Imbibition Driven by a Temperature Gradient

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In this work, we have theoretically studied the imbibition process in a cylindrical capillary under a constant, longitudinal temperature gradient, G. A closed-form analytical solution has been obtained and the Washburn law (valid for the isothermal case) has been found to hold for $G = 0$. The space and time evolution of the interface is strongly dependent on surface tension and the viscosity with temperature. By using reported data for an organic oil (squalene), we showed how imbibition can be accelerated when the temperature gradient is negative.

In nature and in technology, there are many systems where fluids spontaneously penetrate into capillaries or porous materials.[1–3] In isothermal cases, spontaneous flow occurs essentially due to driven capillary pressure, p_c, which originates from surface tension, σ. In a cylindrical capillary, this pressure is $p_c = 2\sigma \cos\theta/a$, where a is the radius of the capillary and θ is the contact angle of the liquid–air interface with the solid surface (see Fig. 1). Due to the dominant viscous force within the capillary it is reasonable to consider the flow as a Poiseuille-like flow. In this case, the imbibition front without considering gravity, $h(t)$, advances in accordance with the well-known law[4] $h^2 = (\sigma a \cos\theta/2\mu)t$, where t is the time elapsed to reach the distance h, μ is the dynamic viscosity and ρ is the fluid density, all considered constants in an isothermal case. On the other hand, when the imbibition occurs in a fully saturated porous sample, the radius a given in the previous relation, has to be changed by the size of the effective hydraulic pore, d_h, which is a function of the permeability of the porous medium, K_m.

In a more general case, imbibition can occur when the capillary or the host porous material are differently heated. In an oil reservoir, for example, this is a common situation because there exists a geothermal gradient which can be assumed as constant close to the earth's surface. Also, in the same context, during enhanced oil recovery, heat is frequently introduced into the reservoir by injecting steam or hot water, generating strong temperature gradients around the injection zones.[5] Other types of systems where capillary forces are influenced by temperature gradient, are those referred to as microfluidic pumping systems,[6–9] where spontaneous upward flows are produced on vertical walls under temperature gradients by surface tension gradients (Marangoni films). The treatment of these systems is the main motivation for searching the possibility of several kinds of imbibition flow within a capillary under longitudinal temperature gradients.

Because some samples of porous materials can be considered to be made of a bundle of capillary tubes of mean pore size, d_h, determining the effect of temperature gradient on the flow in a single capillary is a fundamental step that could be useful in understanding the capillary flow in these more complex systems. Thus, we consider the temperature variations in both surface tension and dynamic viscosity as the cause of the notable changes in the behavior of the flow in a cylindrical capillary under the temperature gradient G, without considering gravity. Later, we shall discuss a working case where theoretical predictions show strong differences between isothermal and non-isothermal imbibition processes. Finally, at the end of this letter, we shall discuss the main conclusions and limitations of our approach.

Here, we perform a theoretical study in order to show the effect on capillary flow of a constant temperature gradient imposed along a cylindrical capillary, $G = \Delta T/H$, where $\Delta T = (T_u - T_0)$ is the temperature difference between the upper (T_u) and lower (T_0) tips of the capillary and H is its length. This is shown in Fig. 1. For such a type of creeping flow with a Reynolds number smaller than unity, the fluid during its advance achieves automatically the solid temperature distribution. Thus, the diffusive regime is assumed to be dominant. This last condition is commonly met, for instance, in thin capillaries because the diffusive time $t_D = a^2/\alpha_f$ (α_f is the thermal diffusivity of the fluid) is very small compared with the transit time, $t_I = a/|dh/dt|$.

Assuming that gravity is absent and that a Hagen–Poiseuille flow is developed within a thin pipe by capillary pressure, the wall shear stress is $\tau_w = 4\mu\bar{u}/a$,[2] which acts
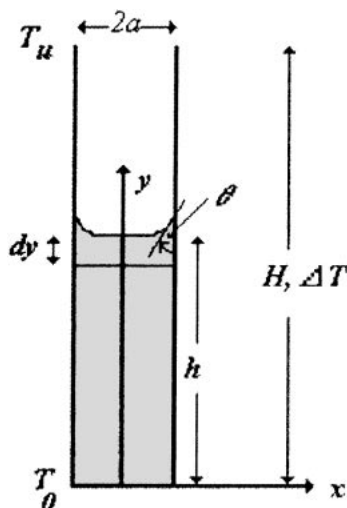

