Nano-channels filling flow of arbitrary cross-sections

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The filling kinetics of liquids in straight nano-channels of arbitrary cross-sections is discussed. We assume that the capillary force induces a fluid flow in channels characterized by their compactness, $C = P^2/A$, where P is the perimeter and A is the cross-sectional area. Analytical expressions for the distance between the capillary meniscus and the capillary inlet, l, as a function of the elapsed time, t, are given for several typical cross-sections. The comparison between our theoretical predictions and a set of reported data for a complex nano-channel [A. Han, J. Colloid Interface. Sci. 293 (2006) 151] allows us to conclude that the model describes well the filling kinetics.

Keywords: Micro and nano- scale flow phenomena; capillary effects; flow in channels.

Se discute la cinética del llenado de líquidos en nano canales de sección transversal arbitraria. Suponemos que la fuerza capilar induce un flujo de fluido en canales caracterizados por su compacidad, $C = P^2/A$, donde P es el perímetro y A es el área de la sección transversal. Expresiones analíticas para la distancia entre el menisco y la entrada del capilar, l, como una función del tiempo transcurrido, t, son dadas para varias secciones tranversales típicas. La comparación entre nuestras predicciones teóricas y un conjunto de datos reportados para un canal complejo [A. Han, J. Colloid Interface Sci. 293 (2006) 151] nos permite concluir que el modelo describe bien la cinética del llenado.

Descriptores: Fenómenos de flujo a micro y nano escala; efectos capilares; flujo en canales.

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1. Introduction

In recent years the study of the capillary penetration of liquids into narrow capillaries becomes a renewed topic of research in science because a fine knowledge of the capillary flow in micro and nano channels under complex operation conditions is critical in microfluidic of biology, medicine and technology, among others. For instance, filling flow driven by surface tension is used to make micro and nano structures by micromoulding and nanomoulding in capillaries (MIMIC and NAMIC) and very complex microstructures can be formed under these procedures [1-5]. Micromoulding and nanomoulding in capillaries are techniques capable of generating microstructures of polymers, inorganic salts, and sol–gel materials on substrates of completely different materials.

To our knowledge, the quasi-steady capillary flow developed in cilindrical capillary tubes, was described a century ago assuming a Poiseuille flow [6-8]. It means that the flow velocity, u, obeys the formula $u \propto \left(R^2/\mu\right)(\Delta p/L)$ where R is the inner radius of the tube, μ is dynamic viscosity of the liquid and Δp is the pressure drop along the length L. Such a flow obeys the conditions postulated in the Poiseuille's law, these are: an incompresible newtonian flow, occurring at a low Reynolds number, through a distance that is substantially longer than its diameter. Typically, in the Poiseuille flow, and consequently in the description of the capillary penetration, when the cross-section of the tube is not circular, the inner radius is substituted with an *ad hoc* hydraulic radius that takes into account the geometry of the channel [1-4]. Models using this approximation give very accurate results for non angular tubes [9] but commonly fail for angulated capillaries [1]. In the present work we use an alternative approach where the use of the hydraulic radius is explicitly avoided.

The aim of this work is to present a theoretical study of the capillary penetration into single, straight channels when non circular cross-sections are assumed. Specifically, we will analyze the capillary flow in straight cylinders of elliptical and rectangular cross-sections which are of interest and utility in many applications. Our start point is the formulation of a motion equation which takes into account the frictional force as a function of the friction factor α, when a constant, initial pressure, p_{in} is present. As will be seen later this term is important because channels of different cross sections have different frictional factors. In particular, we show that in horizontal, angulated channels it is possible to found simple analytical correlations for the front of penetration of liquid, l, as a function of the time elapsed during the penetration, t. Such correlations involve the friction factor α as a linear function of the compactness, $C = P^2/A$, where P is the inner perimeter of the channel and A is its cross-sectional area. Compactness is a fundamental concept used in the area of pattern recognition which is a method very useful in industrial applications to characterize geometrically an object.

By the way, the study of the capillary penetration in micro and nano-channels is receiving now increasing attention due to its relevance to fundamental questions regarding fluid behavior at spatial scales where surface forces dominate over the body forces. The analysis of phenomena in this limit is essential to extend our knowledge about this type of fluid flows [10].

