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Imbibition in pieces of paper with different shapes

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We study theoretically and experimentally the wicking process in pieces of blotting paper of different shapes: rectangular and triangular strips and square strips where radial imbibition occurs. We find that in rectangular strips the dynamics of the capillary penetration yields the classical Washburn law while in the other cases logarithmic corrections respect to this law are obtained. Our experiments are in good agreement with the theoretical predictions.

Keywords: Surface-tension-driven instability; flow through porous media

Estudiamos teórica y experimentalmente el proceso de absorción capilar de fluidos en pedazos de palpel filtro de diferente forma: tiras rectangulares y triangulares, y tiras cuadradas donde, ocurre imbibición radial. Encontramos que en las tiras rectangulares la dinámica de la penetración capilar da la ley clásica de Washburn, mientras que en los otros casos se obtienen correcciones logarítmicas respecto a dicha ley. nuestros experimentos dan un buen acuerdo con las predicciones teóricas.

Descriptores: Inestabilidades originadas por tensión superficial; flujos en medios porosos

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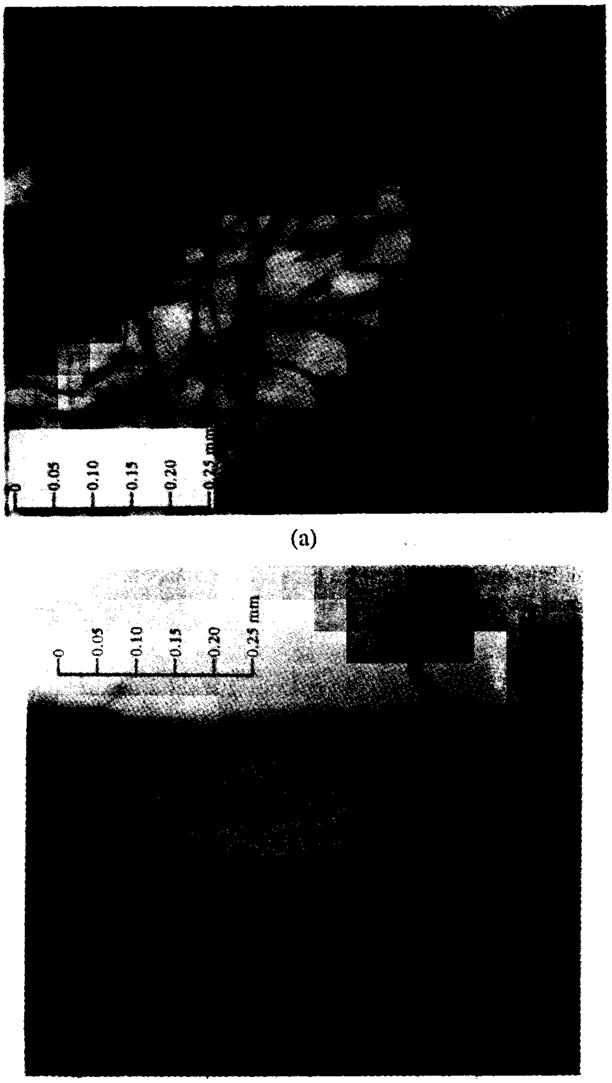
1. Introduction

Capillary penetration of liquids into fibrous materials, like paper or fabric, is of great importance in fundamental and applied physics [1–8]. In the first case, the theoretical and experimental studies show a complex interaction between the liquid and the isotropic [1–6] or the anisotropic [7,8] fibrous matrix. In the second case, there are many processes on these materials, such as textile and paper treatment, chromatography, drying ink, etc., where the knowledge of the fluid penetration under several physical and chemical conditions is critical [9,10]. Moreover, many of the main results of this phenomenon in fibrous materials, also known as imbibition, can be extended to granular porous media, like sand and rocks, and consequently to a very wide range of other areas such as underground water and petroleum exploitation [11–13].

