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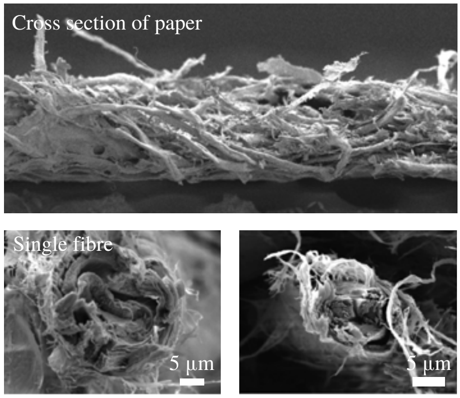
Flow of liquids through paper



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The flow of liquids through paper is challenging to model due to the complexity and disordered layout of the fibre matrix. The expanding use and capability of microfluidic paper-based analytical devices (μPADs) and their requirement for precision has increased the need to accurately predict the flow of liquids through paper. Many studies have developed models and revealed some of the physical mechanisms responsible for the flow behaviour, but we still lack a complete understanding, particularly in relation to how the fluid fills the various voids with a wide range of shapes and sizes in the fibre matrix of paper. In the featured article, Chang *et al.* (*J. Fluid Mech.*, vol. 845, 2018, 36–50) used a combined experimental and theoretical approach to uncover the importance of the liquid filling the intra-fibre pores, showed that this results in deviation from the flow behaviour predicted by the Lucas–Washburn equation and developed a model which accounts for this effect.

Key words: capillary flows, microfluidics, porous media

1. Introduction

The emergence and rapid growth of microfluidic paper-based analytical devices (μPADs) has led to an increased demand for high precision modelling of the capillary flow of liquids through paper and an understanding of the physical mechanisms that influence the flow, to enhance the design of devices that can produce consistent and accurate measurements. In μPADs, the use of paper as the substrate introduces numerous advantages, such as the ease of manufacturing, low cost, disposability and capability to generate flow without external pumps, which make them well-suited for a broad range of user-friendly testing, especially in locations with limited access to resources. Devices have been developed for medical diagnostics, environmental testing and other areas, and there have been many excellent review articles on μPADs (see Martinez *et al.* 2010; Yetisen, Akram & Lowe 2013; Cate *et al.* 2015; Gong & Sinton 2017). These μPADs rely on precise and predictable flow of liquids through the paper

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substrate but modelling this flow can be complicated since paper is a complex porous medium that consists of randomly interwoven fibres of varying length and width. In spite of much research on liquid flow through porous media, and paper specifically, we still lack the ability to generate precise flow predictions for many situations and do not have a complete understanding of the physical mechanisms that influence the flow.

Capillary flow of liquids through porous media was first described one hundred years ago by Lucas (1918) and Washburn (1921), and also by Bell & Cameron (1906). Lucas (1918) and Washburn (1921) independently derived the relationship using Poiseuille's equation, the Laplace pressure and a number of assumptions. They proposed that capillary flow through porous media can be approximated by a bundle of cylindrical capillary tubes, and found that the distance the liquid travels into the medium, l, advances diffusively in time and is given by $l^2 = (\sigma \cos\theta/2\mu)Rt$, where σ is the surface tension, θ is the contact angle, μ is the liquid viscosity, R is the tube radius and t is time. It is clear that assuming porous media to be made up of bundles of cylindrical tubes is not realistic for many cases, particularly paper, and Washburn (1921) pointed out the limitations of this expression stating that it is not applicable for cases where the pores in the media are not equivalent to cylindrical pores and cases where the pore contains an enlargement or ends in a pocket.

In spite of the limitations, the 'Lucas–Washburn' expression is commonly used to predict liquid flow through paper (see Gong & Sinton 2017), probably due to its simplicity and the experimental confirmation by many researchers that the penetration distance is often linearly proportional to the square root of time. Many researchers have applied it with empirically determined parameters, particularly contact angles and 'effective pore radius' values, and found good agreement, but this approach does not always yield precise predictions or reveal the underlying physical mechanisms.

Some physical mechanisms that are not included in the Lucas–Washburn relation and apply to paper involve the swelling of fibres (see Schuchardt & Berg 1991; Masoodi & Pillai 2010; Kvick 2017), inertial effects and uneven filling of smaller versus larger inter-fibre pores (see Szekely, Neumann & Chuang 1971; Sorbie, Wu & McDougall 1995; Schoelkopf *et al.* 2002), film flow (Roberts *et al.* 2003), evaporation (see Amaral *et al.* 1994; Balankin *et al.* 2013), pores that change in size (Reyssat *et al.* 2008) and, importantly, the effect that is uncovered in the featured article by Chang *et al.* (2018): the filling of the intra-fibre pores within the fibres. The questions answered are: (i) does the filling of the intra-fibre pores influence the flow of liquids through paper? And if so, (ii) is there a simple way to model this effect?

