

Multiple interconnected capillary tubes model for description of advanced features of capillary flow in paper strips.

Emanuel Elizalde, Raul Urteaga, Thomas Duriez, and Claudio L.A. Berli

emalde@gmail.com; urteagar@santafe-conicet.gov.ar;
thomas.duriez@gmail.com; cberli@santafe-conicet.gov.ar

Abstract: Detailed study of capillary wicking in paper has become an important issue in the field of paper-based microfluidics. Based on a recent model of multiple interconnected capillary tubes presented by Cummins *et al.*, we discuss the most appropriate pore size distribution to reproduce the experimental data. We demonstrate that this model can predict correctly the dynamic of the flow front under an exhausted reservoir condition. This phenomenon has not been discussed in literature previously and constitutes the major claim of this work. However, the model needs a suitable pore size distribution as an input in order to match experimental saturation curves. We use state-of-the-art machine learning tools to find a pore size distribution that justifies the observed features of capillary flow in filter paper strips. Experimentally, we measure the light transmittance of the paper strip as a function of time and position during wicking. Through a appropriate calibration this optical signal can be converted to porous medium saturation. The pore size distribution found can be used to reproduce all the dynamics along the different flow regimes.

Key-Words: paper-based microfluidics ; capillary imbibition ; pore distribution

Introduction: Capillary imbibition of paper has been well described by the classical model of Lucas-Washburn, which assumes that the porous media can be considered as a bundle of straight capillary tubes (Figure 1-a). The model correctly predicts the dynamics of the wetting front ($l \propto \sqrt{t}$). However, it predicts complete saturation of the porous media behind the wetting front while experiments show that the advancing front is diffuse. Considering a broad pore size distribution (Figure 1-b), the Lucas-Washburn model predicts that large pores fill more quickly than small ones. The introduction of this modification can also explain the smooth varying saturation curve; nevertheless SEM images [1] show that liquid advance first through smallest pore spaces. Bico & Quèrè [2] propose a bidisperse porous media to explain this behavior. Their model consists in two interconnected capillary tubes of different radius. Under these conditions, the large tube work as a source of liquid for the small one. Then, the \sqrt{t} dependence on movement of the small meniscus is coupled to the movement of the large meniscus with the same dependence. However, with only two pore sizes they cannot explain the soft saturation profile observed experimentally.[3] Recently, Cummins *et al.*[4] propose a multiple interconnected capillary tube model (Figure 1-c) extending the proposal of Bico & Quèrè. Here we derive an analytical solution for this model. For N tube radius $r_1 \leq r_2 \leq \dots \leq r_N$ with fractional flow areas f_1, f_2, \dots, f_N the position L_j of the meniscus in the j -th tube satisfies:

$$\sum_{k=1}^j f_k \frac{dL_k}{dt} = \frac{CB_j}{L_j - L_{j+1}} \quad (1)$$

where $B_j = (r_j^{-1} - r_{j+1}^{-1}) \sum_{i=1}^j r_i^2 f_i$ and C is a constant dependent of substrate-liquid system variables. The solution for this system of equations has the form $L_j = L_j^0 \sqrt{t}$, where L_j^0 can be calculated from:

$$L_j^0 = L_{j+1}^0 + \left[\frac{1}{L_{j+1}^0 - L_{j+2}^0} \frac{B_{j+1}}{B_j} - f_{j+1} \frac{L_{j+1}^0}{B_j} \sum_{k=1}^j \frac{B_j}{f_k L_k^0} \right]^{-1} \quad (2)$$

On the other hand, when the inlet reservoir exhaust its content, the velocity of the advancing front abruptly decrease, but does not stop. Danino & Marmur [5] observed this behavior for the radial case and speculated that during this secondary motion the liquid flow from large pore (high capillary pressure) to small ones (low capillary pressure). However, they did not model this characteristic of the capillary flow on paper. In this work we propose that the multiple interconnected capillary tube model can also explains the liquid front movement under exhaust reservoir condition. For this propose it is necessary to change the boundary condition of the model, which implies $B_N = 0$ and then solve equations (1) through Runge-Kutta methods.

The model requires a pore size distribution as input. Cummins *et al.* construct a pore size distribution based on data from a mercury porosimetry. Using this distribution in the analytical solution we show that the saturation

curve predictions does not reproduce the experimental data (Figure 2-b). We use state-of-the-art machine learning techniques to find a suitable pore size distribution that explain both regimes at the same time.