From the experimental point of view, imbibition has been mainly studied in filter or blotting papers where various methods to measure this spontaneous liquid absorption have been proposed. With these methods, the liquid transport into this materials is monitored by physical changes, such as weight, transmittance, visualization, etc. These methods can provide information only on the absorption kinetics and two types of geometries have been used to study this process: unidirectional penetration, from a large fluid reservoir, in rectangular strips and radial penetration from the finite perimeter of a circle of radius r_0 into the fibrous assembly [1,3,4]. The results obtained from these two types of systems can be compared and the main difference is a logarithmic correction in the radial imbibition respect to the unidirectional case. Unidirectional penetration of liquid into rectangular strips clearly shows that the kinetics of the process follows the Washburn law, which is a relation between the distance from the fluid source to the imbibition front, y, and the elapsed time to reach this front, t, such that $y \sim \sqrt{t}$ [14] and where the constant of proportionality is a function of the properties of the material. The radial imbibition follows, as an asymptotic limit for small distances, the Washburn law [3,4]. In this work we will study the imbibition in triangular samples of paper, as a fluid dynamics problem in a porous medium and not as an extension of the fluid motion into parallel plates or into many capillary tubes and our results are compared with the well studied cases of imbibition in rectangular strips and radial imbibition. We found that in triangular shapes logarithmic corrections are also obtained having the Washburn law as an asymptotic limit for small distances. Experiments with water as the wetting fluid show a good agreement with the theoretical predictions for all the geometries here treated. This work is divided as follows: in next section we will introduce our theoretical approach to study the triangular geometries and we also study the unidirectional and radial penetration. After that, in section III we will present experimental studies and the comparison with the theoretical predictions. Finally in section IV we give the main conclusions and perspectives of this work.

2. Theoretical treatment

Despite the complex structure of paper (see Fig. 1), the spontaneous and uniform penetration of a fluid into paper of any shape, when it is horizontal, can be described with the fluid dynamics equations, *i.e.*, through the Darcy equation, the



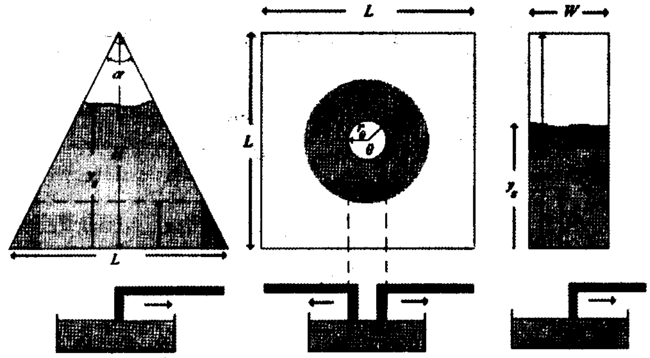
(a)

(b)

FIGURE 1. a) Photograph showing the surface details of the blotting paper used in our experiments. Note the complex structure of this isotropic sample and b) lateral view of the blotting paper where we can see its average thickness, $D \simeq 0.38$ mm. Both photographs were taken with a $10\times$ microscope.

mass conservation equation and the Young-Laplace equation for the capillary pressure difference at the imbibition front in the material. Incidentally, when the fluid penetration is not uniform (there is a complex saturation in the paper), for instance in samples of vertical paper strips, the treatment is more complex because the permeability is a function of the saturation itself [2]. This case will not be considered in this work.

The problem of the unidirectional imbibition in triangular strips of blotting paper was previously studied experimentally by measuring the time evolution of the imbibition front [15]. This last study clearly showed that the evolution front obeys a different behavior than that valid for rectangular strips. However, no theoretical treatment was developed. Therefore, in order to give a better understanding of this problem in the following lines we study the case of imbibition from the basis



(a) (b) (c)

FIGURE 2. Schematics of the horizontal capillary penetration of a liquid into: a) triangular piece of paper of upper angle α, b) radial penetration from the initial perimeter $r = r_0$, and c) unidirectional penetration in a strip of length L. In all cases the paper has thickness D and height H and the fluid penetrates as is shown in the depicts.

of triangles. Experimentally it is noted that the imbibition front is nearly flat (parallel to the basis where the fluid penetrates), *i.e.*, unidirectional. This observation will be very useful in order to build a model for the evolution of this front and the corresponding velocity. Afterwards, we will treat the other geometries.