Fig. 1. Schematic view of the imbibition phenomenon in a cylindrical capillary of radius a and length H under the temperature gradient $G = \Delta T/H$.

on the inner surface of the capillary. Here, $\overline{u}$ is the mean velocity of the flow. The motion equation is then obtained by balancing the (frictional) viscous force with the capillary force, resulting in

$$\int_0^{h(t)} \tau_w dA = 2\pi\sigma(h(t))a\cos\theta. \quad (1)$$

Here, the viscous term has to be integrated from 0 to $h(t)$ because it acts on each differential element of area $dA = 2\pi a dy$, adjacent to the capillary's inner wall, while the capillary force itself acts only on the entire free surface πa^2 at $y = h$. This type of approach has been successfully employed for an isothermal case in a series of studies related to imbibition (For instance, see refs. 10–13). In the present case, as was mentioned above, the viscosity is affected by the spatial change in temperature along the capillary, but the surface tension depends on the instantaneous height, $h(t)$, of the free surface (imbibition front), where temperature is known $T(y = h) = Gh$. Assuming that contact angle θ does not change with temperature or front movement, eq. (1) takes the form

$$4\frac{dh}{dt}\int_0^{h(t)} \mu(y)dy = a\sigma(h(t))\cos\theta. \quad (2)$$

To solve this equation by introducing the existence of a temperature gradient, we assume a linearized form of the dependence of μ and σ on temperature, such that $\mu = \mu_0(1 + 1/\mu_0(d\mu/dT)Gy)$ and $\sigma = \sigma_0[1 + 1/\sigma_0(d\sigma/dT)Gh]$, where μ_0 and σ_0 are the values of dynamic viscosity and surface tension at a reference temperature, $T = T_0$, respectively. By substituting both relations in eq. (2) and evaluating the integral, we obtain

$$\frac{\left[h + \frac{Gh^2}{2\mu_0}\frac{d\mu}{dT}\right]}{\left[1 + \frac{Gh}{\sigma_0}\frac{d\sigma}{dT}\right]}\frac{dh}{dt} = \frac{a\sigma_0\cos\theta}{4\mu_0}. \quad (3)$$

This nonlinear differential equation has an exact solution, which can be expressed in a simple form using dimensionless variables $[\xi = h/a$ and $\tau = (\sigma_0\cos\theta/4a\mu_0)t]$ and dimensionless parameters that take into account the changes in viscosity and surface tension with temperature.

$$A = \frac{Ga}{2\mu_0}\frac{d\mu}{dT} \quad \text{and} \quad B = \frac{Ga}{\sigma_0}\frac{d\sigma}{dT} \quad (4)$$

Under the initial condition $\xi = 0$ at $\tau = 0$, the solution is

$$\frac{1}{2B^3}\left[2B^2\xi + AB^2\xi^2 - 2AB\xi + 2(A - B)\ln(1 + B\xi)\right] = \tau. \quad (5)$$

This expression can be analyzed in the asymptotic limit $B\xi \ll 1$, by expanding the logarithm, resulting in

$$\xi \simeq \sqrt{2\tau} - \frac{2(A - B)}{3 + 2\sqrt{2\tau}(A - B)}\tau. \quad (6)$$

This result shows that the Washburn law ($\xi \simeq \sqrt{2\tau}$) is obtained immediately if the temperature gradient vanishes, making both A and B equal to zero. The same law arises when $A = B$, where both effects identically compensate. The relative changes in the evolution of the imbibition front

under temperature gradients with respect to the isothermal Washburn law will be determined based on the relative values of parameters A and B. These aspects will be discussed later on by analyzing a particular case. Our treatment considers the cases of positive or negative gradients and two isothermal cases with different temperatures.

