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Wettability effects on the dynamics of fluid displacement through capillary tubes

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Abstract We have performed experiments where one fluid is displaced by another immiscible one in capilarry tubes. The fluids have different wettability properties for the tube walls. The purpose of this work is to study the effect of the wettability on the velocity of the meniscus at the interface between the two fluids. The flow is induced by a small (or zero) pressure drop which is of the same order of magnitude (or smaller) than the capillary pressure difference between the fluids. We measure variations with time of the displacement velocity of the meniscus: we compute from this variation the capillary pressure value. Under strong wetting conditions (kerosene displacing air) the variations of the dynamic capillary pressure p_c^d with velocity are very small, in agreement with the predictions of the Hoffman-Tanner theory. Under weaker wetting conditions (water displacing cyclohexane), we have measured the variations of p_c^d as a function of the velocity; these variations are one order of magnitude larger than those predicted by Cox.

Introduction

Mixed wettability deals with the exchange of wetting and non wetting fluids and is an essential notion for the study of diphasic flow in porous media (enhanced oil recovery, hydrogeology)[1].

However it is difficult to analyze, due to the addition of surface state effects, as well as the complex interna1 geometries.

Experimental models involving diphasic flow in capillary tubes illustrate the basic interplay between viscous and capillary effects. The latter are particularly

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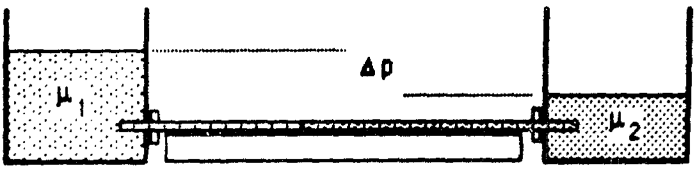
important when they are of the same order of magnitude as the applied pressure head.

The present work is part of a program involving various configurations of capillary tube flows.

1. Experimental procedure

The experimental system was designed to put in a horizontal position a capillary tube, connected with two reservoirs, where the level of the liquids could be controlled.



The experiments on displacement of immiscible fluids were realized with a constant, Ap, between the entrance and the exit of the tube.

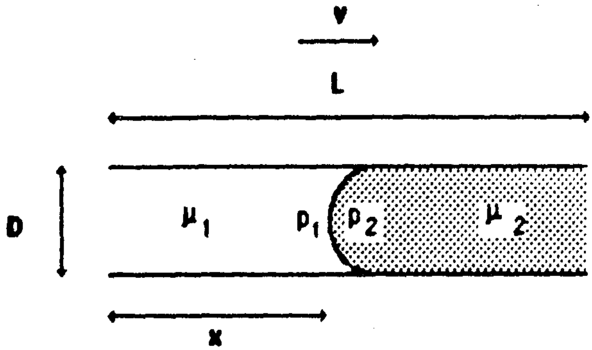
A system connected to a computer allowed us to record the interface position as a function of time.

The capillaries used are of length $L = 85.0 \pm 0.1$ cm and their inside diameter is $D = 0.110 \pm 0.004$ cm. The pressure difference, Ap, was measured with an accuracy of ± 50 dyne/cm^2.

From the Washburn[2] equation for two-phase flow in a capillary tube we obtain

$$[\mu_1 x + \mu_2 (L - x)]dx = \frac{D^2}{32}(\Delta p + p_c^d)dt \qquad (1)$$

Where



We define the dynamic capillary pressure by the following relation

$$p_c^d = p_2 - p_1 = p_c^e - p(v) \tag{2}$$

where $p_c^e = 4\gamma \cos\theta / D$ is the static capillary pressure and 7 is the interfacial tension and have introduction additional pressure term $p(v)$ opposing the flow due to the presence of the meniscus. It results from a sum of two effects: the pinning of the triple contact line and the recirculation zones in the meniscus neighbourhood.