2.1. Triangular strip

In order to analyze the imbibition in the triangular pieces of paper, we consider pieces of length L, height H, thickness D and upper angle α, see Fig. 2a. The characteristic size of the pores is d and the surface tension of the fluid is σ. In Fig. 2a we also show the imbibition front which is flat and parallel to the basis. Because the paper is very thin, the problem will be treated in two dimensions, the gravity field will not be considered. In the first place, the capillary penetration into the paper is due to the capillary pressure difference Δp originated by the surface tension. In this case the Young-Laplace equation is

$$\frac{c_1 \sigma}{d} = -\Delta p, \tag{1}$$

where c_1 is a dimensionless constant related to the contact angle, θ, between the fibres of the porous material and the liquid. Assuming that the steady-state flow in the paper is unidirectional and that the filtration velocity is v, the Darcy equation has the form

$$\frac{dp}{dy} = -c_2 \mu \frac{v}{d^2}, \tag{2}$$

where c_2 is another dimensionless constant. The mass conservation is given by the relation

$$Av = A_s \frac{dy_s}{dt}. \tag{3}$$

where A and A_s are the transversal areas at heights y (within the wet zone) and y_s (just the imbibition front), respectively.

By combining Eqs. (2) and (3) we obtain that

$$\frac{dp}{dy} = -\mu c_2 \frac{A_s}{Ad^2} \frac{dy_s}{dt}, \tag{4}$$

and the integration of Eq. (4) gives the total pressure drop over the imbibed zone. This quantity is

$$\begin{aligned}\Delta p &= -\frac{\mu c_2}{d^2} \frac{dy_s}{dt} \int_0^{y_s} \frac{A_s}{A}\, dy \\ &= \frac{\mu c_2}{d^2}(H - y_s)\frac{dy_s}{dt} \ln\left(\frac{H - y_s}{H}\right).\end{aligned} \tag{5}$$

Here we have used the ratio

$$\frac{A_s}{A} = \frac{H - y_s}{H - y}, \tag{6}$$

which is a consequence of the uniform thickness of the sample and the flat imbibition front in the triangles. Now, by using Eq. (5) in Eq. (1), we find the differential equation

$$\frac{c_1 \sigma d}{\mu c_2} = -(H - y_s)\frac{dy_s}{dt} \ln\left(\frac{H - y_s}{H}\right), \tag{7}$$

this can be written in a simpler form through the dimensionless variable, $\xi = (H - y_s)/H$. So, we have

$$\frac{c\sigma d}{\mu H^2} = -\xi \ln \xi \frac{d\xi}{dt} = -\frac{1}{4} \ln \xi^2 \frac{d\xi^2}{dt}, \tag{8}$$

where we have made $c = c_1/c_2$, clearly this quantity will determined from experiments. The solution of Eq. (8), with the boundary condition $\xi = 1$ at $t = 0$, is

$$\xi^2 \left[\ln \xi^2 - 1\right] + 1 = \frac{4c\sigma d}{\mu H^2} t = \tau, \tag{9}$$

where $\tau = t/(\mu H^2/4c\sigma d)$ is the dimensionless time.