Figure 2 shows the steady-state temperature field experimentally obtained with an infrared camera, from a cylindrical capillary glass, under a temperature gradient $G = -1250$ K/m. The image is shown as visual evidence of the existence of an actual constant temperature gradient along the capillary. After the gradient was imposed, it was necessary to estimate the changes in imbibition rate under temperature gradients with respect to the isothermal case. Here, we have considered a substance called squalene,[6] an organic oil with the chemical formula $C_{30}H_{50}$. The selection of this fluid is also based on the notable changes in surface tension and viscosity reported for this substance at wide temperature intervals.[6] Squalene has thermal diffusivity $\alpha_f = 10^{-7}$ m²/s. If it penetrates into a capillary of radius $a \simeq 2.5 \times 10^{-4}$ m, its diffusive time is $t_D = 0.5$ s, which is much smaller than the imbibition time which at a similar characteristic distance is approximately around 10 s.

Figures 3 and 4 show the plots of surface tension and dynamic viscosity, respectively, as a function of T in the range of 288.15 K $\leq T \leq 303.15$ K. These data were taken
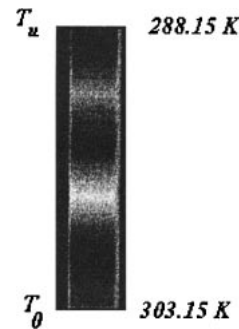

Fig. 2.   Isotherms in a cylindrical capillary of radius a under a temperature difference $\Delta T = (T_u - T_0) = -15$ K along H. In this case, we have a negative gradient because the lower tip is warmer than the upper one.
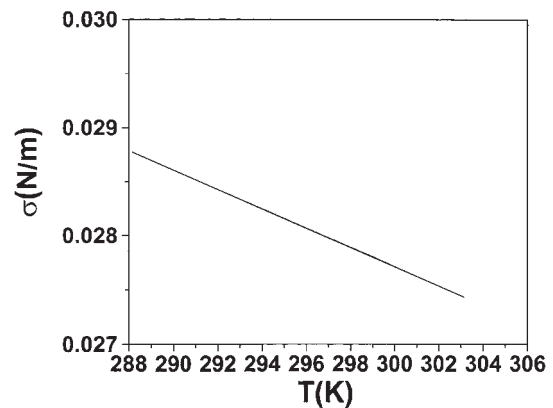

Fig. 3.   Squalene surface tension as a function of temperature for a temperature interval between 288.15 and 303.15 K. Note, in this case, the approximately linear behavior of $\sigma(T)$.
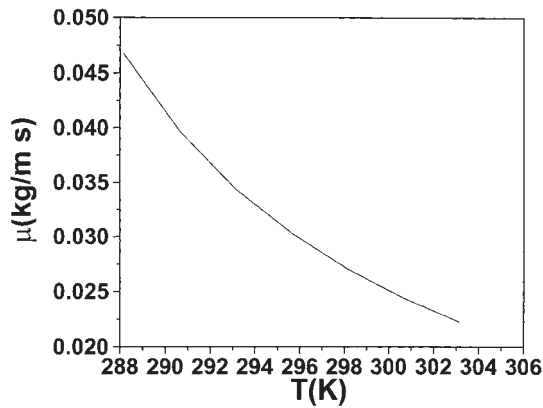
Fig. 4.   Variation in dynamic viscosity $\mu(T)$ as a function of temperature for the same interval and substance used in Fig. 3.

from Ludviksson *et al.*[6] In both plots, we notice that both quantities decrease as temperature increases. Given the way in which σ and μ evolve when T changes, the determination of the dimensionless gradients A and B is straightforward. In Fig. 5, we have plotted these dimensionless quantities for $G > 0$, $G < 0$ and $G = 0$. It is important to note that the graphs of A are not symmetrical due to the difference in reference μ_0 between cases $G > 0$ and $G < 0$. The plots for the evolution of the imbibition fronts in a capillary with the aforementioned radius are shown in dimensionless form for several temperature gradients in Fig. 6. In fact, the dimensionless plots in Fig. 6 correspond to a) isothermal imbibition for $T = 303.15$ K (thin continuous line), b) isothermal imbibition for $T = 288.15$ K (bold continuous line), c) imbibition under a positive gradient $G = 1250$ K/m when the lower tip is at 288.15 K (dashed line) and d) imbibition under a negative gradient $G = -1250$ K/m when the lower tip is at $T_0 = 303.15$ K (dashed curve), as in Fig. 2.

It should be noted that the imbibition at $T = 303.15$ K is faster than that at $T = 288.15$ K. Clearly, this is due to the low viscosity of the fluid at high temperature and to the very slow change in surface tension at this interval. Moreover, we graphically note that when the gradient is positive, the imbibition is similar to that occurring for case a). This indicates that a high viscosity exerts a strong damping effect on the flow in addition to the weak driven capillary force. Conversely, when the gradient is negative, i.e., when the fluid goes from the warmer region to the cooler, the imbibition is faster.