Integrating both sides of eq. (1) we obtain

$$\int_0^{t_i} p_c^d dt = \frac{32}{D^2}\left[\frac{\mu_1 - \mu_2}{2} x_i^2 + \mu_2 L x_i\right] - \Delta p t_i \tag{3}$$

Using the experimental data (x_i, t_i) we plot the right hand side of (3) $A(t)$, as function of time. The derivative of the curve obtained is p_c^d.

2. Experimental results

In a first series of experiments we used air, kerosene and water as fluids. Ten experiments were done with the following stages: a) kerosene displacing air (imbibition); b) water displacing kerosene (imbibition); c) kerosene displacing water (drainage).

2a. Imbibition: Kerosene displacing air

Here $\mu_1 = 1.65$ cp, $\mu_2 \sim 0$ cp. We kept the pressure difference Ap = 0. One set of values of the $x(t)$ experimental data is displayed on figure **1.** The velocity **of** the interface decreases as the amount of kerosene in the tube increases, because the average viscosity increases.

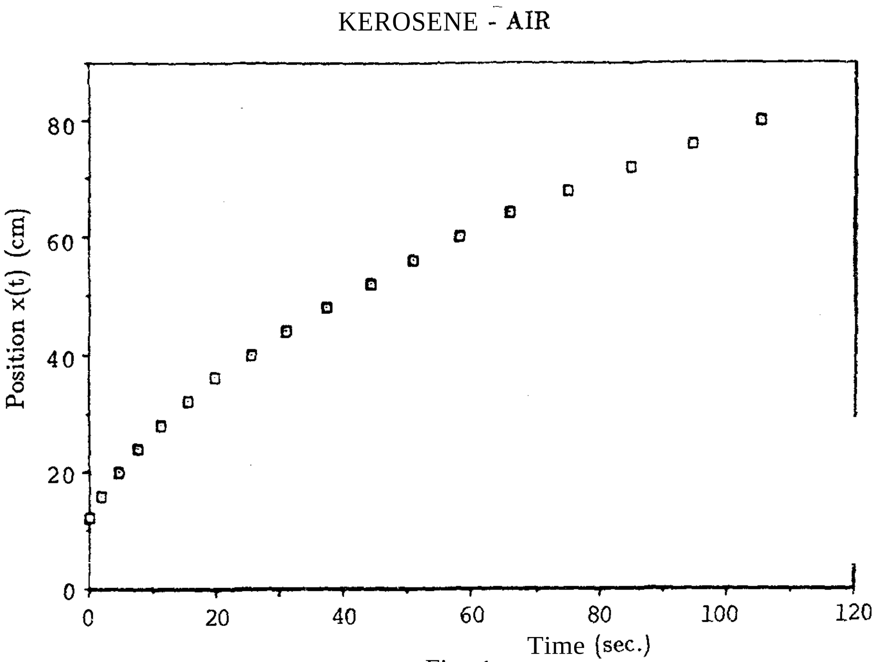


Fig. **1**

In each case, p_c^d was evaluated for velocities ranging from 2.7 cm/sec to 0.3 cm/sec (**Ca max** - 10^{-3}; Ca min $\sim 10^{-4}$, where **Ca** $= \mu v/\gamma$ is the capillary number).

As in this case is $\mu_2 = 0$ and $\Delta p = 0$, from **(3)** we obtain:

$$A(t) = \frac{16}{D^2}\mu_1 x_i^2(t)$$

$A(t)$ is plotted in figure **2** and it can be seen that it is a straight line which, according to (3), indicates that p_c^d has a constant value for this range of velocities. Its value is, for all the experiments done, **1180** $\pm$ 50 dyne/cm^2.