For small dimensionless heights we assume that $\xi = 1 - \varepsilon$, where $\varepsilon = y_s/H$, and by using $\ln(1-\varepsilon) = -\varepsilon - (1/2)\varepsilon^2 - O(\varepsilon^3)$, we obtain

$$\varepsilon^2 = \frac{2c\sigma d}{\mu H^2} t, \tag{10}$$

or

$$y_s^2 = \frac{2c\sigma d}{\mu} t. \tag{11}$$

This last relation is the well known Washburn law [14], and is valid for imbibition in strips of homogeneous porous media with constant transversal area. So, at the initial stage of the imbibition the front obeys the same behavior than that in strips with constant transversal area (as will be illustrated later on). Plots of the Washburn law and of Eq. (8), for several triangles (α's), are shown in Fig. 3a. In this case the most important result, which can be seen in the curves corresponding to the front evolution of triangles is the smooth change in slope (change in the front velocity), at a certain time (or at a certain height). In Fig. 3b we have plotted the front velocities (curves) for the cases here considered. There, we note easily

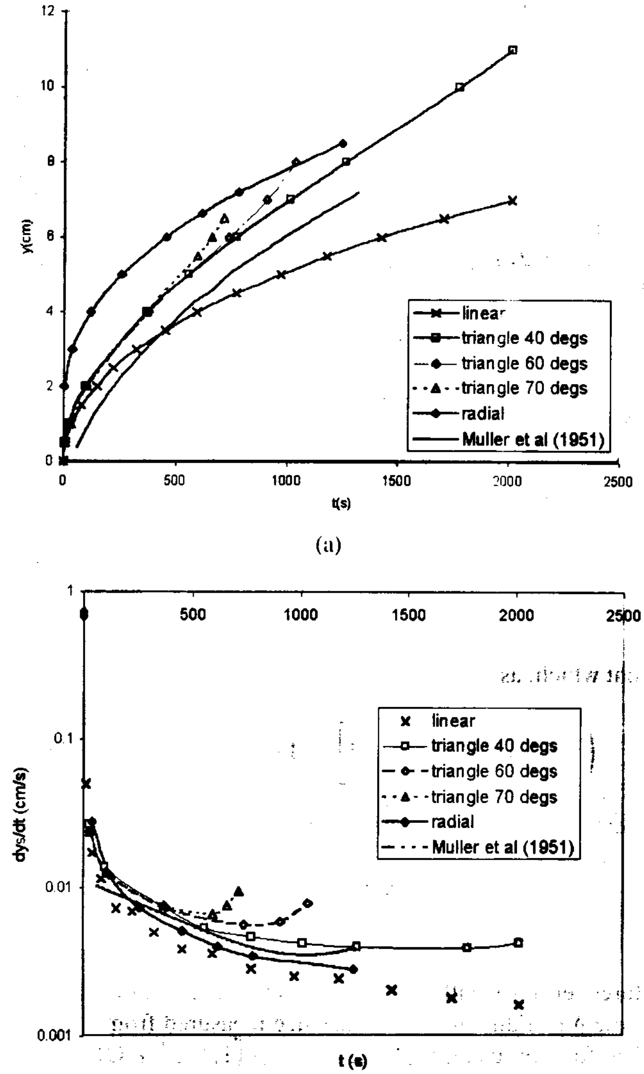


FIGURE 3. a) Evolution of the imbibition front as a function of time for: triangular shapes with angles $\alpha = 40°$ (□), 60° (◇) and 70° (△), for radial imbibition with $r_0 = 1.5$ cm (♦) and for unidirectional imbibition in rectangular strips with $L = 5$ and 10 cm and $H = 10$ cm (×), b) semilog plot of the velocity front, dy_s/dt, as a time function for the several geometries considered in a).

the increase of the front velocity (dy_s/dt) in the triangles. The evaluation of the time where the increase starts, t^*, can be obtained from Eq. (9). This time is

$$t^* = \frac{1 - 3/e^2}{\dfrac{4c\sigma d}{\mu H^2}} \tag{12}$$

and the height where this increase occurs is given by

$$\frac{y_s^*}{H} = 1 - \frac{1}{e} \simeq \frac{2}{3}. \tag{13}$$

To our knowledge, this result has not been predicted for radial imbibition or imbibition in rectangular strips and it is a consequence of the triangular shape of the sample.