The behavior observed here for the imbibition under a negative gradient is, in some sense, similar to that observed for upward flows, which also occur under negative gradients. However, theoretical studies for upward flows[6–9] do not take into account variations in viscosity with temperature. Our study in this sense provides some idea about the relative importance of this quantity in relation to the properties of such types of flow.

Recent experiments on real rocks have shown that imbibition (and consequently oil recovery) is effectively accelerated when the temperature of a porous sample is increased.[5] This study does not present any theoretical models for explaining this behavior. This latter case and the ones where temperature gradients are imposed can be treated with the present model by determining the mean size of
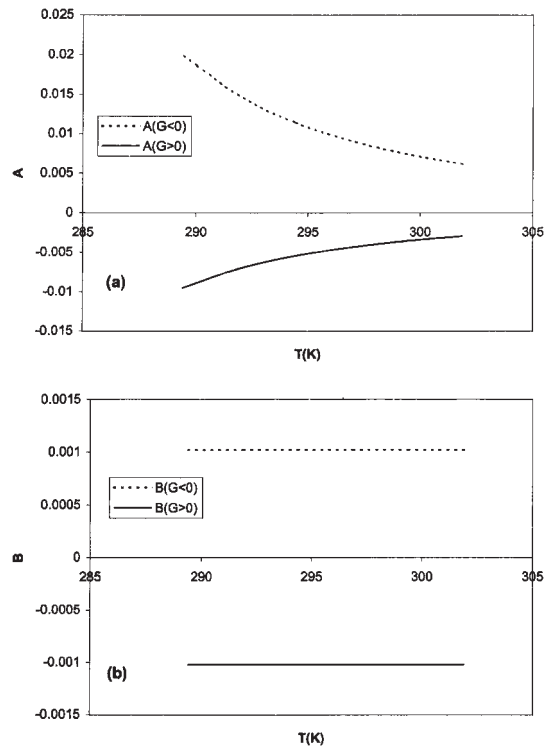

Fig. 5.   (a) Plot of the dimensionless parameter A as a function of T for cases of $G < 0$ and $G > 0$ for squalene and gradient given in the text. (b) Plot of the dimensionless parameter B as a function of T for the same cases referred to in (a).
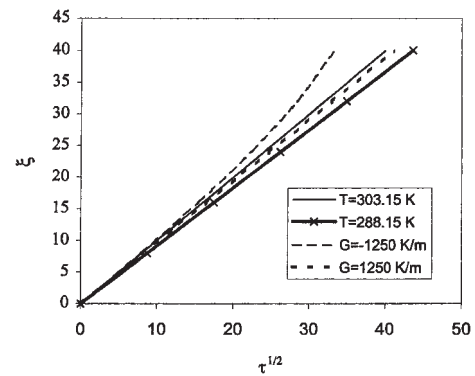

Fig. 6.   Dimensionless time evolution of the imbibition fronts for cases discussed in text with constant temperature and with constant positive ($G = 1250$ K/m) or negative gradients ($G = -1250$ K/m) in a single cylindrical capillary. Notice that imbibition is faster at $T = 303.15$ K (thin line) than at $T = 288.15$ K (bold line). Moreover, a positive gradient slows imbibition (dashed line) while a negative gradient accelerates imbibition (upper dashed curve).

hydraulic pores and replacing it with the radius of capillary, a.

In this letter, we have reported how temperature gradient affects the front dynamics within a cylindrical capillary. Using a balance of force equation, we have evaluated the relative importance of temperature gradient, surface tension and dynamic viscosity in fluid motion. For squalene, we found that a negative gradient accelerates flow during imbibition, due to the low viscosity specific for these types of changes in temperature. In particular, the data used for this substance has permitted us to observe only noticeable changes for the dimensionless gradient A and almost a

constant value for gradient B in the indicated temperature range. Through these quantities, we have shown that the acceleration/deceleration of imbibition depends strongly on the type of fluid and the size of the capillary. Future experimental investigation will be very useful to prove our predictions. Work along this line is now in progress.

Acknowledgments

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