To reach all these goals in next Section we derive the motion equation for the capillary penetration in straight channels, after, in Sec. 3 we formulate the Poiseuille flow for straight channels of arbitrary cross-sections in a dimensionless form, it let us to define the friction factor, α, to compute the linear relation between α and the compactness C and the dependence of l on $\alpha(C)$. Using this latter formula, in Sec. 4 we make a comparison between our model and that related to the use of the hydraulic radius, for an actual flow developed in an irregular nano-channel [4], in order to show the goodness of our method. Finally, in Sec. 5 we give the main conclusions of this work.

2. Capillary penetration in capillary channels

The main assumption in our treatment is that a Poiseuille flow occurs in horizontal capillary channels during the capillary penetration because in this type of problems the Reynolds number is low [11]. So, the resulting motion equation for the capillary penetration, where an initial pressure p_{in} is present, has the form [12,13]

$$Ap_{in} + f_\sigma - f_\mu = 0, \tag{1}$$

here f_σ is the capillary force, and f_μ is the frictional viscous force. The explicit form of each term is given by

$$f_\sigma = P\sigma\cos\theta, \qquad f_\mu = \alpha\mu v l, \tag{2}$$

where σ is the surface tension, θ is the angle of contact, α is the dimensionless hydraulic resistance term, v is the mean velocity of the flow and l is the position of the fluid front. The inclusion of the capillary force in the form

$$f_\sigma = P\sigma\cos\theta \tag{3}$$

has been used successfully by several authors for simple geometries (cross-sections) [12,13]. The substitution of each term yields the differential equation for the front position, l,

$$Ap_{in} + P\sigma\cos\theta - \alpha\mu l\frac{dl}{dt} = 0. \tag{4}$$

Now, if we use the compactness dimensionless parameter [14]

$$C = \frac{P^2}{A}, \tag{5}$$

which is a way to characterize the cross-sectional area, the solution of Eq. (4) has the form

$$l = \sqrt{\frac{2\left(\frac{P^2 p_{in}}{C} + P\sigma\cos\theta\right)}{\alpha\mu}t}, \tag{6}$$

where, again, it is explicitly introduced the capillary cross-section through the explicit dependence on C and α. Equation (6) is a generalization of the well-known Washburn law which is valid for circular channels [8]. Moreover, for more complex cross-sections, recently has been found a relationship between α and C [15] which will be discussed in the next section.

3. Fluid flow in non circular cross-sections

It is obvious that the hydraulic resistance term α should be different for different geometries. The basis to build the explicit dependence of $\alpha(C)$ is that the flow developed during the capillary penetration is a Poiseuille flow, *i.e.*, a one-dimensional flow. Under these circumstances it is possible to introduce the dimensionless hydraulic resistance as

$$\alpha \equiv \frac{R_{hid}}{R^*_{hid}}, \tag{7}$$

by definition, the hydraulic resistance R_{hid} is given by

$$R_{hid} = \frac{\Delta p}{Q}, \tag{8}$$

in this expression Δp is the pressure drop along the channel and Q is the volume flow rate of fluid in the channel. Additionally, through a dimensional analysis one found that the hydraulic resistance should be a quantity of the form

$$R^*_{hid} = \frac{\mu L}{A^2}, \tag{9}$$

where L is the distance penetrated by the liquid and A, in a general way, is the cross-sectional area of the channel

$$A = \int_\Omega dxdy, \tag{10}$$

where Ω is the inner contour of the channel. Thus, using (8) and (9) in Eq. (7), we found that

$$\alpha = \frac{A^2}{(\mu L/\Delta p)\,Q}. \tag{11}$$

Due to the flow in the channel obeys a Poiseuille flow whose velocity is

$$\mathbf{v} = u(x,y)\mathbf{e}_z, \tag{12}$$

where $\mathbf{e}_z$ is the unit vector along the flow direction, we have that the motion equation is

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)u(x,y) = -\frac{\Delta p}{\mu L}. \tag{13}$$