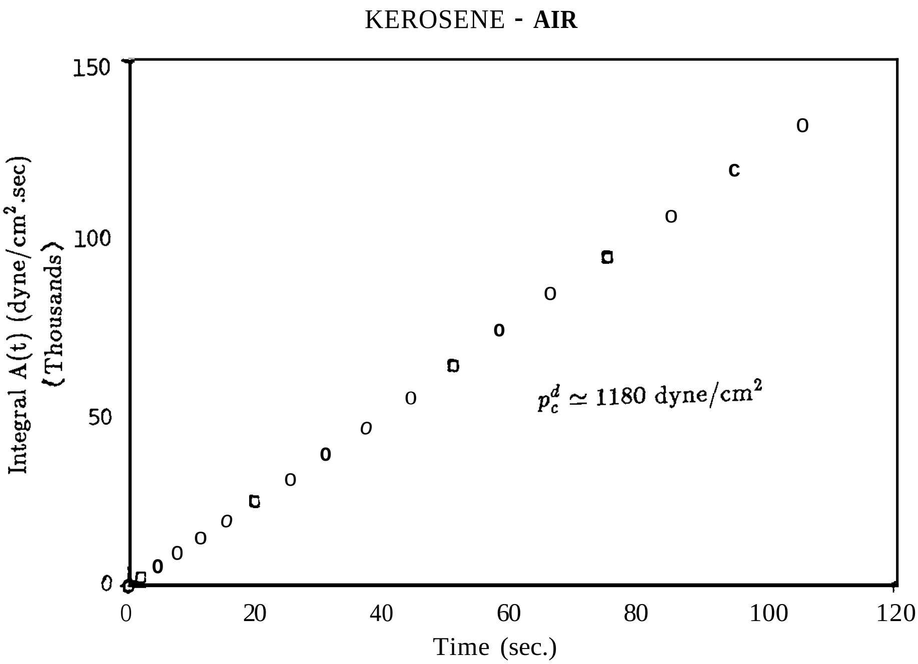


Fig. 2

The static capillary pressure p_c^e, was evaluated from the capillary ascension of kerosene, in a vertical tube. Its value is 1330 ± 50 dyne/cm^2.

From (2), it appears that, for this range of velocities, $p(v)$ is 150 dyne/cm^2.

Let us compare these results with the predictions of the Hoffman-Tanner Law valid for Ca<< 1 and small contact angles[3].

$$p_c^d \sim p_c^e(1 - 10Ca^{2/3}) \tag{4}$$

For a liquid displacing a gas and Ca between 10^{-4} and 10^{-3}, eq.(4) gives a variation in the order of 10% of p_c^d ($\sim$ 120 dyne/cm^2 in the present case). This value is approximately of the same order as the difference between p_c^d and p_c^e.

Therefore we conclude that our results are in agreement with the Hoffman-Tanner law within the experimental errors.

2b. Imbibition: Water displaçing kerosene

In this case $\mu_1 = 1$ cp, $\mu_2 = 1.65$ cp. The experiments were also done with **Ap** = 0. One set of values of $x(t)$ is shown in figure *3*. In figure 4, we plot the corresponding $A(t)$.