2. Overview

In order to determine the role played by the intra-fibre pores, Chang *et al.* (2018) first used scanning electron micrographs to determine the nature and size of these void spaces, as shown in figure 1. They determined the size of the intra-fibre spaces and compared with the inter-fibre spaces. A key feature of this study is the clever use of a sheet of fabric made up of fibres that do not contain any intra-fibre pores, as shown in figure 1(*i*), to enable comparison of liquid flow through media with and without intra-fibre pores. They also selected silicone oils as the liquid in their experiments to prevent any swelling and evaporation, so as to only investigate the influence of the intra- and inter-fibre pores.

Past work has demonstrated that the liquid front observed while liquid is flowing through paper does not indicate that all of the paper behind this front is completely

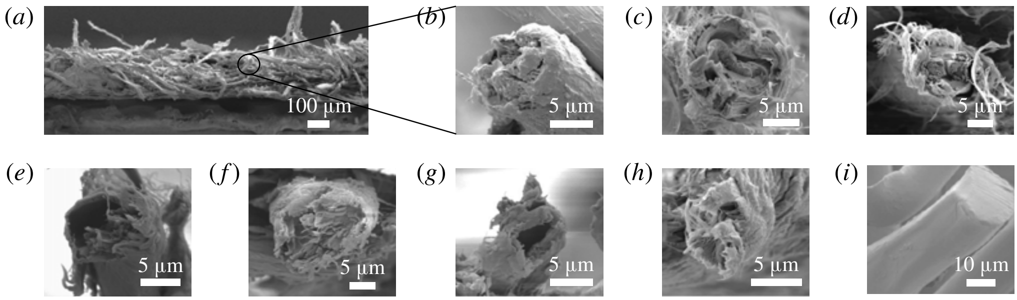

FIGURE 1. Scanning electron microscope (SEM) images of single fibres of the various papers and non-woven fabric. (*a*) Cross-sections of the specimen of Filter II (Whatman grade 5). Cross-sections of single fibres in (*b*) Filter II (Whatman grade 5), (*c*) Filter I (Whatman grade 4), (*d*) Filter III (CHM F1005), (*e*) chromatography (Advantec No. 50), (*f*) Kent paper, (*g*) watercolour paper, (*h*) kraft paper and (*i*) non-woven fabric (Chang *et al.* 2018).

saturated with the liquid (Bico & Quéré 2003). This was also confirmed in a recent study by Walji & MacDonald (2016) where they observed post-wetting flow into paper after the visual liquid front reached the end of the strip. This effect is often attributed to the slower filling of smaller inter-fibre pores as a result of the distribution of pore sizes in a complex fibre matrix like paper. But the work by Chang *et al.* (2018) showed that the fabric made from fibres with no intra-fibre pores had a negligible amount of post-wetting flow even though the fabric would presumably have a distribution of inter-fibre pore sizes as well; therefore, the intra-fibre pores contributed to the post-wetting flow. This is an important finding because visualization of the inter-fibre pore size distribution has been used in the past to quantify post-wetting flow, but the intra-fibre pore size should also be considered and may have a dominant influence on the flow behaviour.

Their experiments showed that there was not a linear dependence between l^2 and t, as predicted by the Lucas–Washburn expression, and they attributed the slowing of liquid penetration to the slower filling of the intra-fibre pores.

Chang *et al.* (2018) modelled the influence of the intra-fibre pores by modifying the geometry of the cylindrical tube model proposed by Lucas (1918) and Washburn (1921), and included a small slit along the side of the cylinder. Their model included two new parameters, t_c and ψ, which correspond to the time required to fill the intra-fibre pores, and the volume ratio of intra- to inter-fibre pores, respectively, and can be attained from visualization and experiments. Their model showed good agreement with the experimental results and further confirmed the importance of considering the flow into the intra-fibre pores.

3. Future

There are several ways in which the intra-fibre pore dependence will impact the development of μPADs. For example, Chang *et al.* (2018) found that paper with a large inter-fibre pore and small intra-fibre pore volume fraction had a fast flow speed. By employing papers with varying volume fractions we can predictably vary the flow speed, which is a tool that can be used in μPAD design for enhanced flow control. By expanding our understanding of the physical mechanisms driving the flow through paper we can model the flow more precisely, which will lead to more accurate μPADs.

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The results will also influence the study of liquid flow through porous media. For example, some past research on the effect of swelling has assumed that the volume of liquid entering the fibres is directly correlated with the amount of swelling. However, this work reveals that liquid can fill intra-fibre pores without causing swelling.

Future work could combine the effects of swelling, evaporation, intra-fibre flow and inter-fibre size distribution, since many fluids of interest are aqueous and all of these mechanisms can impact the flow of aqueous liquids through paper. It would also be interesting to compare the filling of the small intra- and inter-fibre pores to see if there is any fundamental difference in the flow behaviour. It was previously thought that intra-fibre pores may not provide a continuous path for the fluid through the paper, but the scanning electron micrographs in this work show that might not be the case.

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