2.2. Radial imbibition

In this case, the imbibition starts from a finite radius, $r = r_0(t = 0)$, and after the imbibition fronts are concentric circles of radius $r(t)$ (Fig. 2b). The fundamental equations are the same than those in the previous case and we will only change the areas ratio given by Eq. (6) in the previous subsection. Because the imbibition front, at the distance r_s, has the area $A_s = 2\pi r_s D$ we have now that $A_s/A = r_s/r$, where $A = 2\pi r D$ is an area in the wet zone such that $r_0 \leq r \leq r_s$. Therefore, in this case, the Darcy equation yields for the total pressure drop

$$\Delta p = -\frac{\mu c_2}{d^2}\frac{dr_s}{dt}\int_{r_0}^{r_s}\frac{A_s}{A}\,dr = -\frac{\mu c_2}{d^2} r_s \ln\frac{r_s}{r_0}\frac{dr_s}{dt}. \quad (14)$$

Introducing this result in the Eq. (1) we obtain

$$\frac{c\sigma d}{\mu} = r_s \ln\frac{r_s}{r_0}\frac{dr_s}{dt}, \quad (15)$$

and solving this differential equation we obtain the imbibition front which, as a time function, obeys the relation

$$\left(\frac{r_s}{r_0}\right)^2\left[\ln\left(\frac{r_s}{r_0}\right)^2 - 1\right] + 1 = \frac{4c\sigma d}{\mu r_0^2}t = \tau', \quad (16)$$

where τ' is the dimensionless time defined as

$$\tau' = \frac{t}{\frac{\mu r_0^2}{4c\sigma d}}.$$

Moreover, for small lengths we assume that $r_s/r_0 = 1 + \delta$ where δ is a dimensionless distance measured from $r = r_0$. Therefore, by using $\ln(1+\varepsilon) = \varepsilon - (1/2)\varepsilon^2 + O(\varepsilon^3)$, we obtain for small dimensionless distances that

$$\delta^2 = \frac{4c\sigma d}{3\mu r_0^2}t, \quad (17)$$

which corresponds once again to the Washburn law. The plots of the imbibition front and its velocity are shown in Fig. 3.

2.3. Linear imbibition

In Fig. 2c we show the schematics of the unidirectional imbibition in rectangular strips of blotting paper. Once again, the governing equations are the same as above and we again change the areas ratio which in this case is constant, $A_s/A = 1$. Then, the Darcy and mass conservation equations yield the total pressure drop

$$\Delta p = -\frac{\mu c_2}{d^2}\frac{dr_s}{dt}\int_0^{y_s} dy = -\frac{\mu c_2}{d^2} y_s \frac{dy_s}{dt}. \quad (18)$$

This result leads, as above, to the differential equation

$$\frac{c\sigma d}{\mu} = y_s \frac{dy_s}{dt}, \quad (19)$$

and, finally, by solving this differential equation, we find (taking into account that $y_s = 0$ at $t = 0$), the Washburn law

$$y_s^2 = \frac{2c\sigma d}{\mu}t, \quad (20)$$

this curve is shown in Fig. 3. In the next section we present some experiments in order to prove our theoretical results for all these geometries.