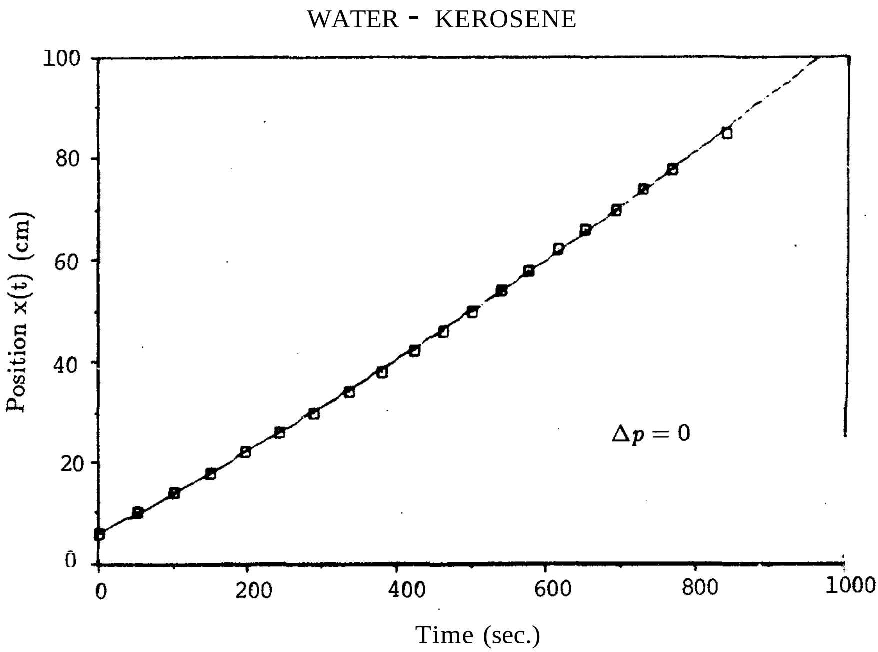


Fig. *3*

The velocity of the interface increases from *0.08* cm/sec (Ca = 3×10^{-5}) to *0.13* cm/sec (Ca = 4×10^{-5}) as the water replaces kerosene in the tube.

Again, it can be seen that in this range of velocities p_c^d is constant. Its value, given be the slope of the linear variation, is, for all cases, around *210* dyne/cm^2. The static capillary pressure, p_c^e, was evaluated by measuring the pressure difference Ap necessary to immobilize the interface. The resulting value was *550* ± *50* dyne/cm^2.

From *(2)* we found that the value of $p(v)$ in this range of velocities is constant and approximately *350* dyne/cm^2.

WATER - KEROSENE

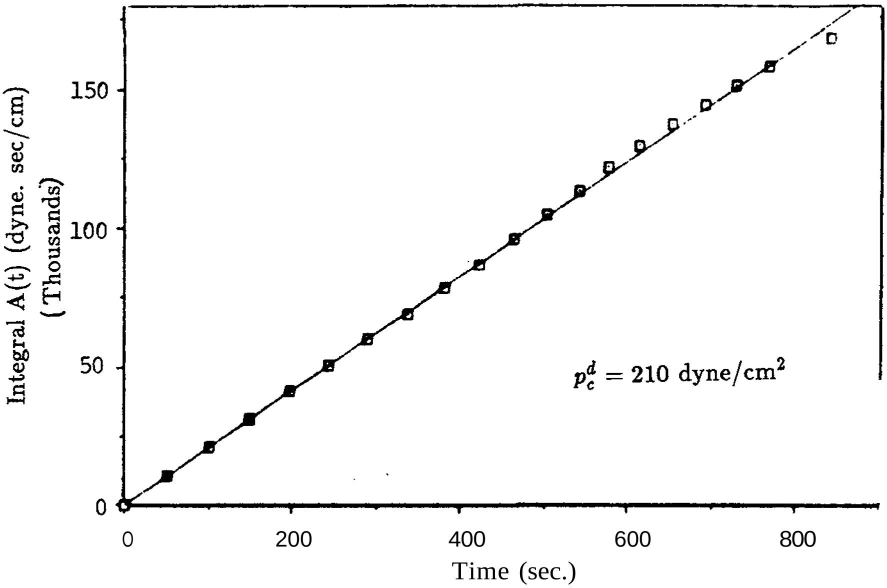


Fig. 4

2c. Drainage: kerosene displacing water

In order to be in the drainage regime we applied a constant pressure difference Ap = 1800 dyne/cm^2. The range of velocities is from 0.25 cm/sec to 0.4 cm/sec, (Ca max = 8×10^{-3}, Ca min = 5 x 10^{-3}.

In figs. 5 and 6, we plot, for one of the experiments done, the variations of $x(t)$ and $A(t)$ respectively.

From these variations, we again obtain in this case that, for these velocities, p_c^d is constant. Its value is -920 dyne/cm^2.

