider the stability analysis and conclude that we have two important cases: a very *slow* and a relative *large* variation of the surfactant concentration on the wetting-surfactant layer adhering on the Hele-Shaw plates. In the last part of Section 2, we pointed out the corresponding conclusion in the two mentioned cases:

1) In the case of an almost constant surface tension (for a very slow variation of the surfactant concentration) we get instability if the surface tension is large enough.

2) In the case of a variable surface tension (for a large variation of the surfactant concentration) we obtain instability if the corresponding Marangoni number *or* the surface tension are large enough.

However, our result is far from being rigorous. It is not so clear what "small" and "large" variation of the surfactant concentration mean. The main mathematical arguments here are the boundary conditions for the perturbations of the basic solution, analyzed in *Remark* 1 and the equation of the perturbed interface.

In the case of horizontal cell – Section 3 – we describe a Hele-Shaw flow with a slip condition on the plates. We obtain the slip condition in terms of the variable surface tension on the layer surface – see the formula (64). A stability analysis is performed and we get the formula (53) for the growth constant of the perturbation, which contains a new term depending on the Marangoni stress on the wetting-layer surface. We get the dispersion formula (65) and the relation (67), therefore the stability conditions are obtained in terms of the displacing velocity V: *The flow is stable if V is less than a critical value, even if the displacing fluid is less viscous.* However, we need the particular condition (63), because the surface tension on the wetting-layer surface is not the same for both displacing liquids. This result is not in contradiction with the experiment reported by Chan and Liang in [2], as we pointed out also in *Remark* 7.

We emphasize that in both considered cases, the displacing velocity is appearing in the stability condition of the flow (even if it is related with other parameters). The variation of the surface tension is not the only factor giving the instability and some problems are not clarified, concerning a more exact description of the relations between all parameters of the flow.

Therefore, further studies are necessary to clarify the effects of the wetting-surfactant layer on the Hele-Shaw cell, especially for the vertical Hele-Shaw cells.

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