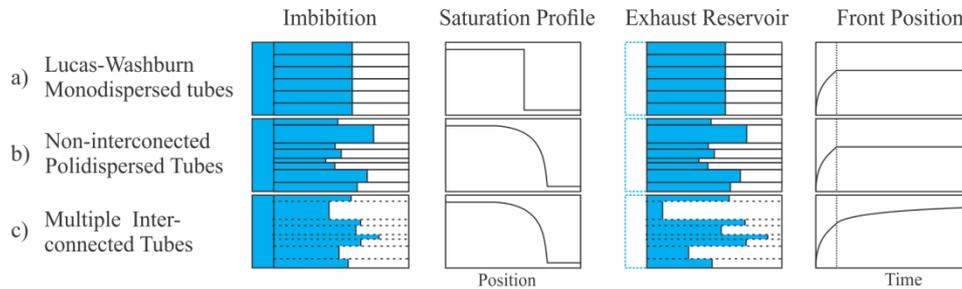


Figure 1- Schematics representation of porous media model and predicted features.

Experimental: Paper strips of Whatman chromatographic paper (grade 3mm Chr) of $5 \times 60 \text{ mm}^2$ were cut by a craft-cutter. Strips were laminated leaving the last 10 mm open to the atmosphere in order to form de inlet area. Plastic pouches were sprayed with a hydrophobic spray based on fluorinated acrylics (Imper Ital Protect, Muntex SA, Munro, Argentina) previous to lamination. The devices were horizontally placed over a white backlighting LED panel. The measurement of the optical transmittance permits an indirect determination of saturation at real time. Using an analytical scale we obtain a calibration curve which permits the conversion of transmittance to saturation. After a certain time the inlet area was dried in order to induce the exhausted reservoir condition. All the process was registered by using a high resolution digital camera (Canon EOS Rebel T5) focused vertically at a distance of around 20 cm, connected to a PC. The spatial resolution was $36 \mu\text{m}$ and the time resolution was 12 frames per minute. Images were analyzed in order to obtain transmittance by subtraction and normalization.

Results and discussion: Figure 2-a shows a color map of the experimental saturation as a function of time and the position along the strip, i.e. the measured temporal profilogram of porous media saturation. At $t=140\text{s}$ the exhaust reservoir regime starts. It can be observed that the liquid front continues to advance even when the reservoir is empty. Figure 2-b presents the simulated temporal profilogram obtained using the pore size distribution proposed by Cummins et al. based on mercury porosimetry. The predicted saturation profiles are not in agreement with experimental data, while the predicted velocity of the front is higher than experimental one. Here we propose that the exhausted flow regime become critical to find a pore size distribution that satisfies the behavior observed experimentally. We implement a genetic programming method to find the function $f(r)$ that minimize the square meaning error between experimental and simulated profilograms. The result is showed in Figure 2-c.

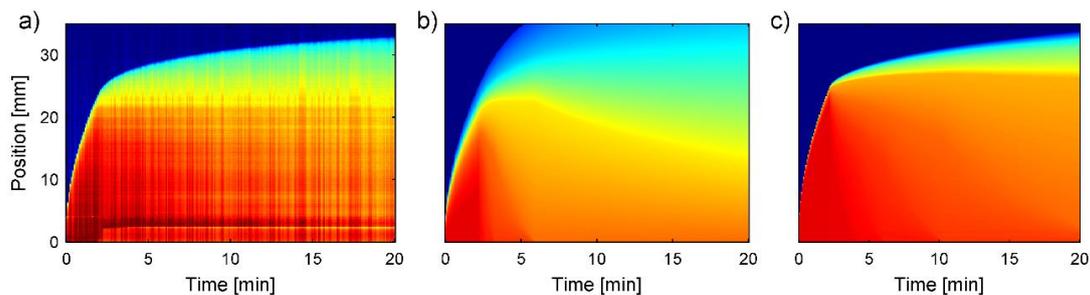


Figure 2- Profilograms of saturation: (a) experimental, and simulated using (b) the pore size distribution proposed by Cummins and (c) the distribution funded through genetic programming.

Conclusion: In this work we propose that the multiple interconnected capillary tube model can be used successfully to describe two advanced features of the capillary imbibition of paper: the saturation profile and the advance of liquid front during exhausted reservoir regime. This features, in combination with state-of-the-art genetic programming methods could be used to characterize the pore size distribution of paper sample.

References and acknowledgements: Authors acknowledge the support of CONICET (PIP 0317), ANPCyT (PICT 315) and UNL (CAI+D 12G/328).

References: [1] R.J. Roberts et al., Journal of Pulp and Paper Science **29**, 123 (2013).

[2] J. Bico and D. Quèrè, Europhysics Letters **61**, 348 (2003)

[3] R. Williams, Journal of Colloid and Interface Science **79**, 287 (1987).

[4] Cummins et al., Analytical Chemistry **89**, 4377 (2017).

[5] D. Danino and A. Marmur, Journal of Colloid and Interface Science **166**, 245 (1994).