While in the former experiment

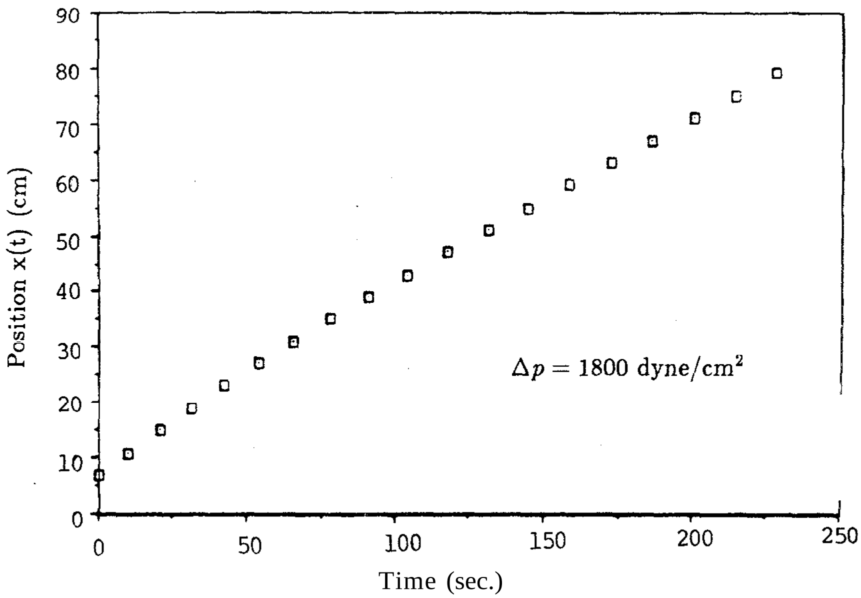
$$p_c^d = p_c^e - p(v) \ ,$$

In this case both p_c^e and $p(v)$ oppose the flow and then

$$p_c^d = p_2 - p_1 = -p_c^e - p(v) = -920 \ \ \text{dyne/cm}^2 \ .$$

With $p_c^e = 550$ dyne/cm^2, we obtain $p(v) = 350$ dyne/cm^2 indicating that $p(v)$ is independent of the sign of the velocity and is the same for drainage and imbibition.

KEROSENE - WATER



Big. 5

2d. Dynamic capillary effect

The purpose of this second series of experiments was to study how p_c^d changes with velocity. The fluids used water and cyclohexane. The wettability of these two fluids is similar, so the static capillary pressure, $p_c^e = 300$ dyne/cm^2, is low. This allows a large range of p_c^d values in the velocity range where x and t recordings could possibly be affected.

In fig. 7 the experimental data for $x(t)$ from one out of the **22** experiments done are shown.