The knowledge of the velocity field $u(x,y)$ in the solution of Eq. (13) allows us to give the hydraulic resistance in the form

$$\alpha = \frac{A^2}{\left(\frac{\mu L}{\Delta p}\right)\int_\Omega u(x,y)dxdy}. \tag{14}$$

The introduction of the non dimensional variables $\xi{=}x/D$, $\eta = y/D$ and $v = u/u_c$, where D is a characteristic size of the cross-section, yields

$$u_c = \frac{D^2}{\mu}\frac{\Delta p}{L} \tag{15}$$

and the Poiseuille Eq. (13) is transformed in

$$\frac{\partial^2 v}{\partial \xi^2} + \frac{\partial^2 v}{\partial \eta^2} = -1. \tag{16}$$

Finally, the friction factor is

$$\alpha = \frac{\left(\int\limits_{\Omega^*} d\xi d\eta\right)^2}{\int\limits_{\Omega^*} v d\xi d\eta}\,. \tag{17}$$

In this later expression Ω^* is the dimensionless contour, the integrals involve only dimensionless parameters and consequently the friction factor depends only on the cross-section shape but does not depend on the size of the channel. In the next section we will give the corresponding values of α for several cross-sections.

3.1. Circular channel

In this case $D = R$, where R is the radius of the circular channel, thus $\xi = x/R$, $\eta = y/R$ and we found that for this case the dimensionless velocity resulting of the solution of Eq. (16) is

$$v = \frac{1}{4}\left(1 - \zeta^2\right), \tag{18}$$

where $\zeta = \sqrt{\xi^2 + \eta^2}$ is the dimensionless radial coordinate and the corresponding friction factor derived from Eqs. (17) and (18) is

$$\alpha = \frac{(\pi)^2}{\frac{\pi}{8}} = 8\pi. \tag{19}$$

Finally, from Eq. (5) it is easy to evaluate the compactness, such evaluation yields

$$C = \frac{(2\pi R)^2}{\pi R^2} = 4\pi, \tag{20}$$

however in this case α and C are not explicitly related, *i.e.*, C is a constant.

3.2. Elliptic channel

For elliptic channels, of major semi-axis a and minor semi-axis b, is possible found the velocity from Eq. (16), in the form

$$v = \frac{\epsilon^2}{2\left(1+\epsilon^2\right)}\left[1 - \xi^2 - \left(\frac{\eta}{\epsilon}\right)^2\right], \tag{21}$$

here $D = a$, $\xi = x/a$, $\eta = y/b$ and $\epsilon = b/a \le 1$. From Eq. (17) and the velocity field (21), the friction coefficient now is

$$\alpha = \frac{\pi^2\epsilon^2}{\frac{\pi\epsilon^3}{4(1+\epsilon^2)}} = 4\pi\left(\epsilon + \frac{1}{\epsilon}\right), \tag{22}$$

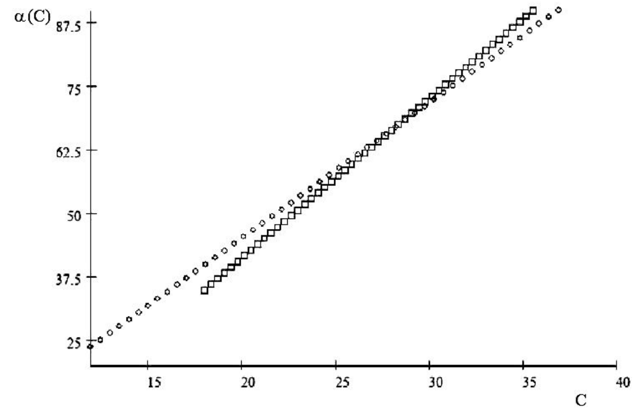

FIGURE 1. Plot of the friction factor α as a function of the compactness C for elliptic (◯) and rectangular (□) cross-sections.

or using

$$\gamma = \frac{a}{b} = \frac{1}{(b/a)} = 1/\epsilon, \tag{23}$$

the new form of α is given by

$$\alpha(\gamma) = 4\pi(\gamma + \gamma^{-1}). \tag{24}$$

The compactness involves elliptic integrals whose simplest form is

$$C(\gamma) = \frac{16}{\pi}\gamma\left(\int\limits_0^{\pi/2} d\theta\sqrt{1 - (1-\gamma^{-2})\sin^2\theta}\right)^2. \tag{25}$$