3. Experiments

Imbibition experiments in horizontal pieces of blotting paper, of mean thickness $D = 0.38$ mm, have been made in order to compare with the theoretical results for three geometries previously considered. In the first place, the triangular pieces of paper were cut so that the basis of the triangles always has $L = 10$ cm, its angles were $\alpha = 40°$, $60°$ and $70°$ and its corresponding heights were $H = 13.74$, 8.66 and 7.14 cm. Rectangular pieces were $L = 5$ and 10 cm in length and $H = 10$ cm in height. We have folded the basis of the triangles and rectangles as is depicted in Fig. 2a and 2c to induce the water filtration from a big container. Finally, the radial imbibition was carried out in paper with $L = 30$ cm and initial radius $r_0 = 1.5$ cm. In the radial imbibition we have brought the water to the perimeter of the horizontal sample of paper by using also a circular piece of the same blotting paper which touched the water reservoir and the perimeter of the paper (Fig. 2b). In all cases we used deionized water (at ambient temperature 24°C) which has a surface tension $\sigma \sim 72$ gr/s^2, density $\rho = 1$ gr/cm^3 and dynamic viscosity $\mu = 0.92$ cp. The front evolution (zone between the dry and wet zones) was recorded with a CCD video camera. After that, we have digitized the film, frame by frame, and we have measured the time evolution of the front, each $1/30$ s, with a spatial resolution of ± 1 mm. Typical duration of experiments in these samples was from 500 s up to 2500 s.

At least four experiments were performed for each geometry (and for each angle α). In Fig. 3 we show the experimental results (symbols) for each geometry. The basic experiment was the imbibition in the rectangular strips (where the experimental results are the same for $L = 5$ or $L = 10$ cm) because in this case we can evaluate easily the factor $A = 2c\sigma d/\mu$ by fitting Eq. (20) to the experimental points. So, the quantity A takes the value $A = 2.71 \times 10^{-2}$ cm^2/s. In this quantity we do not know *a priori* the *effective* value of $2cd$ which, as aforementioned, is related to the effective angle of wetting (θ) between the fibres and the liquid. In the rectangular strips our experiments indicate that $2cd = 0.003$ mm. By the way, it is generally accepted that $2cd \sim \cos\theta d$ and that in blotting paper $\theta \simeq 87°$ [4,6,8], therefore d will be $d \sim 0.05$ mm. Looking at Fig. 1a we note that it is a very good representative value for the pore size. Afterwards, we have estimated that $B_\alpha = 4c\sigma d/(\mu H_\alpha^2)$, for the triangle with angle α, and $C = 4c\sigma d/(3\mu r_0^2)$, for radial imbibition. We have obtained these values through the experiments and by using the value of $2cd$ obtained in the experiments of the rectangular strips. The obtained values for triangles by using both methods are very similar between them ($B_{40°} = 2.88 \times 10^{-4}$ s^{-1}, $B_{60°} = 7.24 \times 10^{-4}$ s^{-1} and $B_{70°} = 1.06 \times 10^{-3}$ s^{-1}) but C, in the radial imbibition, is underestimated (using the value of $2cd = 0.003$ mm) by an order of 2.5 times. So, the best value was $C = 6.43 \times 10^{-2}$ s^{-1}. This difference may be originated by the complex interaction between the fibres and the fluid where, for instance, the wetting angle changes as a function of the filtration velocity [5]. In Fig. 3a we show all the theoretical curves for the imbibition fronts which fitted the experimental data and yield the constants previously discussed. The agreement is very good. There we also have included the experimental data, for a triangle (continuous curve without symbols), from Muller *et al.* [15]. Likewise, in Fig. 3b we show the velocity fronts as a time function for all cases here treated including that by Muller *et al.* (again, the continuous curve without symbols). The main result in this case is the appreciable increase in the velocity just at $y_s^*/H \simeq 2/3$ for all the triangular shapes. So, the experiments give a clear confirmation of our model in relation to the triangular geometry.

4. Conclusions

In this work we have analyzed the spontaneous imbibition in pieces of paper with several simple shapes. We have found that the theoretical predictions are in good agreement with the experimental measurements. Experiments using water were made in order to determine the free parameters of the model. A new result was the prediction of the increase of the front velocities in the triangular geometries which was proved through the experiments. It is also important to note that similar equations for the evolution of the imbibition fronts were obtained for cases of imbibition in triangles and radial imbibition. More work, specially that related to more complicated geometries, to spontaneous penetration from several boundaries and to the use of three-dimensional samples of other porous media is in progress.

5. Acknowledgments

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