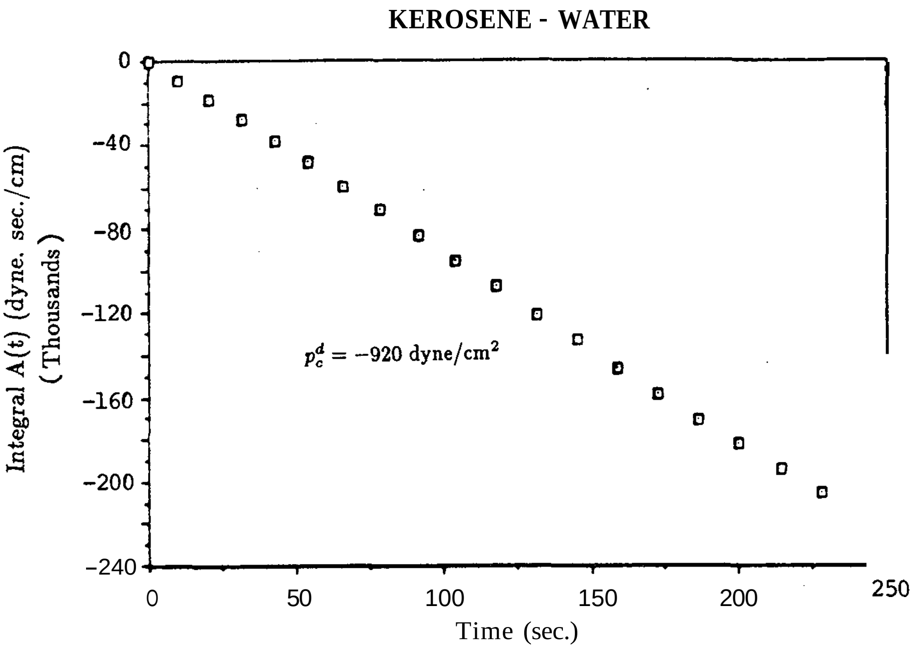


Fig. 6

This is the case of a fluid displacing a second one with $\lambda = \mu_1/\mu_2 \simeq 1$, $\mu_1 = \mu_2 \cong 1$ cp, so $x(t)$ is a straight line. From (3) one gets

$$p_c^d = \frac{dA(t)}{dt} = \frac{32}{D^2}\mu_2 L \frac{dx}{dt} - \Delta p$$

p_c^d is displayed in fig. 8. It takes a constant value for velocities smaller than 0.1 cm/sec (Ca $\sim 5 \times 10^{-5}$); from then onwards it decreases as velocity increases.

Ca ranges from 3×10^{-6} to 10^{-3}, and we found that p_c^d varies from **50** dyne/cm^2 to **-600** dyne/cm^2.

It can be seen that when v $\rightarrow 0$, $p_c^d \rightarrow 50$ dyne/cm^2, while $p_c^e \sim 300$ dyne/cm^2. That is $p(v) \sim$ **250** dyne/cm^2 for $0 < v < 0.1$ cm/sec.

In conclusion, our experimental results suggest that, in the equation

$$p_c^d = p_c^e - p(v)$$

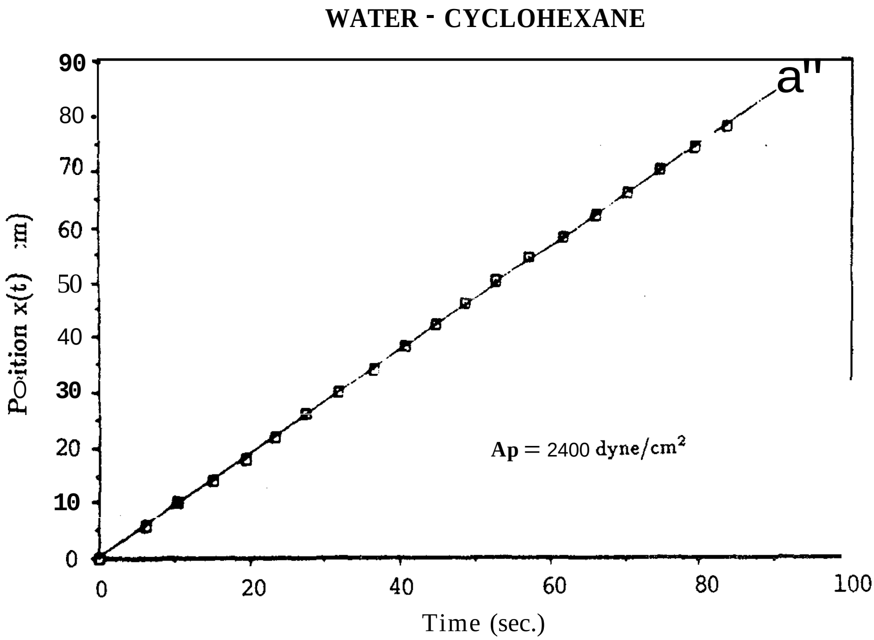


Fig. 7

$p(v)$ would not be merely proportional to $\cos\theta_d$ but it could also be expressed as:

$$p(v) \sim \cos\ \theta_d + f(v)$$

where f (v) is an additional contribution whose physico-chemical origin is unknown.