The relationship between α and C, $C(\alpha)$, or conversely, $\alpha(C)$ is obtained from the substitution of $\gamma(\alpha)$ obtained from Eq. (24) and introducing it in Eq. (25). It yields [15]

$$C(\alpha) = \frac{1}{2\pi^2}\left(\int\limits_0^{\pi} d\theta\sqrt{\alpha - \sqrt{\alpha^2 - (8\pi)^2}\cos\theta}\right)^2. \tag{26}$$

The function $C(\alpha)$ is obtained by expanding $C(\alpha)$ (Eq. (26)) around $\alpha = 8\pi$, and after, solving for α as a function of C, *i.e.*,

$$\alpha(C) = \frac{8}{3}C - \frac{8}{3}\pi + O\left([C - 4\pi]^2\right). \tag{27}$$

In Fig. 1 we plot this linear function $C(\alpha)$ for elliptical cross-sections (symbol ◯). In this same plot we also show the plot for rectangular cross-sections (symbol □). The introduction of Eq. (27) in Eq. (6) allows us to give an expression for the front of the capillary penetration l, as a function of time t,

$$l = \sqrt{\frac{3\left(P^2 p_{in} + PC\sigma\cos\theta\right)}{4\left(C^2 - \pi C\right)\mu}t}. \tag{28}$$

This equation is the Washburn law for an elliptic channel. In a subsequent section we will applied this result in the analysis of the filling flow of a nano-channel.

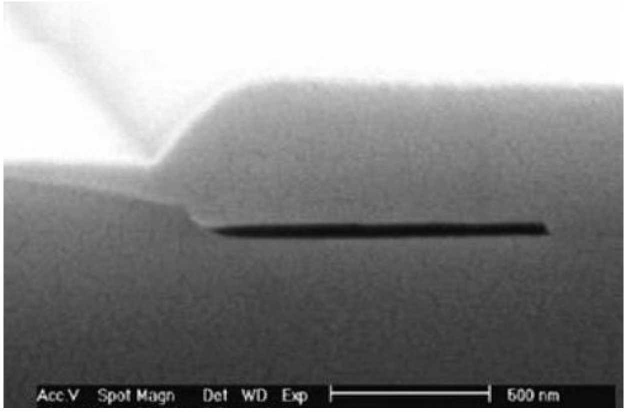

FIGURE 2. Scanning by SEM of the cross-section of a nano-channel [4]. The black zone at the middle part has a size w=900 nm and height $h = 50$ nm.

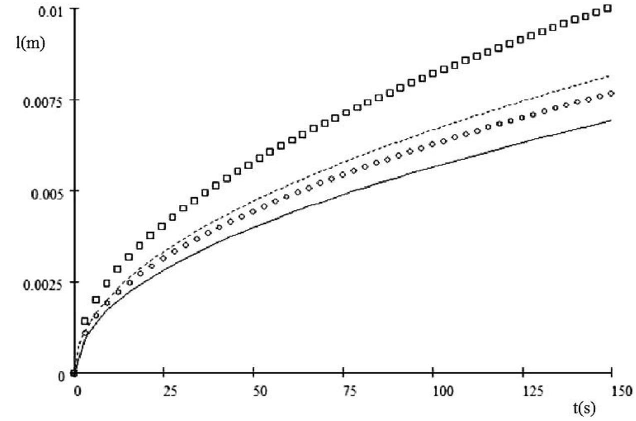

FIGURE 3. Plot of l vs t: continuous line corresponds to the Washburn law using the hydraulic radius R_H (Eq. (32)); □ experimental data from Han *et al* [4]; ◯ elliptic cross-section using Eq. (28) and dashed line rectangular nano-channel with $w = 900$ nm and height $h = 50$ nm Eq. (31).