In this case of similar wetting, the decrease of $p(v)$ when v grows is an order of magnitude larger than that predicted by Cox[4] for θ_d.

This difference could be attributed to the term f (v).

3. Discussion

This behaviour of p_c^d is similar to that of friction forces in solids. We have also observed this analogy in the existence of hysteretic behaviour when using increasing and decreasing applied pressure. A different pressure gradient is needed

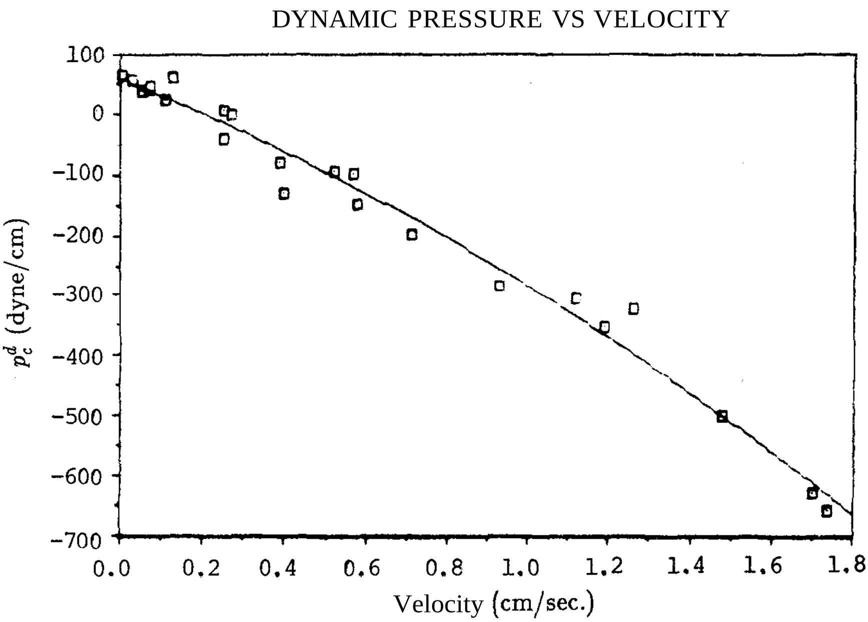


Fig. 8

in order to set a meniscus in motion than that needed in order to stop it (difference between static and dynamic friction).

The instance of a liquid displacing another has been theoretically studied by Cox and experimentally by Fermigier[5]. Both agree that for Ca $< 10^{-3}$ the dynamic contact angle does not vary with Ca.

However, our observations indicate that p_c^d starts to decrease from Ca $\simeq 5 \times 10^{-5}$. If we accept that any variation on p_c^d is due to a variation in the dynamic contact angle, our results are not in agreement with Cox's predictions and with the measurements by Fermigier.

If, on the contrary, we accept that the decreasing value of p_c^d can not be merely described by the contact angle variation but that there can be other contributions to its decrease, our results can be in agreement with those mentioned.

Therefore, this is an open problem (that requires aditional experimentation) and where the existence of wettability (liquid-solid interaction) requires precise definitions not always well understood on the dynamic point of view.

References

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Resumo

Realizamos experiências em que um fluido é deslocado por outro, imiscível, em tubos capilares. Os fluidos têm diferentes propriedades de molhabilidade com relação às paredes do tubo. O objetivo deste trabalho é estudar o efeito da molhabilidade sobre a velocidade do menisco na interface entre os fluidos. O escoamento é induzido por uma pequena (ou nula) queda de pressão da mesma ordem da grandeza (ou menor) que a diferença de pressão capilar entre os fluidos. Medimos variações temporais da velocidade de deslocamento do menisco, e a partir daí calculamos o valor da pressão capilar. Sob condições fortes de molhabilidade (querosene deslocando ar), as variações da pressão de p_c^d como função da velocidade; estas variações são uma ordem de grandeza maiores do que as previstas por Cox.