3.3. Rectangular channels

If the channel now has a rectangular cross-section of height $2w$ and width $2h$ with $w \neq h$, the aspect ratio is $\gamma = w/h \geq 1$. Obviously, $P = 2(w + h)$, $A = wh$ and the solution of Eq. (16), *i.e.*, the velocity, is [15]

$$v = \frac{4}{\pi^3} \sum_{n=1,3,5,...}^{\infty} \frac{1}{n^3} \left[1 - \frac{\cosh\left(\frac{n\pi\xi}{2}\right)}{\cosh\left(\frac{n\pi\gamma}{2}\right)} \right] \sin\left(n\pi\eta\right), \quad (29)$$

in this relationship $D = h$, $\xi = x/h$, $\eta = y/h$ and the friction factor α now is [15]

$$\alpha(C) \approx \frac{22}{7}C - \frac{65}{3} + O\left([C - 18]^2\right). \quad (30)$$

The plot of this later equation is given also in Fig. 1 (symbol □). The corresponding Washburn law for rectangular channels under an initial pressure p_{in} is derived simply by using Eq. (30) in Eq. (6), it yields

$$l = \sqrt{\frac{42\left(P^2 p_{in} + PC\sigma\cos\theta\right)}{\left(66C^2 - 455C\right)\mu} t}. \quad (31)$$

Notice the substantial algebraic difference between this last equation and that derived for elliptic channels (Eq. (28)).

4. A comparison with a nano-channel flow

Han *et al.* [4] have studied experimentally the kinetics of the capillary penetration of various liquids that fill nano-channels. Although, as can be seen in Fig. 2, the cross-section of a typical nano-channel is very complex, Han *et al.* have assumed that such channel has a rectangular cross-section along its axis. Here we are interested in their results reported for filling flow with deionized water. In such experiments the position of the liquid meniscus was followed and its evolution was found as proportional to the square root of time, $l \sim \sqrt{t}$, which is the Washburn law [8,11]. The width of the channel was $w = 900$ nm and the mean height $h = 50$ nm. For water, the surface tension is $\sigma = 0.072$ N/m, the dynamic viscosity $\mu = 1$ centipoise and the contact angle $\theta = 68°$ and in the experiments there is no an initial pressure, *i.e.*, $p_{in} = 0$.

In Fig. 3 we plot several curves to describe the evolution of the front of water, l, as a function of time, t. The curve made with symbols □ corresponds to the experimental data, the dashed line was made by assuming that the capillary is effectively a rectangular channel of width $w = 900$ nm and height $h = 50$ nm and compactness $C = 80.22$. In this case we used the Eq. (31) to determine such a evolution. The plot made with symbols ◯ was made by assuming that the cross-section of the nano-channel was an ellipse of semi-axis $a = w/2 = 500$ nm, $b = h/2 = 25$ nm and compactness $C = 114.75$ (Eq. (28)). Finally, the continuous line corresponds to the Washburn law given for a rectangular channel, this law goes like

$$l = \sqrt{\frac{R_H \sigma \cos\theta}{2\mu} t}, \quad (32)$$

where $R_H = A/P = 2(w + h)/wh$ is the hydraulic radius.

It is clear that the better approximation to the experimental data is that given by the dashed line which corresponds to the assumption that the cross-section of the channel is a rectangle. The worst approximation is that given by the Washburn law based on the hydraulic radius but it is comprehensible because it is a very simple correlation. Surprisingly, in this case the flow in an elliptic nano-channel is more resistive than the flow in the rectangular channel with similar dimensions, the reason for this behavior is the very high value of the compactness for the elliptic channel.

5. Conclusions

Through the use of a one-dimensional motion equation, that includes the friction factor $\alpha(C)$, has been modeled the capillary penetration into channels of complex cross-sections. As a particular case, this model was used to describe the capillary penetration into nano-channels. Our calculations in a narrow nano-channel show that actually the capillary penetration is modulated by the competition between friction and the capillary driven force and both of them are critically dependent on the cross-section or, equivalently, on compactness. The comparison between the experimental data and our model is good and better than the one that uses the hydraulic radius, however there is a significant difference between the actual data and the results of the model. This difference could be also originated by the manner as the material surfaces of the nano-channels were prepared because the contact angle is very sensitive to changes in the properties of the materials. Other possible error involved in such a difference could be due to the corners of the channels because there the flow can be very slow due the shear stresses which compete with the capillary force. More studies along this line are now conducted.

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